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MODULUS OF RAPTURE (MOR) OF PORCELAIN BY SUBSTITUTION OF QUARTZ WITH RICE HUSK ASH (RHA) AND PALM OIL FUEL ASH (POFA) AT DIFFERENT TEMPERATURES

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ABSTRACT

This paper investigates to effects RHA and POFA in porcelain body as substitute materials for quartz. In order to collect experimental data, the effects of ground RHA and POFA replacement rate from 0 wt% up to 25 wt% in porcelain body was studied. The RHA and POFA were grounded in a ball mill until the median particle size was reduced to about 50 µm. They were used to substitute quartz in porcelain body from 0 wt% to 25 wt%. The mixed powder was pressed into pellets at mould pressure of 91 MPa. All the pellets were sintered at a temperatures of 1000 ºC, 1100 ºC, 1200 ºC and 1300 ºC for the soaking times of 2 hours. It was found out among other things that the Modulus of Rapture (MOR) of the samples increases with increase in substitution and also with the increase in temperature. With a value of 35 MPa the MOR of the samples containing 20 wt% sintered at a soaking time of 2 hours were higher than the standard porcelain. This could be attributed to the formation of larger amount of SiO_2 glass, in the presence of fluxing components such as MgO, CaO and Na_2O), more abundant in samples containing RHA and POFA.

Keywords: Quartz, Temperature, SEM, XRF, XRD

INTRODUCTION

The main phase of the porcelain body is made up from heterogeneous glassy matrix containing closed irregular shaped porosities due to gas bubbles and the melted residues of quartz grains, and numerous needle shaped mullite crystals <3 µm length. The MOR of porcelain bodies have been experimentally studied by many researchers (Hamano *et al.,* 1991; Ece and Nakagawa 2002; Mattyasovszky-Zsolnay, 1957; Wiedmann, 1959; Weyl, 1959; Noh 2014), because of its economic importance in ceramic industry. It is known fact that quartz grains in different sizes have significant effects on mechanical strength of porcelain bodies (Hassan *et al.,* 2015). Especially, it is proposed that bending strength of the porcelains increases with an increase in inter planar spacing of quartz crystals, so the quartz is under the tensile stress, and consequently, the glassy matrix surrounding the quartz grains is a compressive stress which acts as pre-stress, improving mechanical strengthening (Noh *et al.,* 2016)). Quartz grains embedded in the porcelain glassy matrix have a deleterious effect on the mechanical strength mainly because of its transformation during cooling (Noh *et al.*, 2017) which results in the development of stresses which initiate fracture (Jamo *et al.,* 2014). The thermo-mechanical properties of whiteware bodies change greatly during the reconstructive and the displacive transformation of free silica due to change in volume (Bragança *et al.,* 2006).

Several researchers tried to improve the mechanical properties of porcelain ceramics by replacing quartz with other materials viz; sericitic pyrophyllite, kyanite, bauxite, sillimanite sand, alumina, RHA, silica fume and fly ash. Although the alumina in different forms has a favourable influence on the mechanical properties of white-ware due to the formation of primary mullite, it lowers the recrystallization of secondary mullite due to an increase in the viscosity of the glassy phase. On the other hand, Stathis *et al.,* (2004) showed that silica-rich glass favours recrystallization of mullite at low temperature and its dissolution at high temperature. Some improvements in the mechanical properties were also observed by several authors (Junior *et al.,* 2009; Sanchez *et al.,* 2006; Junior *et al.,* 2008; Mohamad *et al.,* 2018; Usman *et al.,* 2015), through the reduction of the particle size of quartz and non-plastic materials. Noh *et al.,* (2014) reported that dissolved quartz in the glassy phase and cristobalite phase precipitation has a deleterious effect on the mechanical properties of porcelain ceramic. The use of the combination of RHA and POFA simultaneously to replace quartz in porcelain ceramic is still lacking. Additionally, replacing quartz with RHA and POFA is expected to have impact on the parameters such MOR and sintering temperature of the porcelain body. Hence, the present research wishes to investigate the influence of the combined effect of RHA and POFA as a substitute for quartz on the MOR of porcelain at different temperatures.

MATERIALS AND METHODS

Experimental

The RH was thoroughly washed with distilled water in order to remove adhering soil and dust. After that it was dried in an oven at 100°C for 24 hours. Then the dried husk was subjected to the chemical treatment; 2M HCL, 5% solid at 25 °C before calcinations to increase silica content. After the leaching process, the treated husk was washed with distil water and then dried again. The treated husk was then subjected to calcinations at 700°C for six (6) hours, after which it was subjected to the XRF analysis. The machine used for the analysis was XRF Bruker S4 Pioneer which was operated at 60 KV

The POFA was dried in an oven at 100 ºC for 24 hours. After that it was grinded in a ball mill to reduce the needed particle size to improve reactivity. The milling time was approximately 90 minutes at 200 rpm. Afterwards, the materials were subjected to a set of sieves less than 50 µm in order to remove the particles coarser than 50 µm. The untreated POFA was heated at a temperature of 600 ºC for 1.5 hours in an electric furnace to remove excess carbon.

Porcelain powder was grounded separately in a ball mill. The powder was sieved using sieve shaker and dried in an oven. The RHA and POFA were gradually incorporated into the body of porcelain powder from 5 %wt, to 25 %wt (Table 1). The composition was mixed using a ball mill for one and half hours. 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 the soaking times of 2 hours at a heating rate of 5 °C per minute. The compressive strength was determined. The chemical composition of the POFA was studied using X-Ray Fluorescence (XRF) machine while the amorphous structure of RHA and POFA was identified through XRD and the microstructural features were studied by SEM**.**

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

Sample name	Kaolin	Feldspar	Quartz	RHA	POFA
AP1	50	25	25	0	0
AP2	50	25	20	3	2
AP3	50	25	15	6	4
AP4	50	25	10	9	6
AP5	50	25	5	12	8
AP6	50	25	0	15	10

RESULT AND DISCUSSION

X-ray fluorescence (XRF) analysis was used for the chemical analysis. Hence the amount of chemical elements can be observed (Table 2). The presence of various elements within the raw materials can be seen from the table. This table shows the result of XRF analysis of kaolin, feldspar, quartz, RHA and POFA. It is evident that SiO_2 is the major composition in all the raw materials they are: kaolin, feldspar, quartz, RHA and POFA with 69.3 wt%, 72.7 wt%, 99.4 wt%, 93.7 wt% and 66.9 wt% and then followed by alumina with 24.3 wt%, 16.4 wt%, 0.2 wt%, 2.1 wt% and 6.4 wt% respectively.

Table 2: X-Ray Fluorescence (XRF) Analysis

Sample	Content											
Oxides	SiO_2	Al_2O_3	K_2O	P_2O_5	CaO	MgO	CO_2	SO_3	FeO_3	Na_2O	TiO_2	LOI
RHA	93.70	2.11	1.18	0.96	0.81	0.53	010	0.45	-	-	-	0.16
POFA	66.91	6.44	5.20	3.72	5.56	3.13	-	0.33	5.72	0.19	-	2.30
Kaolin	69.30	24.30	2.44	-	-	-	0.10	-	0.27	-	0.27	0.36
Feldspar	72.70	16.40	0.50	2.42	-	-	-	6.87	0.40	0.29	-	0.10
Quartz	99.40	0.22	-	-	-	-	0.10	-	-	-	-	0.28

Figure 1 shows the volume shrinkage of the sintered samples containing RHA and POFA. Lower temperature (1000 °C to 1100 °C) favours lower volume shrinkage values. The volume shrinkage increases with the increase in replacement until complete replacement of quartz by RHA and POFA. The maximum volume shrinkage was achieved with this range of temperatures with approximate values of 13%, 15%, replacement of quartz by RHA and POFA. While for the temperature of 1200 °C the maximum of 20% value of volume shrinkage was recorded. At the temperature of 1300 °C the maximum volume shrinkage was recorded with a value of 22% on complete replacement of quartz by RHA and POFA. The wetting liquids act on solid particle to eliminate porosity and reduce interfacial energy, since higher energy solid vapour interfaces gradually and replaced by lower energy solid-solid interfaces with a total decrease in free energy occurring on sintering.

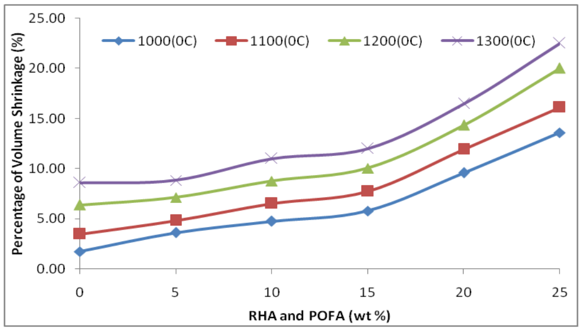

Figure 1: Effect of temperature on shrinkage of the samples with different percentage of RHA and POFA

Considering the temperature, the volume shrinkage increases with the increase in temperature and the replacement of quartz by RHA and POFA. The volume shrinkage increases as the temperature increases from 1000 °C to 1300 °C. The increase in the shrinkage may be attributed to the presence of fluxing alkaline and other oxides in the ashes (RHA and POFA), which enter the liquid phase. Furthermore, previous reports by Jamo *et al* (2014), shows a similar trend where the results of shrinkage with different quartz, mullite and kyanite content sintered at different temperature is in the range of 4% to 8%. In another development Noh *et al* (2017) showed that the shrinkage of the fired samples is associated to other physical properties such as porosity and water absorption. The shrinkage properties as function of firing temperature at the 1200 °C – 1300 °C range. Shrinkage, which initially increases, reaches a maximum value and above 1300 °C decreases due to increase in close porosity. The maximum values are at the temperature of 1200 °C.

Figure 2 shows the result MOR versus RHA and POFA content of the sintered samples. In general, an increasing trend in the strength is observed with the increase in temperature. In conformity with the porosity results presented in Figure 1. The MOR increases at a temperature of 1000 °C from 27 MPa to 29 MPa. At a temperature of 1100 °C the MOR increases from 28 MPa to 33 MPa. While at the temperature of 1200 °C it increases from 29 MPa to 35 MPa. The highest values of MOR occurs at the temperature of 1200 °C on 20 wt%.

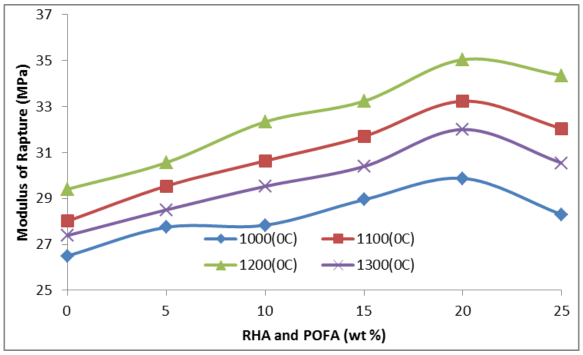

Figure 2: Effect of temperature on MOR of the samples with different percentage of RHA and POFA

The maturing temperature was also reduced by 80 °C, as and the sample containing 20 wt% has 20% increase of MOR over the standard porcelain ceramic. The MOR increases with increase in replacement of quartz by RHA and POFA. The increase may be attributed to the formation of larger amount of SiO_2 glass, in the presence of fluxing components (MgO, CaO and Na_2O), more abundant in samples containing RHA and POFA compared with the samples containing RHA only or POFA only. Again, the more heterogeneous matrix of larger liquid phase arrested the faster increasing trend of MOR (Kobayashi *et al.,* 1991). This phenomenon was also observed by Ece and Nakagawa (2002). As the substitution was increased to 25 wt% the bending strength decreases as result of excess glassy formation.

Consequently, the MOR increases with increase in temperature, as the temperature increases from 1000 °C to 1200 °C. The MOR decreases after reaching maximum values at a temperature of 1300 °C. The increase in MOR with temperature development could also be related to the presence of RHA and POFA along with mullite and the liquid phase, which are responsible for the densification processes and quartz dissolution (Pröbster 1996). The increased amount of liquid phase at higher temperature (above 1200 °C) certainly affects negatively the mechanical strength of the samples, hence, decreasing densification at higher firing temperatures could explain the decrease in MOR. The maximum mullite content is formed at a temperature of 1200 °C.

Figure 4 shows the XRD pattern of the experimental body mixes sintered at different temperatures. In this Figure it is observed that the identified crystalline phases are quartz (ICDD 046-1045), mullite (ICDD 074-4143) and cristobalite (ICDD 039-1425). Consistent with the quantitative XRD analysis (Table 3) the quartz, mullite and cristobalite increases with increase in temperature (1000 °C to1200 °C). The increase in the peaks could be attributed to the reaction of SiO_2 from RHA and POFA with Al_2O_3 as the temperature increases.

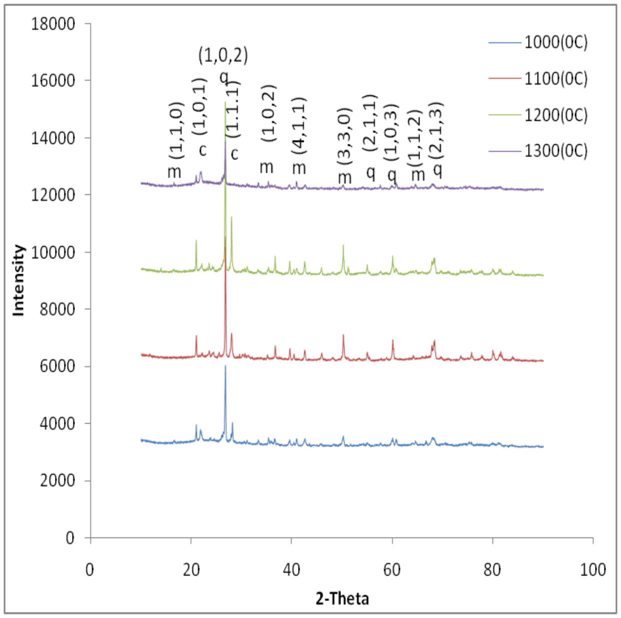

Figure 4: The XRD curves of the samples containing RHA and POFA sintered at different temperatures

Table 3: XRD quantitative analysis of the samples containing RHA and POFA sintered at different temperatures

Temperature (°C)	Quartz (%)	Mullite (%)	Cristobalite (%)	Glassy Phase (%)
1000	35.1	18.5	9.4	27.0
1100	41.4	34.3	12.5	11.8
1200	40.0	38.2	18.8	3.0
1300	36.6	30.0	11.0	22.0

Conclusion

The MOR of the porcelain body was found to increase with increase in soaking time and also with the substitution of quartz by RHA and POFA. The maximum MOR for porcelain samples containing RHA occurred at a temperature of 1200 ºC, on 20 wt% substitution. The wetting liquids act on solid particle to eliminate porosity and reduce interfacial energy, since higher energy solid vapour interfaces gradually and replaced by lower energy solid-solid interfaces with a total decrease in free energy occurring on sintering. At the temperature of 1200 ºC densification takesplace least pores were noticed. The increase in the MOR and the substantial decrease in porosity of the samples containing RHA and POFA, are attributed to the glassy formation and densification of the individual grains during the vitrification process.

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