Effect of firing temperature on triaxial electrical porcelain properties made from Tanzania locally sourced ceramic raw materials

Blasius NGAYAKAMO

MSc Materials Scientist and Engineer at NM-AIST. Department of Materials Energy Science and Engineering. Fields of interest: ceramic raw materials, flux materials and high voltage porcelain insulators.

S. Eugene PARK

Associate Professor and chair, Materials and Energy Science and Engineering at NM-AIST Department of Materials Science and Engineering. Fields of interest: Ceramic and sustainable Energy Engineering.

BLASIUS NGAYAKAMO ▪ The Nelson Mandela African Institution of Science and Technology, Department of Materials Energy Science and Engineering ▪ henryblasius@gmail.com

S. EUGENE PARK ▪ The Nelson Mandela African Institution of Science and Technology, Department of Materials Energy Science and Engineering

Érkezett: 2018. 02. 16. ▪ Received: 16. 02. 2018. ▪ <https://doi.org/10.14382/epitoanyag-jsbcm.2018.19>

Abstract

The study has investigated the effect of firing temperature during the production of technical triaxial electrical porcelain, for electrical insulation applications using Tanzania locally sourced ceramic raw materials. The green triaxial porcelain samples containing 50 wt% of Pugu kaolin, 35 wt% of Same clay and 15 wt% of feldspar were produced and fired at 1200 °C-1300 °C with a heating rate of 10 °C/min (dwell time of 1.5h) and cooled at 10 °C/min to a room temperature. X-ray diffraction technique was used to investigate phases developed in the triaxial electrical porcelain after firing process. The main crystalline phases revealed were mullite and quartz. The technological properties of the triaxial electrical porcelain such as water absorption, apparent porosity, bulk density, bending and dielectric strength were determined for each porcelain sample fired at high temperature. The optimum physical-mechanical and electrical properties were found at 1250 °C. However, the triaxial electrical porcelain properties were found to decrease with the increase in firing temperature.

Keywords: Firing temperature, triaxial electrical porcelain, physical-mechanical and dielectric properties

Kulcsszavak: Égetési hőmérséklet, triaxiális szigetelő porcelán, fiziko-kémiai és diektromos jellemzők

1. Introduction

Triaxial electrical porcelain is composed of clay, feldspar which are locally sourced ceramic raw materials and other filler materials such as quartz and alumina. The raw materials play specific roles in influencing the properties and performance of the final products. Clay $[Al_2Si_2O_5(OH)_4]$ provides plasticity, quartz (SiO_2) maintains the shape of the porcelain structure during firing, and feldspar $[KxNa1-x(AlSi_3O_8)]$ promotes vitrification. The three ceramic raw materials place electrical porcelain in the phase system $[(K, Na)_2O-Al_2O_3-SiO_2]$ in terms of oxide hence referred as triaxial porcelain [1, 2]. Traditional ceramic raw materials are the potential candidate materials for the production of triaxial electrical porcelains. The use of traditional ceramics as raw materials instead of industrial chemicals is highly preferred due to the lower price of the raw materials [3].

The properties of triaxial electrical porcelain are contributed to the variations in the composition of the raw materials, the method of production, and the firing temperature adopted [1]. The sintered triaxial porcelain product contains mullite $(Al_6Si_2O_{13})$ and undissolved quartz (SiO_2) crystals embedded in glassy phase which result from the liquid phase formed by the melting of feldspar in the raw materials [4].

Therefore the desired properties of triaxial electrical porcelains are achieved particularly during the firing process since the technological properties of clay-based ceramics products depend on firing conditions such as temperature [5, 6]. However, other technological properties which are

evaluated to determine the performance of the ceramic product after firing are water absorption, firing shrinkage and bending strength [6]. During the firing process, the triaxial porcelain body undergoes several phase transitions, during which both composition and structure change significantly which influence triaxial porcelain properties at the end of the firing process [5]. Hence, the properties of the triaxial porcelain are mainly influenced by sufficient development of mullite during firing process since the development of mullite in the porcelain is highly associated with firing temperature of the porcelain which should not be below 1150 to 1200 °C for the mullite forming processes to be completed [5, 7]. Therefore, the development of the physical-mechanical and dielectric properties of porcelain are contributed by each phase developed during firing which depends on the concentration and microstructural attributes which are influenced by temperature and the chemical composition of the raw materials which is an important factor because of its effects on porcelain properties [8-11]. Since the effect of firing temperature on the electrical porcelain properties made from Tanzania locally sourced ceramic raw materials is not reported. Therefore, the work intends to evaluate the effect of firing temperature on the triaxial electrical porcelains made from Tanzania locally sourced ceramic raw materials. However, the study focuses also on the phase changes, surface morphology development as well as the physical-mechanical and dielectric properties of the triaxial electrical porcelain sample due to change in firing temperature.

Fig. 1. X-ray diffraction patterns of Pugu kaolin, Same clay and feldspar [16]
1. ábra Pugu kaolin, Same agyag és földpát röntgendiffraktogramjai [16]

2. Experimental procedures

Pugu Kaolin was collected from the Pugu hills, 35 km west of Dar es Salaam, Same clay and feldspar from Same, Kilimanjaro region in the northern zone of Tanzania. The ceramic raw materials were crushed and ball milled to reduce their size. The particle size less than 106 μm was achieved by using sieve shaker Model RX-29-10 digit. The chemical composition of the raw materials was analyzed by using X-Ray Fluorescence (XRF) PANalytical, Model: Minipal4 (PW4030)-Rh X-Ray Tube, 30kV, 0.002mA and the results are presented in Table 1. The examination of the surface morphology of the porcelain sample was carried out by Scanning Electron Microscope (SEM) Model: JEOL JSM-6335F having a resolution of 10 μm at 2kV. The crystalline phase analysis of the porcelain insulator was analyzed by X-ray diffractometer Model: Bruker D2-PHASER-40Kv/44mA. Six triaxial porcelain samples were produced by varying the composition of the locally sourced materials by 50%wt of Pugu kaolin, 35%wt of same clay and 15%wt of feldspar. The powder mixtures were uniaxially compacted into rectangular shapes at 10 MPa. The porcelain green body samples were seasoned at a room temperature for 5 days and they were oven dried at the temperature of 110 $^{\circ}\text{C}$ for 24 hrs. The sintering of porcelain samples was done at 1200, 1250 and 1300 $^{\circ}\text{C}$ for 1.5 hrs at the ramp rate of 10 $^{\circ}\text{C}/\text{min}$ in each firing process. The sintered porcelain bodies were left to cool at 10 $^{\circ}\text{C}/\text{min}$ to room temperature and were subjected to physical-mechanical properties and dielectric strength analysis.

3. Results and discussion

3.1 Chemical composition of the raw materials

The chemical compositions of the raw materials in form of their oxides are presented in Table 1. The study results reveal that both clays have the higher content of silica and alumina. However, feldspar and Pugu kaolin have a higher content of Hematite (Fe_2O_3) compared to Same clay. The literature reports that small amount of coloring oxides such as Fe_2O_3 and TiO_2 less than 0.9% may be accepted for porcelain wares production [12]. However, a considerable high amount of Fe_2O_3 in Pugu kaolin and feldspar may not be accepted as

they may impart yellowish and reddish color in porcelain wares unless beneficiated. Feldspar has considerable higher alkaline oxide K_2O than Pugu kaolin and Same clay. During the sintering process, the alkaline oxide K_2O melts and forms the liquid phase that contributes to densification at higher temperatures due to the formation of the glassy phase. Nevertheless, the quantities of the alkaline oxides depend on the mineralogical nature of the clays and their reactivity during melting of the clay minerals [13]. The alkaline oxides (K_2O and Na_2O) play a significant role towards vitrification, phase transformation and mullite grain growth [14, 15].

Oxides	Pugu kaolin	Same clay	Feldspar
SiO_2	60.0	60.4	57.1
Al_2O_3	30.3	13.9	14.0
Fe_2O_3	3.95	1.40	3.08
MnO	0.021	0.00	0.32
CaO	0.39	0.00	1.0
Na_2O	0.00	0.04	0.20
K_2O	2.14	2.6	12.09

Table 1. Chemical composition of raw materials
1. táblázat Alapanyagok kémiai összetétele

3.2 Mineralogical composition of the raw materials

The X-ray diffraction patterns of the ceramic raw materials before and after firing are presented in Fig. 1 as reported by [16]. The result shows phase compositions of both Pugu kaolin and Same clay are kaolinite, however, Pugu kaolin showed the development of crystalline phases of mullite and quartz at a temperature of 1400 $^{\circ}\text{C}$. In addition, Same clay was observed to form cristobalite and sillimanite above 1200 $^{\circ}\text{C}$. Feldspar contains albite, and microcline, tridymite, and quartz. Since the major components of interests are potassium feldspar ($\text{K}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 6\text{SiO}_2$), sodium feldspar ($\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 6\text{SiO}_2$), and lime feldspar ($\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 6\text{SiO}_2$). However, the results indicate that feldspar deposit contains a high content of potash feldspar compared to soda feldspar which is also supported by the chemical composition by XRF that is K_2O is 12.09% while

Na₂O is only 0.20%. So feldspar deposit is, therefore, a potash feldspar. Feldspar promotes vitrification of the porcelain insulator at the end of the sintering process.

3.2 Characterization of fired triaxial porcelain samples

Fig. 2 presents the results of water absorption, apparent porosity, and bulk density respectively for the porcelain samples versus firing temperature. The figure shows that the best values for physical properties for triaxial electrical porcelain are achieved at the firing temperature of 1250 °C. This might be due to the formation of the liquid phase and densification at this firing range. However, the values of water absorption, apparent porosity, and bulk density were observed to decrease at higher firing temperature. This might be due to the expansion of trapped water bubbles inside the porcelain matrix and change in the composition of the glassy phase [1, 3]. The results of the study are in agreement with the works of [1, 3, 13]. The authors reported that water absorption and bulk density increased due to vitrification and densification of the porcelain samples. However, the physical properties were observed to vary due to the decrease of vitrification range and an increase of firing temperature due to the expansion of trapped water bubbles inside the porcelain sample at high firing temperatures. Generally, the variation of the physical properties of the triaxial electrical porcelain might have been caused by the method of production, chemical and mineralogical properties of the raw materials.

Fig. 2. Physical properties of triaxial electrical porcelain sample versus firing temperature

Fig. 2. Triaxiális szigetelő porcelán fizikai jellemzői az égetési hőmérséklet függvényében

Fig. 3 shows changes in the mechanical strength of triaxial electrical porcelain with firing temperature. The trend shows that the increase of mechanical strength of porcelain sample may be due to increased densification, vitrification and in absence of microcracks. The best mechanical strengths (both bending and compressive strengths) were obtained at 1250 °C. However, the mechanical strengths began to decrease above 1250 °C due to closed pores development and a considerable amount of cracks on the surface of the porcelain samples. The results of the current study are in agreement with the previous studies as reported in the works of Kitouni *et al.*, [13] and Olupot *et al.*, [1]. The authors have reported that the mechanical strength increases due to increased densification with temperature and tends to decrease due to development of pores at high firing temperature. However, the mechanical strength was found to decrease with

the increase of the firing temperature due expansion of closed pores and microcracks [1]. Hence the mechanical strength of a porcelain sample is strongly dependent on the defects such as pores and cracks [13].

Fig. 3. Variation of mechanical strength of triaxial electrical porcelain sample versus firing temperature

Fig. 3. Triaxiális szigetelő porcelán mechanikai jellemzői az égetési hőmérséklet függvényében

Fig. 4 shows the variation of the dielectric strength of porcelain samples fired at 1200, 1250 and 1300 °C. The trend shows that the dielectric strength increases with an increase in firing temperature and began to decrease with further temperature rise at 1300 °C. The increase of the dielectric strength of electrical porcelains is due to increased vitrification range of the electrical porcelains samples. The results of the current study are also reported by Olupot *et al.*, [1]. The authors evaluated ceramic raw materials from Uganda for electrical porcelain production. The authors obtained the highest dielectric strength of 19kV/mm at 1250 °C. However, above 1250 °C, the samples became more porous due to change in the composition of the glassy phase. The dielectric strength was found to decrease with the increase of firing temperature which affected vitrification range and the dielectric properties of the triaxial electrical porcelain.

Fig. 4. Variation of dielectric strength of triaxial electrical porcelain sample versus the firing temperature

Fig. 4. Triaxiális szigetelő porcelán dielektromos jellemzői az égetési hőmérséklet függvényében

In Fig. 5 the X-diffraction pattern of triaxial electrical porcelain is presented. The diffractogram confirms that the mullite and quartz phases are present in the porcelain insulator. Both phases promote the mechanical and dielectric properties

of the porcelain insulator. However, high peaks of quartz may lead to high amount of glassy phase which may lower the dielectric strength of the porcelain insulators but not the mechanical strength of a porcelain insulator which is affected by microcracks. The high amount glassy phase provides free movement of mobile ions such as Na^+ , K^+ , and Al^{3+} which increases the conductivity [17].

Fig. 5. X-ray diffraction pattern of a porcelain insulator fired at 1250 °C
Fig. 5. 1250 °C hőmérsékleten égetett szigetelő porcelán röntgendiffraktogramja

Fig. 6 shows the examination of the surface morphology using the Scanning Electron Microscope (SEM) Model: JEOL JSM-6335F having a resolution of 10nm at 2kV. It was evidenced the densification on the surface of the triaxial electrical porcelain sample after the firing process was completed.

Fig. 6. SEM micrographs showing densification of triaxial electrical porcelain sample at 1250 °C

Fig. 6. 1250 °C hőmérsékleten égetett triaxiális szigetelő porcelán elektronmikroszkópos felvétele

4. Conclusions

In this research work, the effect of firing temperature on triaxial porcelain samples properties was investigated. At the optimum firing temperature of 1250 °C, the best physical-mechanical and dielectric properties were achieved. However, firing beyond 1250 °C resulted in progressive deterioration of the physical-mechanical and the dielectric properties of the electrical porcelain samples. This might have been caused by the development of microcracks and high content of glassy phase caused by high peaks of quartz. So it is imperative to be aware that, the actual firing temperature and its influence on the triaxial electrical porcelain properties depend on the chemical composition of the materials under study. Therefore, the locally sourced materials need to be evaluated from time to time in order to avoid deviation of the desired triaxial electrical porcelain properties during the firing process.

References

- [1] Olupot, P. W. (2006): Assessment of ceramic raw materials in Uganda for electrical porcelain, *KTH*.
- [2] Carty, W. M. – Senapati, U. (1998): Porcelain—raw materials, processing, phase evolution, and mechanical behavior. *Journal of the American Ceramic Society*, 1998. 81(1): p. 3-20. <https://doi.org/10.1111/j.1151-2916.1998.tb02290.x>
- [3] Kasrani, S., et al., (2016): Sintering and dielectric properties of a technical porcelain prepared from economical natural raw materials, *Cerâmica*, 2016. p. 405-412. <https://doi.org/10.1590/0366-69132016623641994>
- [4] Olupot, P. W. – Jonsson, S. – Byaruhanga, J. K. (2013): Effects of the Sintering Process on Properties of Triaxial Electrical Porcelain from Ugandan Ceramic Minerals. *World Academy of Science, Engineering and Technology, International Journal of Chemical, Molecular, Nuclear, Materials and Metallurgical Engineering*, 2013. 7(5): p. 267-273. <http://scholar.waset.org/1307-6892/11565>
- [5] Stubna, I. – Sin, P. – Viljus, M. – Trnik, A. (2014): The effect of the firing temperature on the hardness of alumina porcelain. *Materiali in tehnologije*, 2014. 48(3): p. 331-336. <http://mit.imt.si/Revija/izvodi/mit143/stubna.pdf>
- [6] Njoya, D. – Tadjuidje, F. S. – Ndzana, E. J. A. – Pountouonchi, A. – Tessier-Doyen, N. – Lecomte-Nana, G. (2017): Effect of flux content and heating rate on the microstructure and technological properties of Mayouom (Western-Cameroon) kaolinite clay-based ceramics. *Journal of Asian Ceramic Societies*, 2017. 5(4): p. 422-426. <https://doi.org/10.1016/j.jascer.2017.09.004>
- [7] Jang-Hoon Ha – Jongman Lee – In-Hyuck Song – Sang-Hyup Lee (2015): The effects of diatomite addition on the pore characteristics of a pyrophyllite support layer. *Ceramics International*, 2015. 41(8): p. 9542-9548.
- [8] Olupot, P. W. – Jonsson, S. – Byaruhanga, J. K. (2008): Effects of mixing proportions and firing temperature on properties of electric porcelain from Ugandan minerals. *Industrial Ceramics*, 2008. 28(1): p. 1-10.
- [9] Iqbal, Y. – Lee, W. E. (2000): Microstructural evolution in triaxial porcelain. *Journal of the American Ceramic Society*, 2000. 83(12): p. 3121-3127. <https://doi.org/10.1111/j.1151-2916.2000.tb01692.x>
- [10] Lee, W. – Iqbal, Y. (2001): Influence of mixing on mullite formation in porcelain. *Journal of the European Ceramic Society*, 2001. 21(14): p. 2583-2586. [https://doi.org/10.1016/S0955-2219\(01\)00274-6](https://doi.org/10.1016/S0955-2219(01)00274-6)
- [11] Chaudhuri, S. – Sarkar, P. – Chakraborty, A. (1999): The Electrical resistivity of porcelain in relation to the constitution. *Ceramics International*, 1999. 25(1): p. 91-99. [https://doi.org/10.1016/S0272-8842\(98\)00006-6](https://doi.org/10.1016/S0272-8842(98)00006-6)
- [12] Acchar, W. – Duttra, E. J. (2015): Ceramic Materials from Coffee Bagasse Ash Waste. 2015: *Springer*.
- [13] Kitouni, S. – Harabi, A. (2011): Sintering and mechanical properties of porcelains prepared from Algerian raw materials. *Cerâmica*, 2011. 57(344): p. 453-460. <https://doi.org/10.1590/S0366-69132011000400013>
- [14] Chaudhuri, S., (1974): Influence of mineralizers on constitution of hard porcelain. 1. Mineralogical compositions. *American Ceramic Society Bulletin*, 1974. 53(2): p. 169-171.
- [15] Das, S. K. – Dana, K. (2003): Differences in densification behavior of K-and Na-feldspar-containing porcelain bodies. *Thermochimica Acta*, 2003. 406(1): p. 199-206. [https://doi.org/10.1016/S0040-6031\(03\)00257-0](https://doi.org/10.1016/S0040-6031(03)00257-0)
- [16] Hamisi, H., et al., (2014): Influence of firing temperature on physical properties of same clay and Pugu kaolin for ceramic tiles application. *International Journal of Materials Science and Applications*, 2014. 3(5): p. 143-146. <https://doi.org/10.11648/j.ijmsa.20140305.12>
- [17] Islam, R. A. – Chan, Y. – Islam, M. F. (2004): Structure-property relationship in high-tension ceramic insulator fired at high temperature. *Materials Science and Engineering: B*, 2004. 106(2): p. 132-140. <https://doi.org/10.1016/j.mseb.2003.09.005>

Ref.:

Ngayakamo, Blasius – Park, S.Eugene: *Effect of firing temperature on triaxial electrical porcelain properties made from Tanzania locally sourced ceramic raw materials*
Építőanyag – Journal of Silicate Based and Composite Materials, Vol. 70, No. 4 (2018), 106–109. p.
<https://doi.org/10.14382/epitoanyag-jsbcm.2018.19>