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Effects of Temperature and Substitution of Quartz By Rice Husk Ash (RHA) On the Hardness of Porcelain Body

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Abstract

The effect of temperature and substitution of quartz by rice husk ash (RHA) on hardness of porcelain tile was investigated. Rice husk ash (RHA) was used as a substitute for quartz with different replacement levels (0wt%-25wt%). The mixed powder was pressed into pellets and at pressure of 91 MPa. All the pellets were sintered at the temperatures of 1000℃, 1100℃, 1200℃ and 1300℃ for 2 hours soaking time, at a heating rate of 5℃ per minute. Study on the effect of the microstructure of the porcelain on the Vickers hardness and physical properties has been carried out. The microstructural observations show that as the temperature increases to the grain size becomes smaller because of the quartz conversion to cristobalite due to increase in temperature and substitution of quartz by RHA. The Vickers hardness and bulk density was the highest and the porosity was the least at a temperature of 1200℃ on 20wt% substitution of POFA. Porcelain containing POFA has about 7% weight reduction compared with normal porcelain. The improvement in the properties could be attributed to sharp changes in the microstructural features as a result of increase in mullite and glassy phase simultaneously.

Keywords: Bulk density; Vickers hardness; Porcelain; RHA; Temperature

1.0 Introduction

Rice husks are a residue produced in significant quantities on a global basis. While they are utilized as a fuel in some regions, in other countries they are treated as waste, causing pollution and disposal problems. Due to growing environmental concern, and the need to conserve energy and resources, efforts have been made to burn the husks under controlled conditions and to utilize the resultant ash as a building and ceramic material [1 -4].

The beneficiation of rice generates as by-product rice husk ash that corresponds to about 23% of its initial weight. This ash can be used as a fertilizer in agriculture [4] or as an additive for cement and concrete fabrication [2, 3]. Due to its high silicon content, rice husk has become a source for preparation of elementary silicon [5,6] and a number of silicon compounds [7], especially silica [8,9], silicon carbide [10,11] and silicon nitride [12].

Up to now, little research has been done to investigate the use of RHA as supplementary material in porcelain production in Nigeria. For this reason, this study investigates the influence of temperature on the substitution of quartz by rice husk ash in porcelain composition.

2.0 Experimental Procedures

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 KVP and 50 mA.

Porcelain powder was grounded separately in a ball mill. The powder was sieved using 50µm sieve and dried in an oven. The RHA was gradually incorporated into the body of porcelain powder (Table 1). The composition was mixed using a ball mill for 1.5 hours. The mixed powder was pressed into pellets and bars at pressure of 91 MPa. All the pellets were sintered at the temperatures of 1000°C, 1100°C, 1200°C and 1300°C for 2 hours soaking time, at a heating rate of 5°C per minute. The bulk

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density and Vicker hardness were determined. The chemical composition of the RHA was studied using X-Ray Fluorescence (XRF) while the crystalline structure of the pellets was identified through XRD and the microstructural features were studied by SEM.

The Vicker hardness was carried out according to ASTM-C-1327. The device used for the hardness test was Shimadzu Micro-hardness tester. Before the test, the samples were polished. The samples were sprayed with black colour in order to get the clearer surface microstructure. The magnitude of the test force was 9.81 N while the application of the test load was 0.015 mm/s. The Vickers hardness, *HV*, is given by:

HV = Constant × (Test force / Surface of indententation)

$$\text{HV} = 0.102 \times \frac{\left(\sin\frac{136^{o}}{2}\right)}{d^2}$$

Table 1: Body composition with progressive replacement of quartz by RHA

Sample name	Kaolin	Feldspar	Quartz	RHA
AR1	50	25	25	0
AR2	50	25	20	5
AR3	50	25	15	10
AR4	50	25	10	15
AR5	50	25	5	20
AR6	50	25	0	25

3.0 Resultsand 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 and RHA with 69.3 wt%, 72.7 wt%, 99.4 wt% and 93.7 wt% and then followed by alumina with 24.3 wt%, 16.4 wt%, 0.2 wt%, and 2.1 wt% respectively.

Table 2: Chemical analysis of the minerals

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
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 SEM analysis of the mixed samples sintered at different temperatures. It can be seen from Figure (1a) that the grains are big due to undissolved quartz but as the temperature increases to 1100°C the grains becomes small (Figure 1b), at the temperature of 1200°C (Figure 1c) the quartz dissolved and got converted to cristobalite, more mullite 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 RHA.

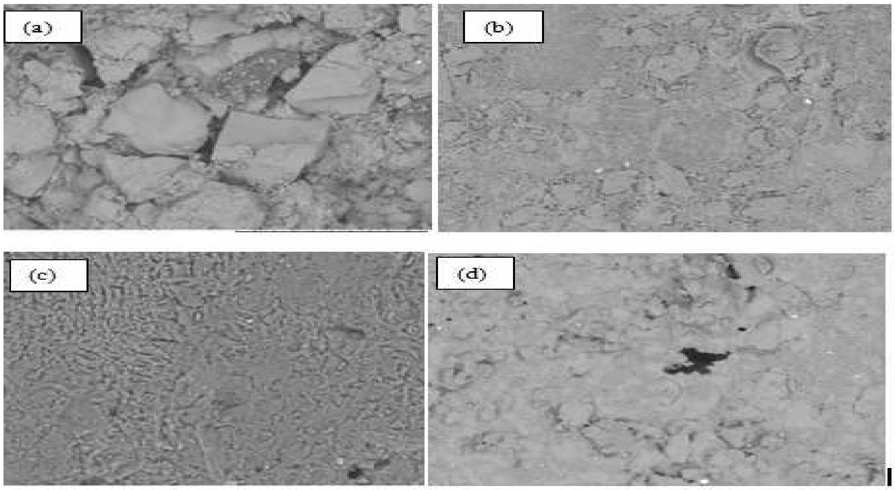

Figure 1: The SEM of the mixed samples containing RHA sintered at different temperatures

As the temperature reaches 1300^{o}C (Figure 1d) pores re-emerged, this could be as result of bloating (i.e., pore volumeexpansion),whicharisesfromhigherpressures (athightemperatures)ofgasessuchasnitrogen,carbonmonoxide and carbondioxideentrappedwithinclosedpores. Similar result was obtained [13,14].

Figure 2 shows the XRD qualitative of analysis for mixed sintered samples at different temperatures. Three major phases were identified are quartz hexagonal (ICDD 046-1045), mullite orthorhombic (ICDD 4143), and cristobalite tetragonal (ICDD 039-1425). The peaks intensity for mullite, and cristoballite with increases between the temperature of 1000°C and 1200°C, and decreases at a temperature of 1300°C. Quartz dissolution is expected as the temperature increases but a reverse rend was noticed here, this could probably be attributed to residual that did not suffer conversion [15]. The quartz decreases while mullite and cristobaliteincreases, as the temperature increases from 1000^{o}C to 1200^{o}C. Above the temperature of 1200^{o}C a reverse trend was noticed. All the three minerals decrease due to over firing.

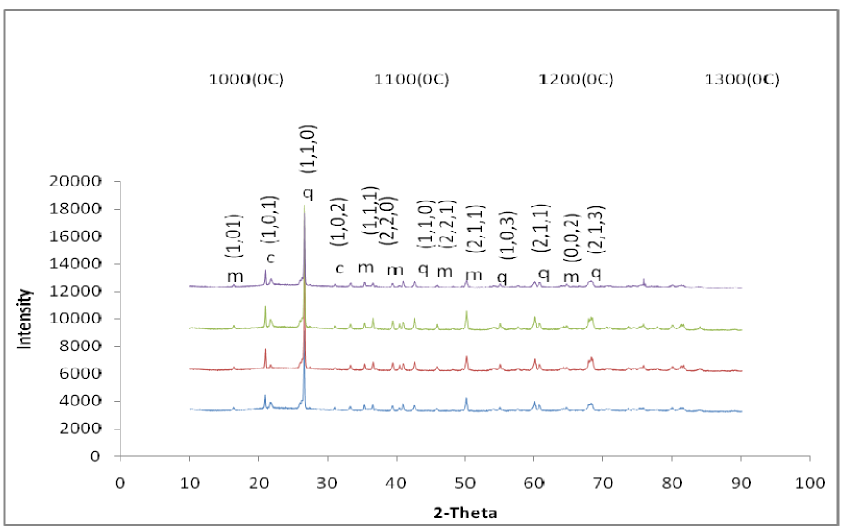

Figure 2: The XRD curves of the mixed samples containing RHA sintered at different temperatures

Figure 3 shows the result of bulk density of the mixed samples containing RHA. As can also be seen from the Figure the result of bulk density of the mixed samples containing RHA has an increasing trend. Consistent with SEM the bulk density increases with the increase in temperature and with the incorporation of RHA. With aapproximate values of 2.28 g/cm^{3} the maximum bulk density was reached on 15 wt% of RHA at a temperature of 1000 ^{o}C, further substitution causes the bulk density to decrease. At the temperatures of 1100 ^{o}C, 1200 ^{o}C and 1300 ^{o}C the maximum was reached on 20 wt% of RHA with approximate values of 2.38 g/cm^{3}, 2.42 g/cm^{3} and 2.35 g/cm^{3} respectively. This could be as a result of increase in mullite which mainly originates from clay, feldspar and RHA [16].

In terms of temperature the bulk density increases as the temperature increases between the temperatures of 1000 ^{o}C to 1200 ^{o}C. The increase could be due to decrease in internal pores as the substitution increases which causes the densification to increase [17]. However, as the temperature increases to 1300 ^{o}C the values for bulk density drops to a value of 2.10 g/cm^{3} this could be as a result of bloating [18].

The result of Vickers micro-hardness of sintered samples is shown in Figure 4. In line with the studies earlier carried out [19], the result of Vickers hardness increases with the temperature increase and also with increase of quartz substitution by RHA. In conformity with the bulk density result presented in figure 3 the bulk density increases with increase substitution of quartz by RHA. The maximum hardness was achieved with approximate values of 443 HV, for 1000 ^{o}C on 15 wt% of RHA. While for the temperatures of 1100 ^{o}C, 1200 ^{o}C, and 1300^{o}C the maximum value of Vickers micro-hardness was achieved with approximate values of 892 HV, 1052 HV and 641 HV respectively on 20 wt% substitution of quartz by RHA. SiO_2 from RHA makes up part of the crystalline phases of porcelain that develop the needed hardness to the porcelain.

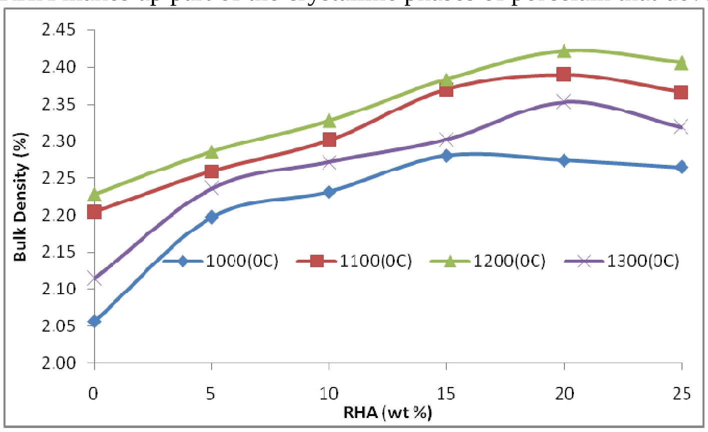

Figure 3: Effect of temperature on bulk density of body mixes with different percentage of RHA

A considerable part of SiO_2 and Al_2O_3 passes into molten feldspar, which increases the strength of the glassy phase. This is in conformity with the XRD both quantitative and qualitative, and also SEM analysis supportive. A similar conclusion had been also reached that the hardness in particular and mechanical strength in general of porcelain is such as hardness is influenced mainly by stresses set up in the glassy phase [20-22].

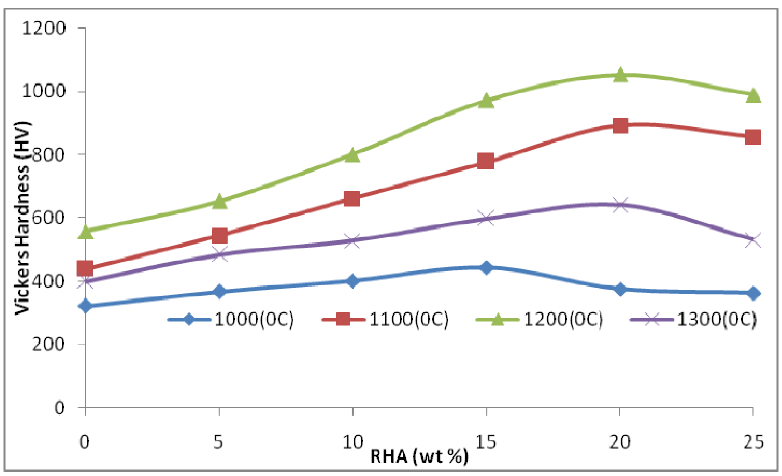

Figure 4: Effect of temperature on Vickers micro-hardness of body mixes with different percentage of RHA

4.0 Conclusion

The microstructure of porcelain body were significantly affected with the increase temperature and substitution of quartz by RHA. The grains become smaller as the temperature and substitution increases. The bulk density increases from 2.04g/cm^3 to 2.42g/cm^{3}as the temperature increases from from 1000 ^{o}C to 1200 ^{o}C, as well as the substitution of quartz by RHA.Hence the maximum hardness was achieved at a temperature of 1200^{o}C with a value of 1052HV on 20wt% substitution of quartz by RHA.

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6.0 References

[1] Prasad, C. S., Maiti, K. N., & Venugopal, R. (2001). Effect of rice husk ash in whiteware compositions. *Ceramics International*, 27(6), 629-635.

[2] Rodríguez de Sensale, Gemma. "Strength development of concrete with rice-husk ash." *Cement and Concrete Composites* 28, no. 2 (2006): 158-160.

[3] Givi, Alireza Naji, Suraya Abdul Rashid, Farah Nora A. Aziz, and Mohamad Amran Mohd Salleh. "Contribution of rice husk ash to the properties of mortar and concrete: a review." Journal of American science 6, no. 3 (2010): 157-165.

[4] Prasad, C. S., Kedar Nath Maiti, and R. Venugopal. "Effect of substitution of quartz by rice husk ash and silica fume on the properties of whiteware compositions." Ceramics international 29, no. 8 (2003): 907-914.

[5] Della, V. P., I. Kühn, and D. Hotza. "Rice husk ash as an alternate source for active silica production." Materials Letters 57, no. 4 (2002): 818-821.

[6] Narciso-Romero, F. J., and F. Rodriguez-Reinoso. "Synthesis of SiC from rice husks catalysed by iron, cobalt or nickel." Journal of materials science 31, no. 3 (1996): 779-784.

[7] Kurama, S., and H. Kurama. "The reaction kinetics of rice husk based cordierite ceramics." Ceramics International 34, no. 2 (2008): 269-272.

[8] Rahman, I.A., "Preparation of Si3 Nby corbothermal reduction of digested rice husk, Ceramic International". 20, (1994): 195–199.

[9] Real, Concha, Maria D. Alcala, and Jose M. Criado. "Preparation of silica from rice husks." Journal of the American ceramic society 79, no. 8 (1996): 2012-2016

[10] Kato, F. "Recycling waste husk into road construction material. " Mon. Waste 26, no. 12 (2000): 68–71.

[11] Choi, Nak-Woon, Ippei Mori, and Yoshihiko Ohama. "Development of rice husks–plastics composites for building materials." Waste management 26, no. 2 (2006): 189-194.

[12] Kurama, S., N. Ay, S. Turan, and E. Putun. "Effect of active silica on the microstructure of porcelain." In Proc. Turkish Ceram. Society., pp. 279-286. 1998.

[13] Lee, W. E., G. P. Souza, C. J. McConville, T. Tarvornpanich, and Y. Iqbal. "Mullite formation in clays and clay-derived vitreous ceramics." *Journal of the European Ceramic Society* 28, no. 2 (2008): 465-471.

[14] Kitouni, S., and A. Harabi. "Sintering and mechanical properties of porcelains prepared from algerian raw materials." Cerâmica 57, no. 344 (2011): 453-460.

[15] Kamseu, E., C. Leonelli, D. N. Boccaccini, P. Veronesi, P. Miselli, Giancarlo Pellacani, and U. Chinje Melo. "Characterisation of porcelain compositions using two china clays from Cameroon." *Ceramics international* 33, no. 5 (2007): 851-857.

[16] Martín-Márquez, Jorge, J. Ma Rincón, and Maximina Romero. "Effect of firing temperature on sintering of porcelain stoneware tiles." *Ceramics International* 34, no. 8 (2008): 1867-1873.

[17] Prasad, C. S., Kedar Nath Maiti, and R. Venugopal. "Effect of rice husk ash in whiteware compositions." *Ceramics International* 27, no. 6 (2001): 629-635.

[18] Pérez, Juan M., Jesús Ma Rincón, and Maximina Romero. "Effect of moulding pressure on microstructure and technological properties of porcelain stoneware." *Ceramics International* 38, no. 1 (2012): 317-325.

[19] Cattani-Lorente, Maria, Susanne S. Scherrer, Patrick Ammann, Marc Jobin, and H. W. Wiskott. "Low temperature degradation of a Y-TZP dental ceramic." *Acta Biomaterialia* 7, no. 2 (2011): 858-865.

[20] Daguano, Juliana KMF, Paulo A. Suzuki, Kurt Strecker, Maria HFV Fernandes, and Claudinei Santos. "Evaluation of the micro-hardness and fracture toughness of amorphous and partially crystallized $3CaO \cdot P_2O_5$–SiO_2–MgO bioglasses." *Materials Science and Engineering: A* 533 (2012): 26-32.

[21] Kordani, Naser, Ali Asadi, and Amin Jabbari. "Optimization of fracture behavior of alumina/silicon carbide nano ceramic." *Neural Computing and Applications* (2012): 1-7.

[22] Beheri, Hanan H., Khaled R. Mohamed, and Gehan T. El-Bassyouni. "Mechanical and microstructure of reinforced hydroxyapatite/calcium silicate nano-composites materials." *Materials & Design* (2012)