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Effect of Clay as Additive Element in Recycling Waste Porcelain and Sintering Temperature

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ABSTRACT

Porcelain tile production has been consistently growing worldwide at an approximate rate of 300 million m^2/year. This implies that there is an increasing demand for porcelain raw materials. The industry's commercial success was due to the constant and continuous increase in both technical and aesthetic performance. Due to the complex nature of its raw materials, vigorous firing process and processing approach, porcelain represent the most intricate ceramic system. This paper studied effect of sintering temperature using recycled porcelain and clay as additive in the production of porcelain on physical and mechanical properties of the new product. Waste porcelain was treated with HCl acid, dried in an oven to remove the moisture content and sieve with 50 µm aperture. Clay was added to the treated porcelain powder at 0%, 2%, 4%, 6%, 8% and 10% mixed with a ball mill machine, dry pressed into pallet at 91 MP and sintered at 1100 °C, 1150 °C and 1200 °C for 2 hours soaking time. Physical and mechanical properties such as volume shrinkage, mass loss, bulk density, water absorption and compressive strength as well as microstructural analysis were determined. At a sintering temperature of 1200 °C and clay composition of 2%, the maximum value of compressive strength was 863.15 MPa, indicating the overall maximum. Furthermore, at this composition, the value of the mass loss, shrinkage, bulk density and water absorption of porcelain waste with clay addition was 5.93%, 64%, 2.53% and 6.09%, respectively.

Keywords: *Clay; porcelain waste; microstructure.*

1. INTRODUCTION

Porcelain is a traditional ceramic material that has been extensively used for different applications such as household, scientific, and engineering purposes. Among the major advantages of porcelain are its low water absorption, durability, translucency, and high mechanical strength [1]. Porcelain was mainly used for household flooring and wall façades because of its cold shocks, high strength against pressure and humidity [2].

Recently, porcelain products have been considered the fastest growing market internationally, this is attributed to their aesthetic and promising technical characteristics [3]. As the world population has witnessed tremendous growth in recent years, the most important economic and environmental solution for the porcelain and ceramic industries is the recycling of materials that do not easily decompose naturally or by atmospheric precipitation, such as waste porcelain [4]. To safeguard the environment and conserve the limited natural resources available, recycling wastes like porcelain is therefore inevitable [4].

Researchers and industries have sorted for improvement of the compressive strength of porcelain and ceramic materials; hence, the use of waste porcelain as coarse aggregate was suggested to quench the researcher's thirst, it was reported that, material containing waste porcelain and sintered at 850°C revealed an increased strength of 20% higher than the normal material [5]. The research further revealed that, apart from the improved mechanical strength, the heat resistance was also increased. In making the final product of porcelain, each component plays a specific role, clay addition helps in developing mullite and crystalline phase during firing, furthermore, it helps in forming by providing plasticity and dry mechanical strength during processing [6]. For feldspars, at low temperature it helps to develop a glassy phase, assisting the sintering process, and enabling virtually zero (<0.5%) open porosity and a low level of closed porosity (<10%) to be achieved. Similarly Quartz is responsible of providing thermal and dimensional stability due to its high melting point [1].

Furthermore, in another research by Guerra et al. [7] reported that, waste porcelain can be used to improve compressive strength. It was further reported that, incorporation of waste porcelain at 9% lead to improvement of compressive strength. Similarly, Cachim [8] relate that, there are variations in some experimental results which were attributed to different types of waste porcelain and nature of disposal. In the last few years, the production of ceramic porcelain has steadily stood at 13 billion square meters [9], this indicated that, there is high demand of raw materials amounting to million tons yearly. Hence, porcelain industries have been reported to have the best technology that proved to recycle their own waste products such as porcelain waste [9]. Porcelain waste has been reported to serve as remedy for the weakness of limestone used in concrete and hence incorporation of porcelain waste improved the concrete heat resistance [2].

De-Silve et al. [10] reported that, clays have been known as the materials used for bricks and porcelain production, the continues mining of clays posed a danger for the loss of virgin topsoil resources. It is further reported that, use of clay bricks for walling and in ceramic industries has been exponentially growing. Research shows that, bricks and porcelain produced with different clay compositions divulged a satisfactory result compared to the conventional standards. Ofarrell et al. [11] reported that, 6% replacement of hot dip galvanizing sludge with clay and sintered at 1020°C yielded a satisfying mechanical and EN standards. Similarly, clay as replacement of industrial wastewater treatment plant sludge at 20% for bricks production fired at 960°C and 1000°C sintering temperature conformed to the Chinese National Standard [12]. Furthermore, it was reported that, clay as replacement of waste glass at 25% and sintered at 850°C, indicate an improvement in the compressive strength for up to 37%. Consequently, to maintain aesthetic requirements, it was reported that, ceramic sludge replacement with clays at 10% and 1050 °C sintering temperature is promising [13]

Currently, most of the studies available in literature regarding recycling and reuse of industrial wastes as alternative raw materials focused on the use of palm oil fuel ash (POFA) and rice husk ash (RHA). In this regard, this study presents the effect of using clay as additive element in recycling of waste porcelain and sintering temperature.

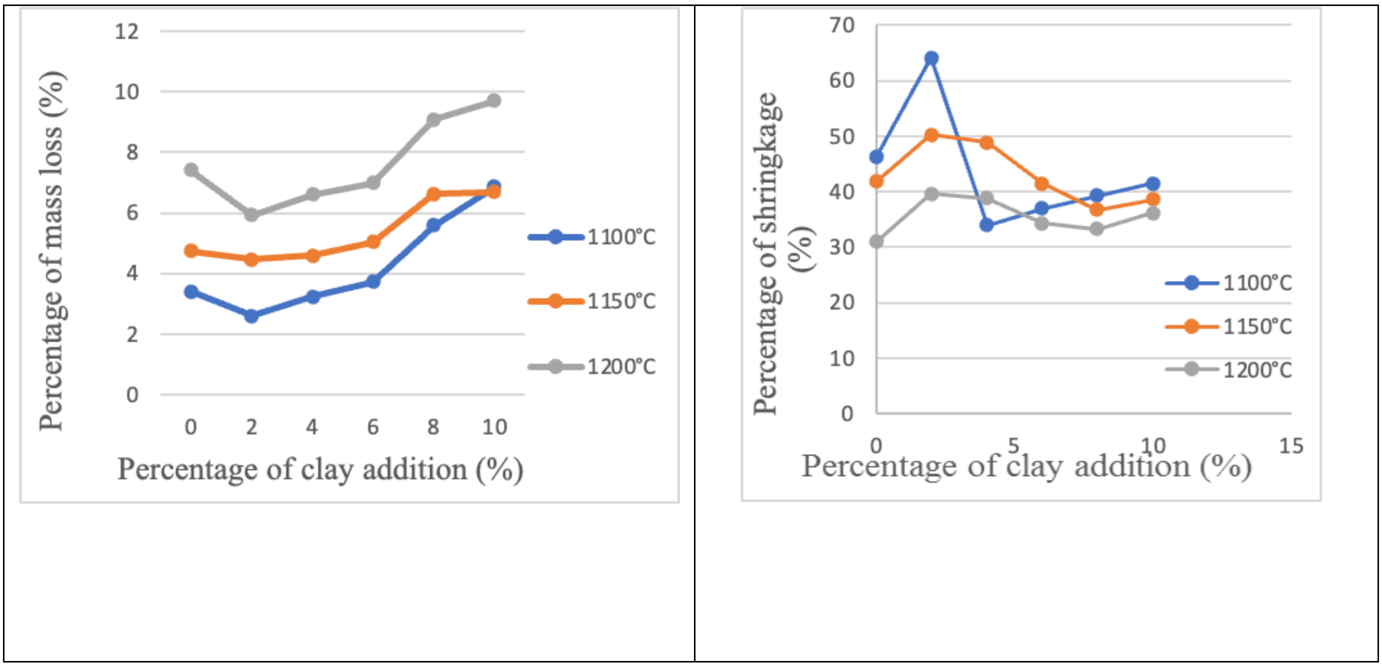
2. MATERIALS AND METHODS

Clay and porcelain waste were crushed, sieved, and treated with HCl acid. The powder was dried in an oven and mixed using a ball mill machine. Polyvinyl alcohol (PVA) was added to each composition as a binder, ground with agate mortar, and dry pressed into pallets using a Hydraulic Pressed Carver at 200 mm diameter by 3 mm thickness in a cylindrical shape. The solid sample is then vacuum pressed with an American Cold Isostatic pressing machine (CIP360) for about 5 min and then sintered at different sintering temperatures of 1100 °C, 1150 °C, and 1200 °C in a Prothern Furnace at a heating and cooling rate of 1°C/min and 5 °C/min respectively. Scanning Electron Microscopy (SEM), X-Ray Diffraction analysis (XRD), volume shrinkage, mass loss, bulk density, and water absorption were used to find the effect of clay and waste porcelain on the physical and mechanical properties of porcelain.

3. RESULTS AND DISCUSSION

3.1. Physical Properties

Physical properties are one of the most important factors considered during production in the ceramic industry. The effect of clay addition in recycling waste porcelain and sintering temperature on volume shrinkage, mass loss, bulk density, and water absorption has been determined and presented in Figure 1 below.



(a) (b)

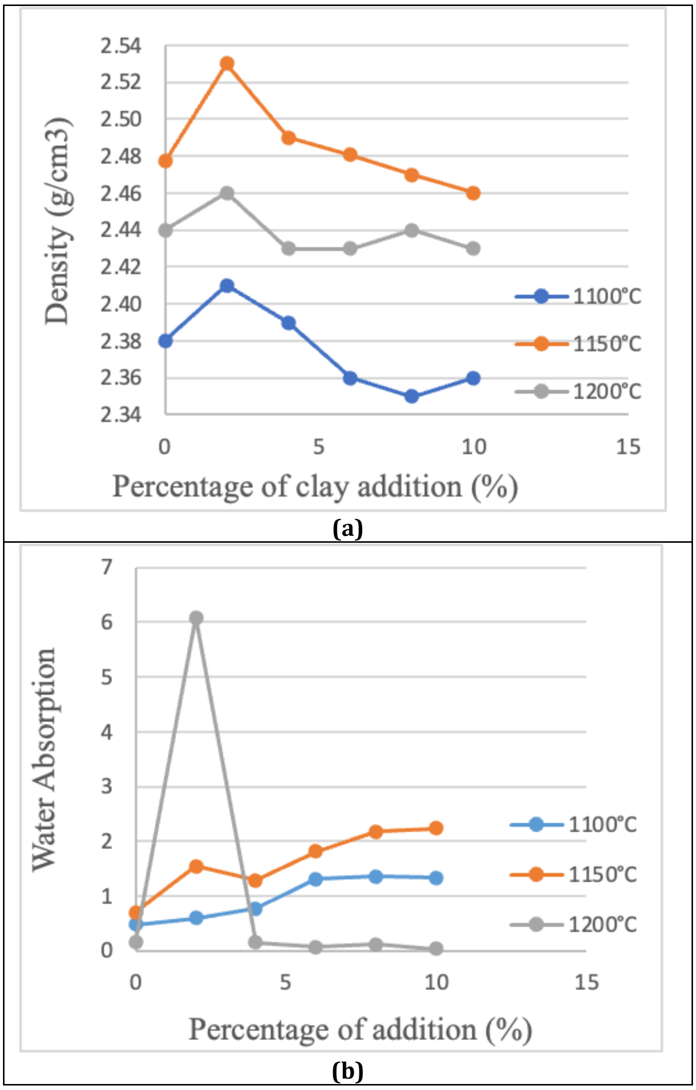
Figure 1 Graph of the percentage of (a) mass loss and (b) volume shrinkage of porcelain waste with clay addition.

Figure 1(a) shows the percentage of mass loss versus percentage of clay addition. It is important to note that mass loss steadily increased (from 2% to 10%) as the percentage of clay addition increased for all the sintering temperatures (1100 °C, 1150 °C and 1200 °C respectively), furthermore, the maximum value of mass loss was achieved at 10% composition for 1200°C. As the sintering temperature increases, the mass of the porcelain is reduced, and hence the porosity is also reduced. Clay addition ensure the plasticity needed for shaping and helps in maintaining the mechanical strength of unfired porcelain as reported by Dondi et al. [14]. This result indicated that, addition of clay on recycling waste porcelain has a great deal for producing lightweight porcelain. Similarly, Figure 1(b) shows the percentage of volume shrinkage versus percentage of clay addition, it is significant to note that, the volume shrinkage decreased as the percentage of clay addition increases for sintering temperatures of 1150°C and 1200°C respectively this is due to the nature of clay that consist of higher amount of SiO_2, meanwhile, the lowest volume shrinkage was reported at 1100°C at 4%, whereas, at 2% within the same sintering temperature the maximum value was recorded as 64% and this is due to the rise in the densification of the porcelain. Furthermore, it was reported that, formation of bloating and micro cracks is eminent as a result of increment of sintering temperature and over firing.

Similarly, Figure 2(a) shows the graph of bulk density versus percentage of clay addition and sintering temperature, from the graph, it can be deduced that, the density increased as the composition of clay increased from 0% to 2% for all the sintering temperatures (1100°C, 1150°C and 1200°C respectively), the maximum value of bulk density was attained at 1150°C and composition of 2% as 2.53 g/cm^3, whereas the lowest value was achieved at 1100 °C, 8% composition as 2.35 g/cm^3, the graph further revealed that, the highest bulk density was achieved at sintering temperature 1150°C for all the percentages of clay addition which is attributed to the acid treatment that enhanced the purity of SiO_2 content and hence improve the bulk density. Consequently, Figure 2(b) shows the graph of water absorption versus composition of clay addition. It is therefore pertinent to note that, at 1200°C and 2% composition the water absorption abruptly increased from 0.15% to 6.09% due the high value of bulk density at 2% composition. At this composition (2%), the highest bulk density and lowest volume shrinkage were achieved, indicating a promising match for an improved technological property. Similarly, at 1100°C and 1150°C, the water absorption increased steadily for all the compositions from 0% to 10%, thus, it can be concluded that, the higher the sintering temperature the lower the percentage of water absorption. Consequently, further addition of clay and increasing the sintering temperature leads to formation of porosity and micro cracks which has a direct proportion to density decrease. Figure 2 shows graph of the percentage of (a) bulk density and (b) water absorption of porcelain waste with clay addition and sintering temperature.

3.2. Mechanical Properties

The major component that determines the mechanical properties of porcelain in the ceramic industry is compressive strength. The result of the compressive strength versus percentage of clay addition is presented in Figure 3. According to Figure 3, the compressive strength increased as the sintering temperature increased. At 1100°C, the highest value of the compressive strength was recorded as 550.26 MPa at 2% of clay addition, similarly, at 1150°C the highest value of compressive strength is 745.49 MPa at 8% of clay addition.



(a)

(b)

Figure 2 Graph of the percentage of (a) bulk density and (b) water absorption of porcelain waste with clay addition and sintering temperature.

The maximum value of compressive strength recorded for 1200°C sintering temperature is 863.15 MPa at 2 % of clay addition, indicating the overall maximum, complete dissolution of quartz and subsequent matrix reinforcement have been reported to be the rationale behind the improvement of compressive strength, another researcher suggested that porosity plays a vital role in determining the strength of porcelain, thus, the improvement was due to complete densification and hence reduced porosity [6]. Mostly the graph shows an increase and decreasing pattern of the compressive strength with subsequent addition of clay and the sintering temperature, which is an indication that, sufficient amount of SiO_2 have been achieve, because presence of excess SiO_2 lead to the reduction of strength.

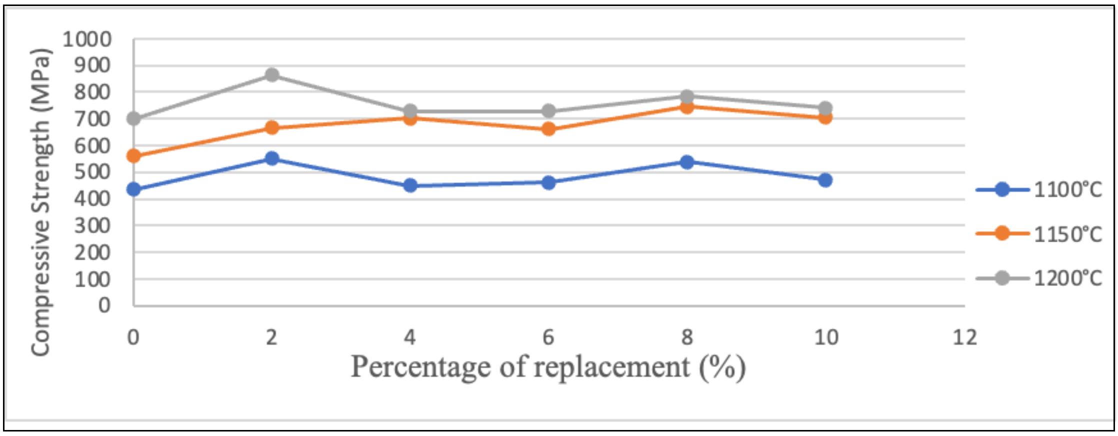


Figure 3 The graph of the compressive strength of porcelain waste with clay addition

3.3. X-ray Diffraction (XRD)

Figure 4 shows the X-ray diffraction (XRD) pattern and XRD analysis of the samples based on three different percentages of clay addition with different temperatures, that is, 1000°C, 1150°C, and 1200°C, respectively. The graph indicates that the major dominant patterns are quartz and mullite for all the different sintering temperatures. From the graph, it is worth noting that the highest peak is that of the quartz element, while the lowest peak is that of the mullite element. Clearly, the graph shows that at sintering temperatures from 1100 °C to 1200 °C there is an imminent decrease in the peak intensities of quartz and mullite. Whereas, at 1100 °C, mullite crystals were present in a minute quantity that is hardly noticed and had higher peak intensities than quartz. Similarly, at 1150°C, there is a more pronounced amount of mullite and very little quartz, whereas at 1200 °C, both quartz and mullite are present, with each of them nearly dominating the order. This is a clear indication that, at this sintering temperature, the crystals are very clear and densification is enhanced.

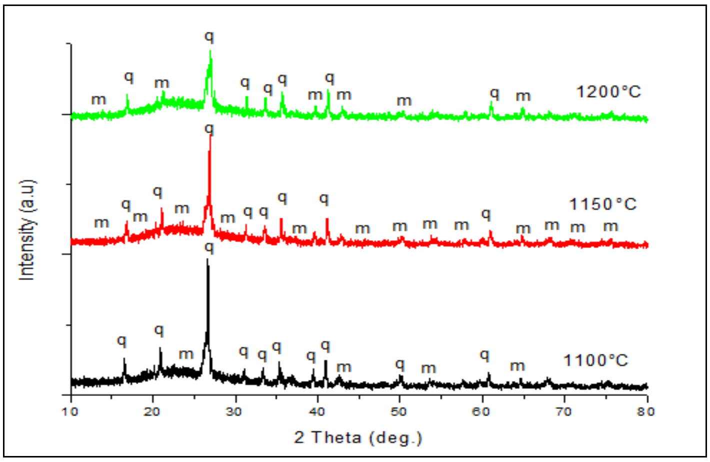


Figure 4 X-ray Diffraction (XRD) pattern of porcelain waste with clay addition.

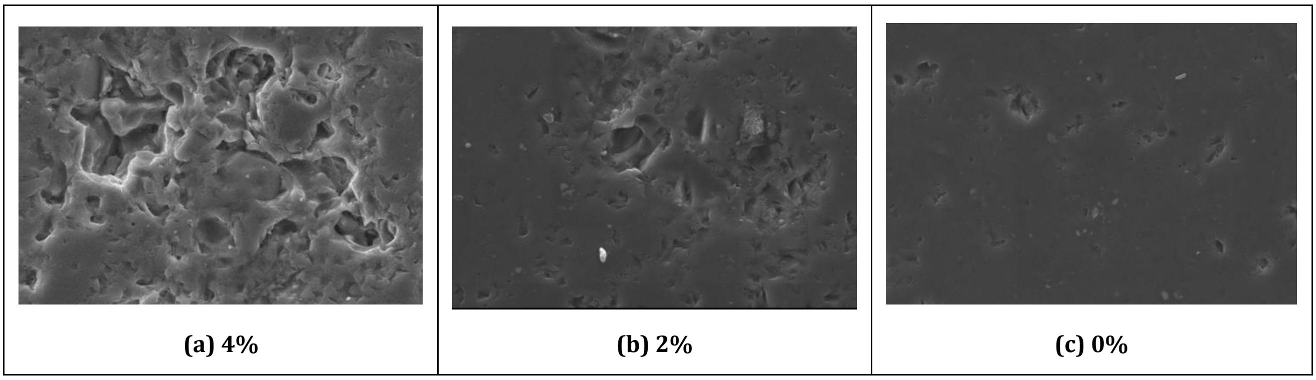
3.4. Microstructure Study

The microstructural analysis of the samples was studied and analyzed using SEM. Figure 5 shows the micrograph of SEM of the sample at 0%, 2% and 4% of clay addition with different sintering temperatures (1100°C, 1150°C and 1200°C).

Figure 5 (a) shows that the sample has more and bigger pores compared to other samples. In Figure 5(b), the sample has a lower size and open porosity due to the lower amount of clay in the sample, and in Figure 5(c), the sample has the lowest size and amount of open porosity due to no addition of clay in the sample, which is a clear indication that clay is a key player in determining the porosity of the sample. Besides that, Figure 6 shows the graph of EDX analysis with a scanning electron microscope (SEM). From that graph, it was observed that the elements of the sample mainly contain silica, aluminum, oxygen, and more; this indicated that the XRD is in line with the result of EDX.

4. CONCLUSIONS

This research presented a study on the effect of clay addition and sintering temperature on the recycling of waste porcelain. From the results, it has been concluded that the optimum value for both the physical and mechanical properties of the sample is 2% clay addition at 1150°C. At this composition, the mass loss, shrinkage, bulk density, and water absorption of porcelain waste with clay addition were 5.93%, 64%, 2.53%, and 6.09%, respectively. Moreover, the sample gives the highest value of compressive strength, which is 863.15 MPa. The X-ray diffraction analysis result shows that the crystalline and glassy phases were obtained in porcelain waste with the addition of clay sintered at 1100°C, 1150°C, and 1200°C. For scanning electron microscopy (SEM) and EDX, the major elements in the sample are silica, aluminum, oxygen, and more. The result suggested that higher clay addition tends to create a porous sample.



(a) 4% **(b) 2%** **(c) 0%**

Figure 5: The micrograph of SEM of the sample at 0%, 2% and 4% of clay addition at all the 3 different sintering temperatures (1100°C, 1150°C and 1200°C).

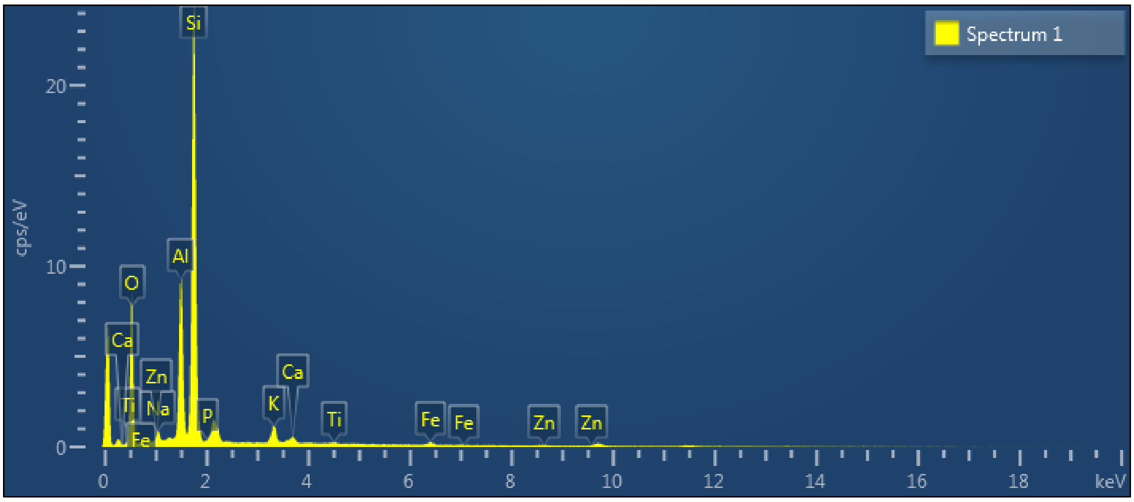


Figure 6: Graph of EDX with Scanning Electron Microscope (SEM).

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REFERENCES

[1] S. G. Durumin-Iya, M. Zaky, Siti Nur Raiedah Binti Mohd Hanafiah Noh, and Siti Fairuz Binti Mat Radzi, “Effect of Quartz Replacement with Palm Oil Fuel Ash for Triaxial Porcelain Composition of 50%: 40%: 10%,” *Int. J. Nanoelectron. Mater.*, vol. 15, p. 1, 2022.

[2] Z. Keshavarz and D. Mostofinejad, “Effects of high-temperature exposure on concrete containing waste porcelain coarse aggregates and steel chips,” *J. Build. Eng.*, vol. 29, p. 101211, 2020.

[3] Y. E. R. Calderón *et al.*, “Development of ceramic porcelain stoneware pastes by the revalorization of Colombian clays subjected to bleaching process,” *Appl. Clay Sci.*, vol. 178, p. 105128, 2019.

[4] O. R. Njindam *et al.*, “Effect of glass powder on the technological properties and microstructure of clay mixture for porcelain stoneware tiles manufacture,” *Constr. Build. Mater.*, vol. 170, pp. 512-519, 2018.

[5] M. Talaei and D. Mostofinejad, “Mechanical properties of fiber-reinforced concrete containing waste porcelain aggregates under elevated temperatures,” *Constr. Build. Mater.*, vol. 289, p. 122854, 2021.

[6] M. Z. Noh, H. U. Jamo, and Z. A. Ahmad, “Effect of temperature and composition of palm oil fuel ash on compressive strength of porcelain,” *Appl. Mech. Mater.*, vol. 660, pp. 173–177, 2014.

[7] I. Guerra *et al.*, “Eco-efficient concretes: The effects of using recycled ceramic material from sanitary installations on the mechanical properties of concrete,” *Waste Manage.*, vol. 29, no. 2, pp. 643–646, 2009.

[8] P. B. Cachim, “Mechanical properties of brick aggregate concrete,” *Constr. Build. Mater.*, vol. 23, no. 3, pp. 1292–1297, 2009.

[9] S. Conte *et al.*, “Glassy wastes as feldspar substitutes in porcelain stoneware tiles: Thermal behaviour and

effect on sintering process," *Mater. Chem. Phys.*, vol. 256, p. 123613, 2020.

[10] G. S. De Silva and E. Hansamali, "Eco-friendly fired clay bricks incorporated with porcelain ceramic sludge," *Constr. Build. Mater.*, vol. 228, p. 116754, 2019.

[11] M. Ofarrell, S. Wild, and B. Sabir, "Pore size distribution and compressive strength of waste clay brick mortar," *Cement Concrete Compos.*, vol. 23, no. 1, pp. 81–91, 2001.

[12] J. Martín-Márquez, M. Rincón, and M. Romero, "Effect of firing temperature on sintering of porcelain stoneware tiles," *Ceram. Int.*, vol. 34, no. 8, pp. 1867–1873, 2008.

[13] S. Yürüyen and H. Ö. Toplan, "The sintering kinetics of porcelain bodies made from waste glass and fly ash," *Ceram. Int.*, vol. 35, no. 6, pp. 2427–2433, 2009.

[14] M. Dondi, M. Raimondo, and C. Zanelli, "Clays and bodies for ceramic tiles: Reappraisal and technological classification," *Appl. Clay Sci.*, vol. 96, pp. 91–109, 2014.