

INFLUENCE OF ADDITION OF RICE HUSK ASH ON PORCELAIN COMPOSITION

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ABSTRACT

Rice husk ash (RHA) has long been known to possess high silica content. The abundance of rice husk as agricultural waste makes it the most promising to be used as a supplementary cementations and ceramic material. In this study RHA was used as an additive material on porcelain body. The rice husk was subjected to the chemical pre-treatment with 2M HCL, 25°C before calcinations to increase silica content of the husk ash. The pressed pellets were sintered at the temperature of 1200°C and soaked for 2 hours. It was found that progressive addition of RHA in a porcelain body resulted in early vitrification of the mixture. The compressive strength was the highest at 40wt% addition of RHA. Porosity was the least at 40wt%. It can therefore be concluded that 40wt% addition of RHA into the porcelain body is the best composition.

KEYWORDS: RHA, Porcelain, Compressive Strength, Bulk Density, Porosity

INTRODUCTION

Rice Husk (RH) is an agricultural residue abundantly available in rice producing countries. Globally, approximately 600 million tonnes of rice paddy is produced each year. Therefore, total global ash production could be as high as 21 million tonnes per year. In Malaysia, for example, the RH produced annually amounts to more than 2 million tons (Motari *et al*, 2009), while in Brazil, 2.5 million tonnes of RH is generated each year (Rodrigues, 2003). In India the production of paddy was about 150 million tonnes as at 2001 (Prasad *et al*, 2001). This attracted the attention of the researchers to think of using it as a recycled material in order to reduce potential environmental problems.

Rice husk is usually discarded and burned in the fields. This is common practice that can lead to a serious environmental damage as silica particles remain suspended in the air, hence a potential cause of respiratory diseases (Siqueira *et al*, 2009), while the release of methane gas into the atmosphere results in the depletion of the Earth's ozone layer (Gastaldini *et al*, 2009). Resources supply and environmental protection are increasingly becoming matters of global concern. It is well known that large amount of RH is left as waste product after the rice production and it has no commercial interest (Amick, 1982). The major contents of RH are organic matters such as ash, 20% lignin, 22%; cellulose, 38%; and pentosans, 18%; and other organics, 2%. Rice husk ash (RHA) is a residue which contains approximately 85–90 wt. % of silica, 10–15 wt. % of carbon, small

amounts of alkalis and other trace elements (Krishnarao and Mahajan, 1995; Gastaldini *et al*, 2009). The presence of silica as SiO₂ in rice husk ash has been known since 1938 (Martin, 1938).

Nowadays silica (SiO₂) has been widely used in vegetable oil refining, pharmaceutical products, detergents, adhesives, chromatograph column packing, and ceramics (Martin, 1938; Brinker and Scherer, 1990; Proctor *et al*, 1995).

The use of RHA in the form of silica in the ceramic field has been studied at a laboratory level by several authors. Prasad (2001) and Kula *et al* (1998) investigated the effect of rice husk ash in traditional whiteware compositions completely replacing the quartz phase without substantial modification to the physical and mechanical properties of the products. Naskar and Chatterjee (2004) investigated the possibility to use this agricultural waste material to synthesize lithium aluminium silicate (LAS) glass ceramics and cordierite.

The approaches mentioned above make use of the substitutive approach by using RHA to substitute quartz. In this study we intend to study the effects of additive influence of RHA on Porcelain body. Hence, the aim of this study therefore is to study the effects of the addition of rice husk ash on the composition of porcelain body.

MATERIALS AND METHOD

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 treated with 2M HCL, 5% solid at 25°C before calcinations to increase silica content. After the leaching process, the treated husks were washed with distil water and then dried again. The treated husk was then subjected to calcinations at 700°C for six (6) hours.

Porcelain powder was grounded separately in a ball mill. The powder was sieved using sieve shaker and dried in an oven. The RHA was also ground in a ball mill. The RHA was gradually incorporated into the body of porcelain powder from 10%wt to 90%wt. The composition was mixed using a ball mill for one and a half hours. The mixed powder was pressed into pellets at a pressure of 3 tonnes. All the pellets were sintered at a temperature of 1200°C for 2 hours soaking time, at a heating rate of 5°C per minute.

The physical and mechanical properties of the pellets such as volume shrinkage, porosity and compressive strength were determined. The chemical composition of the RHA was studied using X-Ri 7

Fluorescence (XRF) machine while its amorphous structure was identified through XRD. Its microstructural features were studied by SEM and FESEM.

RESULTS AND DISCUSSION

XRF analysis is proficient in analyzing material contents inside RHA, hence the amount of SiO₂ can be observed. The presence of various compounds within the RHA and porcelain raw material can be seen in Table 1. It is evident that SiO₂ is the major composition in both porcelain and RHA with 66.50wt% and 93.70wt%, respectively. Followed by alumina with 26.13wt% and 2.11wt%, respectively.

Table 1: Chemical analysis of RHA and Porcelain

| Composition | RHA Wt(%) | Porcelain Raw Material Wt(%) |
|--------------------------------|----------------------|---|
| SiO ₂ | 93.70 | 66.50 |
| Al ₂ O ₃ | 2.11 | 26.30 |
| K ₂ O | 1.18 | 4.60 |
| P ₂ O ₅ | 0.96 | - |
| CaO | 0.81 | 0.45 |
| MgO | 0.53 | 0.18 |
| SO ₃ | - | 0.45 |
| Na ₂ O | - | 0.86 |
| Fe ₂ O ₃ | - | 0.42 |
| Rb ₂ O | - | 0.14 |
| CsO | - | 0.13 |
| Co ₂ | 0.10 | 0.10 |

Figure 1 shows the particle size of RHA. The lower and highest percentages are (d₁₀) and (d₉₀) of the RHA particle size is 2.18 μm

and 11.33 μm, respectively. The average particle (d₅₀) size is 5.35 μm.

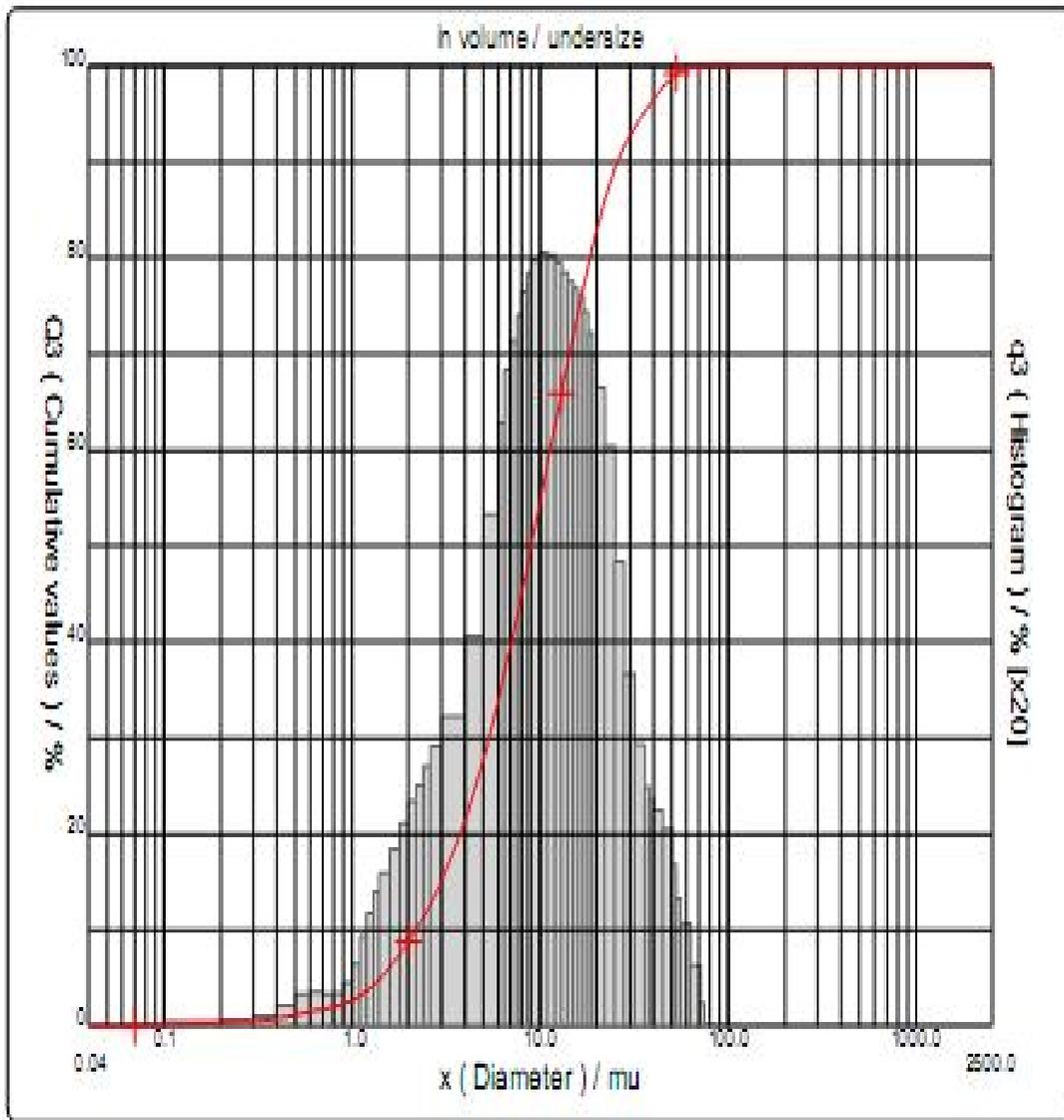


Figure 1: Result of particle size analysis of RHA

X- Ray Diffractometer is a mechanical device for obtaining x-ray intensities as a function of the angle between the incident and the diffracted beams. Figure 2 shows the result of phase diagram (called a diffractogram) which indicated that at 40kV/40A, speed of 0.05°/min and scanning at $10^\circ \geq 2\theta \leq 80^\circ$, the ash was mainly in amorphous

form as indicated by a broad peak centred on 2θ angle of figure 2. The phase concentration is indicated by the peak height, with higher peak representing higher concentration. The amorphous structure is indicated by a background hump at peak position of approximately 25° on the diffractogram.

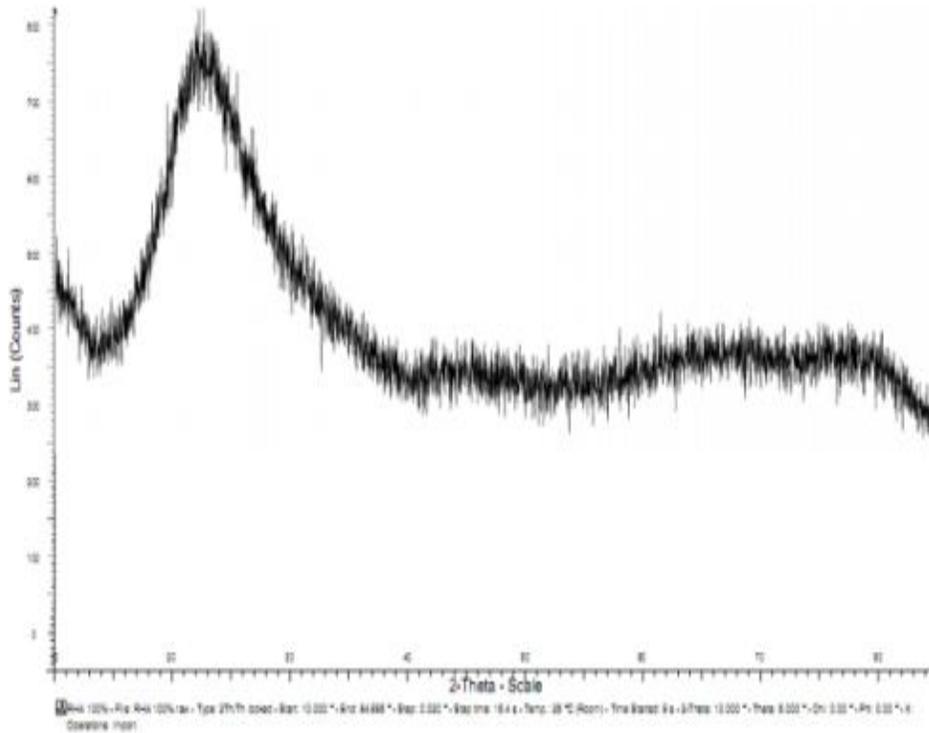


Figure 2: X-ray diffraction of RHA

Figure 3 shows the scanning electron microscopy of the RHA which indicates that the RHA has a porous cellular structure and consists of irregular-shaped particles, with particle lengths of 51.3 and 106µm. Figure 4 shows the picture of treated RHA while Figure 5 shows

untreated RHA. The difference can be seen clearly as the treated RHA looks lighter because of the high silica content while the untreated one looks greyish because of low silica content.

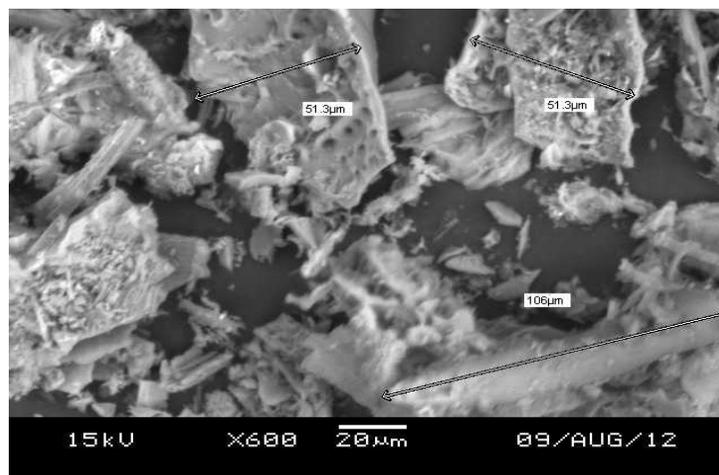


Figure 3: SEM micrographs of RHA



Figure 4: Treated RHA

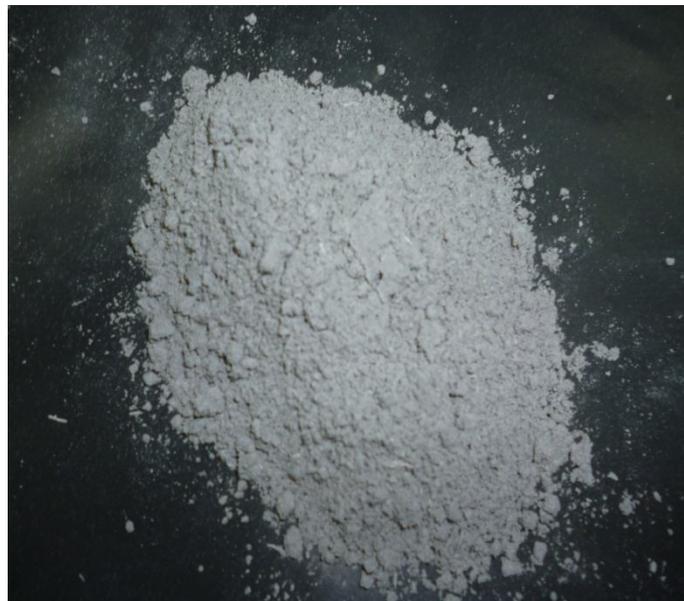


Figure 5: Untreated RHA

Figure 6 shows the volume shrinkage is on the increase with the increase of RHA and the highest shrinkage was at 90 wt% addition of RHA. However, this increase could be attributed to the removal of initial pores, combined with growth of grains and strong bonding forces to a powder compact (James and Thomas, 1994). The voids were filled and the body became more compacted. This could be attributed to the increase in liquid phase with increased RHA content, which was

able to fill the void between particles, and the release of developed gases from the bulk. The solid grains rearrange and draw close to each other to form more homogeneous and continuous bulk, with a fine water absorption and porosity decreases with soaking time (Prasad *et al*, 2003; Perez *et al*, 2012).

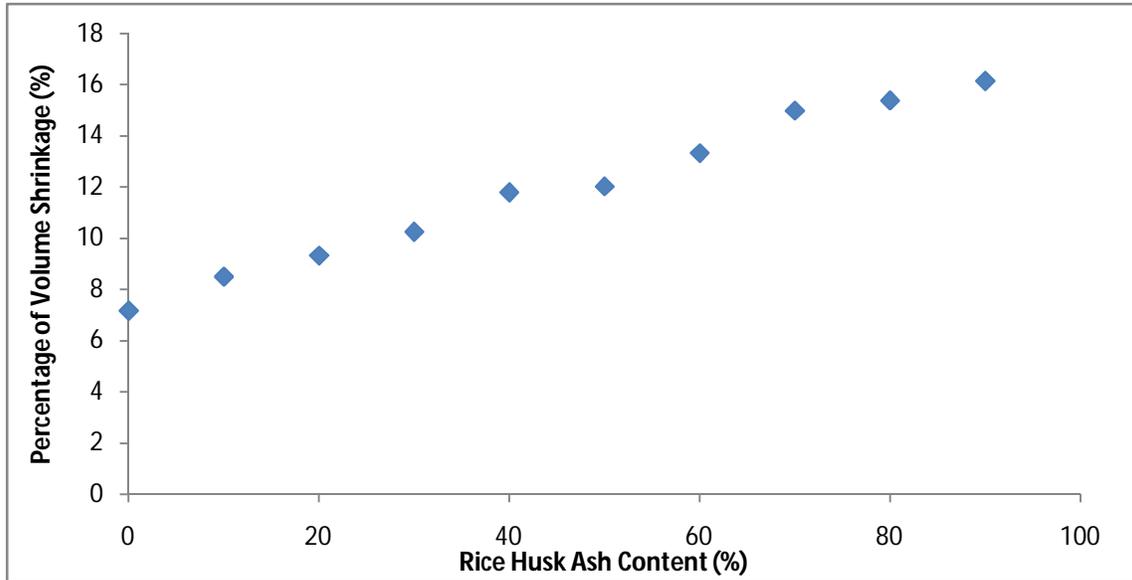


Figure 6: Volume shrinkage Vs RHA content

As shown in Figure 7 the porosity decreases with RHA addition up to 40 wt% after which it began to increase until 80 wt% addition of RHA. This is due to the amorphousness and the high amount of SiO₂. With addition of RHA the quartz starts to dissolve rapidly and produces more SiO₂ content to assist feldspar in dissolving the particles in

porcelain (Prasad *et al*, 2003). It is evident that the least porosity occurs at 40 wt% and highest at 80 wt% addition of the RHA. It is believed that sintering temperature makes the extraction of the amount of silica possible (Nuruddin *et al*, 2009).

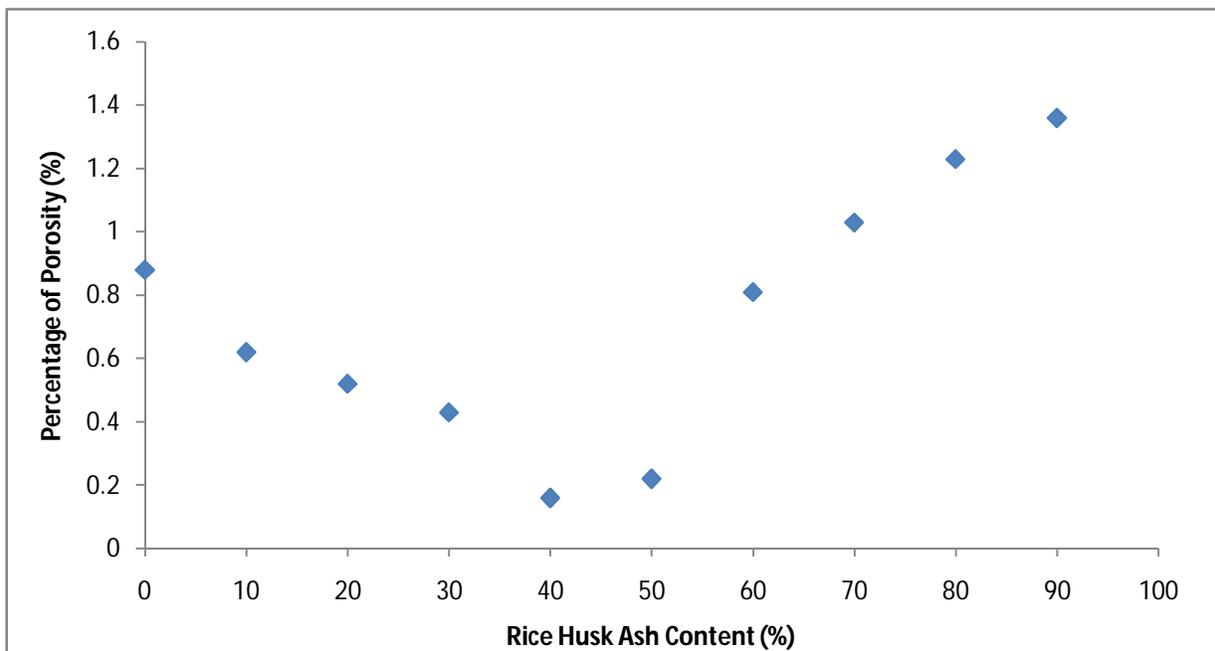


Figure 7: Porosity Vs RHA content

Figure 8 shows the bulk density versus RHA content. The bulk density increases with increase in addition of RHA. The highest bulk density occurs at 40 wt% addition of RHA. Densification is generally achieved

by particles rearrangement and viscous flow. Many sintering mechanisms are contemporarily active (melting, pore coalescence and coarsening) due to wide range of pore and particle size in the green

compacts. However, the densification appears to be rate-controlled by the solubility of solids in the liquid phase and the strong dependence of melt viscosity on temperature and increase in RHA. The bulk density decreases after reaching maximum on further addition of RHA. The decrease in densification is emphasized by coarsening

phenomena. The phase composition seems to reach an equilibrium and the unique transformation are solution / nucleation of mullite and some dissolution of RHA in the liquid phase. Similar trend was obtained by Bribiesa *et al* (1999).

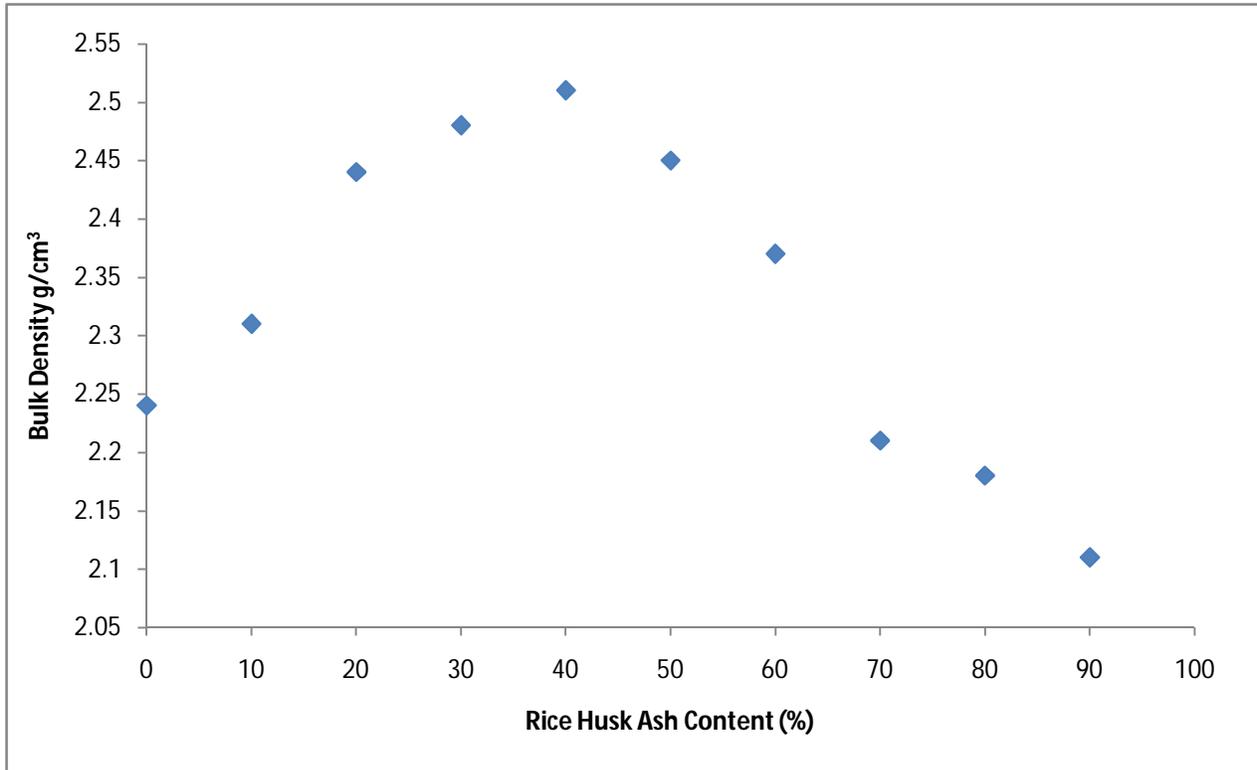


Figure 8: Bulk density Vs RHA content

The results of compressive strength are presented in Figure 9. The results revealed that the compressive strength of the sample with 40 wt% addition of RHA is the highest than that of the control sample. The strength of porcelain has been explained by developing three major theories summarized in the mullite hypothesis by Carty and Senapaty (1998). The felt like interlocking of fine mullites needles are

responsible for the porcelain strength, the matrix reinforcement hypothesized that induced thermal compressive stresses to thermal expansion mismatch lead to compressive strength improvement, and the dispersion-strengthening hypothesis posited that the dispersed particles limit the Griffith flaws, leading to increased compressive strength (Carty and Senapaty, 1998).

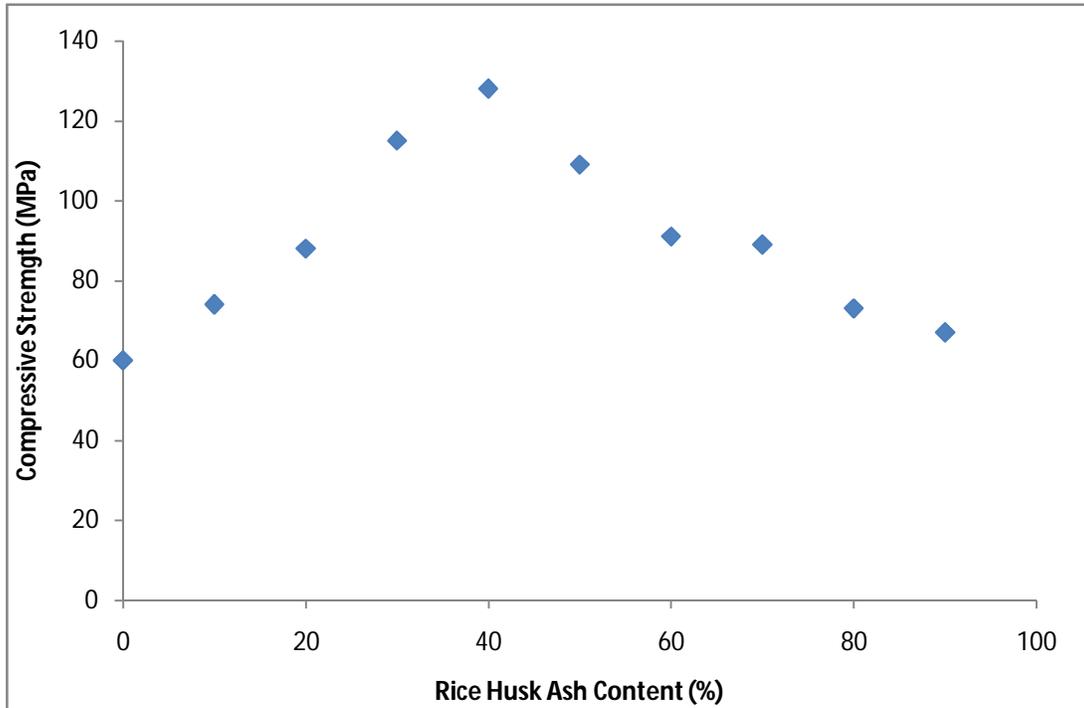


Figure 9: Compressive Strength Vs RHA content

When flux melted, the melting temperature decreases and more glasses are formed, and the pores become smaller and cause porosity and water absorption to decrease, hence the compressive strength increases. Mullite interlocking at higher temperature gives rise to increase in compressive strength (Carty and Senapaty, 1998). Similar results were obtained by Tangchirapat *et al* (2008).

Theoretically, when the water and porosity decrease, compressive strength is expected to increase, and that was exactly what happened, especially between 10% to 40 wt% substitution of RHA. Also, from theoretical perspective, when the bulk density increases, the compressive strength is expected to increase. That was what happened between 10% to 40 wt% substitution of RHA. Further substitution of RHA from 50 wt% to 90 wt% resulted in a graph that is

almost linear, which shows that the increase of RHA within this range contribute to strength of porcelain.

Figure (10a) shows the sintered control sample where the existence of pores can be seen. With the addition of 20 wt% of RHA, densification becomes manifested and the pores drastically become smaller as can be seen from Figure 10b (the arrow pointing to it). In Figure 10c, addition of RHA to 40 wt% resulted in the highest densification with the least pores.

With the 60wt% addition of RHA, pores manifested again as can be seen from Figure 10d. At Figure 10e shows the addition of 90%wt where cracks were developed. This is in conformity with the result obtained by compressive strength and other tests.

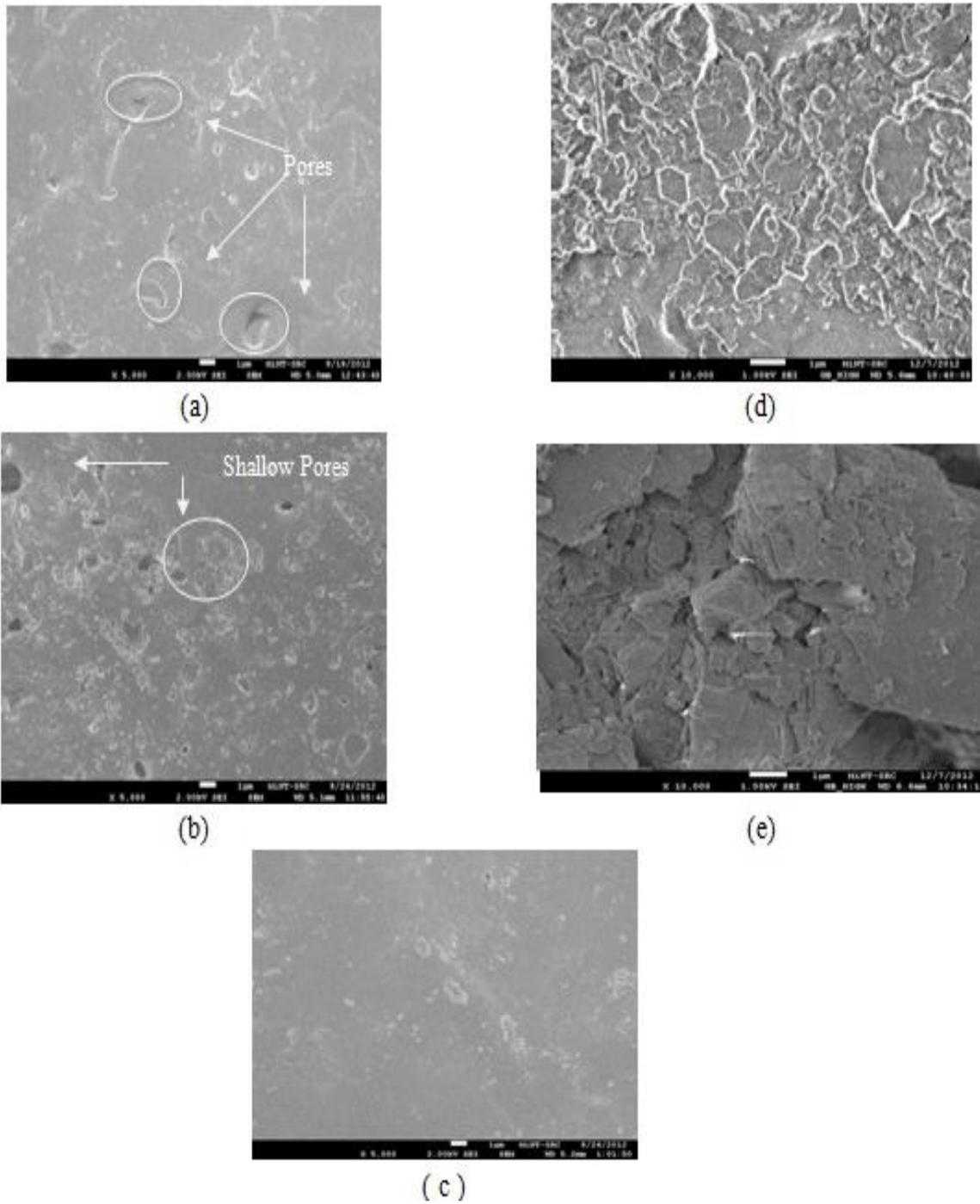


Figure 10: FESEM Micrograph of the samples

CONCLUSION

The RHA sample is completely amorphous, the XRD result revealed that combustion at 700°C produces a very good reactive material. The presence of SiO₂ increases the reactivity of the ash. Since silica from rice husk ash can be prepared under controlled temperature with chemical treatment, it is a potential source of highly active amorphous silica as presented by the XRD result.

Progressive addition of RHA in a porcelain body resulted in early vitrification of the mixtures. The compressive strength result shows that RHA has the potential to be used as an additive material and can improve the strength of the recycled aggregate porcelain. The use of RHA as an additive material on the porcelain body at 40% could give higher compressive strength than porcelain without RHA. This is in conformity with the result obtained from porosity, water absorption and bulk density. It can therefore be concluded that 40wt% addition of RHA into the porcelain body is the best composition.

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REFERENCES

- Amick J.A. (1982), Purification of rice hulls as a source of solar grade silicon for solar cells. *Journal Electrochem Society*, 129, 864–866.
- Bribiesca S., R. Equihua and V. Ilasenor (1999), Photo acoustic thermal Characterization of Electrical Porcelains; Effect of Alumina Additions of Thermal Diffusivity and Elastic Constant. *Journal of the European Ceramic Society* 19, 1979-1985.
- Brinker C.J. and G.W. Scherer (1990), Applications. In: *Sol-Gel Science, The Physics and Chemistry of Sol-Gel Processing*. Academic Press, Inc., San Diego, CA, 839–880
- Carty M.W. and U. Senapaty (1998), Porcelain-Raw Material Processing, Phase Evolution and Mechanical Behaviour. *Journal of the American Ceramic Society*. 81, 3-20.
- Gastaldini A.I.G., G.C. Isaia, T.F. Hoppe, F. Missau and A.P. Saciloto (2009), Influence of the use of RHA on the Electrical Resistivity of Concrete: A Technical and Economic Feasibility Study. *Construction of Building Materials* 23, 3411-3419.
- James A.J. and F.K. Thomas (1994), *Engineering Material Technology*.
- Krishnarao R.V. and Y.R. Mahajan (1995), Effect of acid treatment on the formation of SiC whiskers from raw rice husks. *Journal of European Ceramic Society*, 15, 1229–1234.
- M.K., Naskar, and M., Chatterjee, A novel process for the synthesis of cordierite (Mg₂Al₄Si₅O₁₈) powders from rice husk ash and other sources of silica and their comparative study. *Journal of European Ceramic, Society*, 24(2004), 3499–3508.
- Martin J.L. (1938), *The Desilicification of Rice Hulls and a Study of the Products Obtained*, Louisiana State University, Eunice, LA: MS thesis.
- Motari K.A., M. M., Haslinawati, Z.A. Wahab, H.A.A. Sidek, T.K. Ban and W.A.K. Ghani (2009), Producing amorphous white silica from rice husk. *Journal of Basic Applied Sciences* 1(3), 512-515.
- Nuruddin M.F., N. Shafiq and N.M.L. Kamal (2009), The effects of types of rice husk ash on the porosity of +concrete. *Joint Conferences APSEC-EACEF*, Awana Porto Malai Langkawi, Malaysia.
- Perez M.J., J.M. Rincon and M. Romero (2012), Effect of moulding pressure on microstructure and technological properties of porcelain stoneware. *Ceramics International* 38, 317-325.
- Prasad C.S., K.N. Maiti and R. Venugopal (2001), Effect of rice husk ash in whiteware compositions. *Ceramic Intentional*, 27, 629–635.
- Prasad C.S., Maiti, K.N. and Venugopal R. (2003). Effect of substitution of quartz by rice husk ash and silica fume on the properties of whiteware compositions. *Ceramic International*. 29 (8), 907–914
- Proctor A., P.K. Clark and C.A. Parker (1995), Rice hull ash adsorbent performance under commercial soy oil bleaching conditions. *J. AOCs* 72, 459–462.
- Rodrigues F.A. (2003), Low-temperature synthesis of cements from rice hull ash. *Cement Concrete Res.*, 33, 1525–1529.
- S. Kula, N. Kurama, S., Ay, S. Turan and E. Putun, (1998). Effect of active silica on the microstructure of porcelain, S. Turan, F. Kara, E. Putun, Editors, Proceedings of the International Participation IV on Ceramic Congress, 22–25 September, Eskisehir, Turkey. *Turkish Ceramic Society*, pp. 279–287.
- Siqueira E. J., I.V.P. Yoshoda, L.C. Pardi and M.A. Schiavon (2009), Preparation and Characterization of Ceramic Composites Derived from Rice Husk Ash and Polysiloxane. *Ceramic International* 35, 213-220.
- Tangchirapat W., R. Buranasing and C.J. Chindapsirt (2008), Influence of rice husk-berk ash on mechanical properties of of concrete containing high amount of recycled aggregates. *Construction and Building Materials* 22, 1812-1819.