Effect of cullet on firing temperature and dielectric properties of porcelain insulator

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ABSTRACT

Porcelain insulators prepared from locally available ceramic materials, such as Hosaena clay, quartz, partially replaced feldspar by cullet (rich in alkaline materials). The physical and electrical properties, such as water absorption, apparent porosity, bulk density, linear shrinkage, and dielectric strength as a function of firing temperature, were evaluated. The XRD and AAS measurement results show Hosaenna clay mainly belongs to kaolinite minerals with a sufficient SiO2 (45.60wt. %), Al2O3 (35.52wt. %), and it possesses reasonable plasticity nature. The SEM and Electrical properties measurement shows the porcelain insulators fired at 1150°C & 1200°C showed homogenized primary and secondary mullite with optimum glassy phase and good dielectric strength for practical applications. The primary & secondary mullite and glassy phases were grown early at a firing temperature of 1150 and 1200°C, due to the partial replacement of feldspar by cullet. The porcelain insulator prepared from the composition of 45%clay, 35%feldspar, 10%quartz and 10%cullet and fired at 1200°C showed dielectric strength 8.9kV/mm, water absorption (0.459%), apparent porosity (1.530%), bulk density (4.29 g/cm³), and linear shrinkage (4.18%). Therefore, incorporating economic alkaline materials such as cullet to improve the alkalinity of the locally available clay materials, Hosaenna clay, Arero feldspar, and Arero quartz reduces the firing temperature with excellent dielectric strength.

1. Introduction

In the electric power industry, porcelain insulator plays a vital role during electric power transmission. The porcelain insulators have been using to fasten the electric cable with a sporting pole or tower and resist the flow of electric current from electric wire to the supported metal that has connected to a grounded pole or tower (Meng et al., 2014; Moyo and Park, 2014). The mechanical and electrical insulation performances of the porcelain insulator highly depend on the entire microstructure of the porcelain (Martín-Márquez et al., 2009).

The microstructure of porcelain insulators possess micro-phases, such as crystalline mullite, quartz phase, and an amorphous glassy phase, which grow during sintering (Lee and Iqbal, 2001). Unlike quartz phase, mullite and glassy phases are newly originated phases from the relict of clay and feldspar raw materials at elevated firing temperature (>980°C) (Lee et al., 2008), and their intensity and structure highly depend on firing conditions (firing temperature and dwelling time) as well as quality and quantity of raw materials applied for production (Meng et al., 2016).

Clay, feldspar, and quartz are the raw materials used to produce porcelain insulators (Meng et al., 2012; Olupot et al., 2010; Singh et al., 2017). However, the quality of each material used for porcelain production significantly influences the ultimate properties of porcelain material and its production conditions. For instance, a fluxing material (feldspar) should contain the required alkali oxides level (Na2O + K2O > 12 wt.%) (Moyo and Park, 2014), which are responsible for glassy phase
formation during firing. If it below the required level, it needs a higher firing temperature to get the necessary glassy phase (Merga et al., 2019).

In the study reported by Merga et al. (2019), the porcelain insulator produced from local clay in Ethiopia, such as Babawuha clay, Areró feldspar, and Areró quartz, and better dielectric properties achieved from a batch composition of 45% clay, 45% feldspar, and 10% quartz at firing temperature of 1300 °C (Merga et al., 2019). Hence this high firing temperature (1300 °C) is due to the low quantity of flux material Areró feldspar (Na2O + K2O < 7 wt. %). However, such a high firing temperature is not feasible for mass production at the industrial level. Therefore, this work reports the reduction of the firing temperature without tradeoff the dielectric properties of porcelain insulators using flux-forming oxides such as Na2O, CaO, and SiO2 oxides (Binhussain et al., 2014; Colombo et al., 2003). The flux forming oxides are obtained from waste soda-lime glass.

2. Materials and methods

2.1. Materials

The raw materials used in this study were Hosaena clay, Areró feldspar, Areró Quartz, and cullet (waste-derived glass). The clay used in this study were obtained; Hosaena clay from the Hadiya zone of the southern region (7°33′N and 37°51′E) whereas Areró (feldspar and quartz) from Borena zone of Oromia region (4°45′N and 38°49′E), Ethiopia.

2.2. Materials characterization techniques

The thermal property of the clay sample was analyzed using a simultaneous thermal analyzer (Differential Thermo-Gravimetric Analyzer (DTG)) (model Shimadzu DTG-60H). The DTG/DTA measurement of the clay samples was recorded by heating from room temperature to 1100 °C, at the 10 °C/min heating rate. The plasticity nature of the clay samples was measured using the Casagrande method (L.C.P.C., 1987). The composition of the clay and cullet samples determined by using Atomic Absorption Spectrometry (AAS) (model, spectra AA-20 plus), and loss of ignition (LOI) determined according to Eq. (1).

\[
LOI = \frac{(W_d - W_c)}{(W_d)} \times 100\quad \text{(1)}
\]

where, \(W_d\) = weight of dry sample at 100 °C and \(W_c\) = weight of calcined sample at 1000 °C for 2 h.

X-ray diffraction pattern of clay materials was measured using an x-ray powder diffractometer (XRD), Shimadzu 7000, coupled with a Ni filtered CuKα1 radiation (\(\lambda = 1.5418\)Å) generated by the accelerated voltage of 40 kV and filament current 30 mA. The samples were scanned at Bragg angle range 5–80° in step size of 0.02°. The obtained diffraction data compared with the ICDD PDF-2(International Center for Diffraction Data Powder Diffraction File -2) database and analyzed.

2.3. Porcelain insulator test sample preparation

The green bodies preparation procedure is shown in the schematics as shown in Figure 1; all raw materials were dried, pulverized, and passed through 63 μm Si sieve separately, then mixed according to the desired formulation and dry milled 12hr for uniform homogenization. Consequently, the green body pellets diameter and thickness of 20 mm and 3 mm, respectively, were prepared. Finally, the pellets were fired by a high temperature-controlled furnace at 1100, 1150, 1200, and 1250 °C with a heating rate and soaking time of 6°C/min and 2hr, respectively. The prepared pellets are composed of constant wt. % of clay and quartz, with progressive addition of cullet and Feldspar. The resulting green body is designated as Batch-1, Batch-2, Batch-3 and Batch-4 based on the wt. % of cullet and Feldspar as shown in Table 1.

2.4. Characterization of test samples

The physical properties such as water absorption, apparent porosity, and bulk density of the fired pellets were analyzed using ASTM C338-96, 2010. The dielectric strength of the samples fired at different temperatures was estimated from the breakdown voltage measured by a high voltage testing machine model TERCO HV 1103. The topography and microstructure of the fired pellets scanned by SEM, COXIE3-30, Shimadzu, South Korea. Moreover, the samples were etched with 30% of HF for 30 s and consequently washed repeatedly, dried, and gold-coated before the topography and microstructural image have taken.

Table 1. Prepared batch compositions.

| Materials | Wt.% |
|-----------|------|
|          | Batch – 1 | Batch – 2 | Batch – 3 | Batch – 4 |
| Clay      | 45     | 45    | 45    | 45   |
| Feldspar  | 42.5   | 40    | 37.5  | 35   |
| Cullet    | 2.5    | 5     | 7.5   | 10   |
| Quartz    | 10     | 10    | 10    | 10   |

- Figure 1. Schematics that shows the preparation of cylindrical shape green body porcelain insulator.
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Moreover, the mullite ceramic phases are stable phase (see the chemical reaction shown below) (Garcia-Valles et al., 2010). The phase transformation of the clay results in the release of amorphous metakaolinite into the crystalline mullite (Krupa and Malinari, 2011). The phase transformation from crystalline clay to mullite and amorphous silica (2) (Hammami-Ben Zaied et al., 2015). The 10.89 % mass loss observed by the TGA analysis in Figure 2 corresponds to the loss of water molecules. Besides, the second exothermic DTA curve that appears at 1002.81 °C indicated the recrystallization of amorphous metakaolinite into the crystalline mullite phase (see the chemical reaction shown below) (Garcia-Valles et al., 2010). Moreover, the mullite ceramic phases are stable firing beyond 1500 °C though the density and size of the porosity in the green body is affected (Gong et al., 2014).

3. Result and discussion

3.1. Raw material property analysis

The DTA/TGA data of the clay samples are measured at nitrogen atmosphere and shown in Figure 2. The first intense endothermic peak around 514 °C due to phase transformation from crystalline kaolinite to amorphous metakaolinite phase (Krupa and Malinaric, 2011; Wang et al., 2017). Therefore, the Hosaenna clay exhibits the required plastic character for industrial-scale application.

Table 2. XRD quantitative phase analysis of Hosanna clay.

| Mineral name | Formula                  | PDF #    | Quant (%) |
|--------------|--------------------------|----------|-----------|
| Kaolinite    | Al2(Si2O5)(OH)4          | 00-089-6538 | 92.52     |
| Quartz       | SiO2                     | 00-005-0490 | 5.29      |
| Hematite     | Fe2O3                    | 00-013-0534 | 2.19      |

The XRD diffraction pattern of the Hosaenna clay were measured to identify the types of clay minerals such as kaolinite, illite, smectite, and non-clay minerals present in the sample, the clay scanned at the bulk stage after clay fractionated; and heat-treated at 550 °C (Nzeukou Nzeugang et al., 2018; Senoussi et al., 2016) as shown in Figure 3 and Table 2. The observed crystalline peaks on the bulk and fractionated states of the clay belong to kaolinite clay, quartz, and hematite, as shown in Figure 3a-b. The phase composition of kaolinite clay, quartz, and hematite, 92.52%, 5.29%, and 2.19%, respectively, is presented in Table 2.

Furthermore, the presence of kaolinite clay mineral in the clay sample was confirmed by a spectrum scanned after heated at 550 °C, as shown in Figure 3c. The crystalline kaolinite clay has expected to convert into a metakaolinite amorphous phase at the temperature range of 450–680 °C (Boussen et al., 2016). Hence the amorphous metakaolinite phase formed due to the lack of long-range order and confirmed by a broad humble XRD peak as shown in Figure 3c.

The plastic behavior of clay is an important parameter to be controlled for the production of porcelain insulators, therefore, the Casagrande plasticity test of Hosaenna clay liquid limit, plastic limit, and plastic index, are shown in Table 3. The average plasticity index (PI) found in the range of 14.77 ± 0.64, average plastic limit (PL) 24.57 ± 0.12, and average liquid limit (LL) 39.33 ± 0.58, which are in moderate plasticity according to Holtz and Kovacs diagram (Mahmoudi et al., 2017). Therefore, the Hosaenna clay exhibits the required plastic character for industrial-scale application.

Table 3. Casagrande plasticity test; liquid limit, plastic limit and plastic index.

| Trials   | Trial -1 | Trial -2 | Trial -3 | Average |
|----------|----------|----------|----------|---------|
| Liquid limit (LL) | 39       | 40       | 39       | 39.33 ± 0.58 |
| Plastic limit (PL) | 24.5    | 24.5    | 24.7    | 24.57 ± 0.12  |
| Plastic index (PI) | 14.5    | 15.5    | 14.3    | 14.77 ± 0.64  |

The physical properties such as apparent porosity, water absorbance, bulk density, and linear shrinkage of the porcelain insulator prepared from Hosaenna clay shown in Figure 4. As the firing temperature increase,
the porosity and water absorbance of all batches of the porcelain decrease and reached a minimum value at a firing temperature of 1200 °C; however, the porosity and water absorbance increased while fired above the 1200 °C temperature as shown in Figure 4a, b. The bulk density and linear shrinkage increase as the firing temperature increases to 1200 °C and decreases while fired above this temperature, as shown in Figure 4c, d.

The formation of the liquid phase/glassy phase starts at a firing temperature of 1100 °C. The glassy phase mainly originated from fluxing materials such as feldspar and cullet/waste glass. The firing temperature increases the feldspar and cullet/waste glass melt and fills up open pores/void space found in the microstructure; and new crystalline mullite (primary mullite) grow from clay relict (Belhouchet et al., 2018; Meng et al., 2014, 2016). Therefore, the pore volume decreases and continues up to complete filling of the voids present at 1200 °C. Hence, water absorption, apparent porosity got a minimum value, and density and linear shrinkage reached a maximum. However, raising the firing temperature to 1250 °C, the formation of the liquid phase highly increased. The liquid phase intensity relates with the amount of flux material present in batches (batch-1 < batch -2 < batch -3 < batch -4) (Table -1). The increase of the low viscose liquid phase in the microstructure of ceramics promotes and facilitates the release of trapped gases (Belhouchet et al., 2018). Therefore, the release of trapped gases leads to the formation of a new open-pore to the microstructure result in density reduction; consequently, the porosity and water absorption increased at 1250 °C (Fig. 4a, b, and c).

The dielectric strength (ratio of break down voltage to sample thickness) of all batch samples increases with firing temperature up to 1200 °C and decrease at the temperature of 1250 °C, as shown in Figure 5. A gradual increase of dielectric strength is associated with the densification and mullite formation in insulator bodies. The glassy phase formation in the porcelain originated from flux materials such as feldspar and cullet, and the crystalline mullite phase grows from clay relict, which possesses high electric resistance (Islam et al., 2004). Above the 1200 °C of firing temperature, the dielectric strength of all batches except batch 1 decreases and is associated with the intensity of the glassy phase present.
in the body and crystal defect formed. At the elevated temperature, a 1250 °C excess amount of low viscous glassy phase has formed and results in microstructure and crystalline phases change. Therefore it facilitates easy mobility of ions (Na\(^+\) and K\(^+\)), which leads to decreasing dielectric strength at 1250 °C (Figure 5) (Kasrani et al., 2016). Besides, high concentrations of glassy phase in structure promote the releasing of trapped gases, consequently yield new pore/void space in the porcelain that enhances conductivity. But Batch-1 has a progressive increase of dielectric strength in successive firing temperatures because this batch relatively contains low flux materials as shown in table -1.

Moreover, all batches, B-1 – B-4, showed relatively good dielectric strength for a low voltage transmission line that is in the range of above 6 kV/mm –8.9 kV/mm (Ologunwa, 2020; Olupot et al., 2010) at the firing temperature between 1150 °C–1200 °C. The firing temperature is reduced by 150 °C without compromising the dielectric strength of the porcelain insulator comparing the previous study (Merga et al., 2019). The reduction of firing temperature is due to the partial substitution of low alkaline feldspar by cullet/waste glass. The added cullet substituting the alkaline feldspar facilitates the early formation of the glassy phase responsible for the densification and secondary mullite phase formation.

### 3.3. Crystallographic phase and microstructure analysis

The microstructure and crystallographic phase of the batch-4 sample is shown in Figure 6. Batch-4 shows higher dielectric strength and good physical properties than other batches. Thus batch-4 is selected for microstructure and crystallographic phase analysis. The XRD shows that at early firing temperature, a characteristic peak of mullite, quartz, and feldspar peaks have appeared. Moreover, the mullite phases increase with increasing firing temperature and resulting in quartz and feldspar phases decrease. At a higher temperature, the feldspar peak disappeared, as shown in Figure 6 (a).

The feldspar vanished at the higher temperature due to the complete melting of the feldspar to form a glassy phase, which is confirmed by the formation of humble reflection (amorphous phase) in the XRD, Figure 6 (a) (iv). The SEM image shows that the primary mullite, secondary mullite, and glassy phases at firing temperatures of 1200 °C and 1250 °C, Figure 6(b) –(c).

The crystalline mullite phases may originate from clay relict (primary mullite), and mullite which grows from feldspar relict (secondary mullite). The secondary mullite has an interlocked needle-like structure (Lee and Iqbal, 2001; Martín-Marquez et al., 2010; Meng et al., 2012) that appeared with optimum glassy phase in a homogenized. However, the content of glassy phase increase and adhere on the surface at elevated temperature 1250 °C, as shown in Figure 6(c). The microstructure features agree and confirm the justification asserted for the physical and electrical properties of insulator samples.

### 4. Conclusion

Electrical porcelain insulators produced from locally available Clay materials, i.e., Hosaena clay, Arero feldspar, and Arero quartz cullet...
(waste glass) used to improve the alkaline level of flexing material (feldspar). The raw material characterization result implies that the clay material (Hosaena clay) occupies 45% up to 50% kaolinite type clay minerals with the required level of oxides SiO₂ (48.68wt.%) and Al₂O₃ (33.22wt.%). Moreover, the Hosaenna clay contains a small amount of color-producing and phase-changing transition metal oxides, i.e., TiO₂ (0.27 wt. %) and Fe₂O₃ (1.88 wt.%). Therefore, Hosaenna clay is suitable in ceramic and clay-based industries to produce whitewares and porcelains industries. The prepared insulator samples have a body composition of 45%clay, 10% quartz, (35–45%) feldspar, and (2.5–10%) cullet fired at 1150 °C and 1200 °C satisfy the required technical and electrical properties for insulation application. However, among the prepared batch, batch -4 (45%clay, 35%feldspar, 10% quartz, and 10% cullet) possess excellent technical properties such as water absorption (0.459%), apparent porosity (1.530%), bulk density (4.29 g/cm³), linear shrinkage (4.18%) and electrical property of dielectric strength 8.9kv/mm at firing temperature of 1200 °C. Therefore adjusting the alkaline level of clay, which has low alkaline behavior, with economic waste-derived flex material (cullet) can facilitate the formation of mullite and glassy phases at a relatively low sintering/firing temperature of 1150 °C.

Declarations

Author contribution statement

Andualem Merga, Dinsefa Mensur Adoshe: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.
Tatek Temesgen Terfasa, Adane Muche Abebe: Performed the experiments; Analyzed and interpreted the data; Wrote the paper.
Enyew Amare Zefre, Eaba Beyene: Performed the experiments.
Miresa Tadese: Performed the experiments; Contributed reagents, materials, analysis tools or data.

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Data availability statement

Data will be made available on request.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

References

Belhouchet, K., Bayadi, A., Bellhouchet, H., Romero, M., 2018. Improvement of Mechanical and Dielectric Properties of Porcelain Insulators Using Economic, 8, pp. 28–37.

Binhussain, M.A., Marangoni, M., Bernardo, E., Colombo, P., 2014. Sintered and glazed glass-ceramics from natural and waste raw materials. Ceram. Int. 40 (2), 3543–3551.
Bouchen, S., Sghaijer, D., Chabani, F., Jamoussi, B., Bennouar, A., 2016. Characteristics and industrial application of the lower Cretaceous clay deposits (Bouhedma Formation), Southeast Tunisia: potential use for the manufacturing of ceramic tiles and bricks. Appl. Clay Sci. 123, 210–221.
Colombo, P., Brusatin, G., Bernardo, E., Scartinic, G., 2003. Inertization and reuse of waste materials by vitrification and fabrication of glass-based products. Curr. Opin. Solid State Mater. Sci. 7 (3), 225–239.
García-Valles, M., Alfonso, F., Martínez, S., Roca, N., 2020. Mineralogical and thermal characterization of kaolinitic clays from terra alta (Catalonia, Spain). Minerals 10 (2).
Gong, Lunhun, Wang, Yonghong, Cheng, Xudong, Zhang, Ruifang, Zhang, Heiping, 2014. Porous mullite ceramics with low thermal conductivity prepared by foaming and starch consolidation. J. Porous Mater. 21, 15–21.
Hammani-Ben Zied, A., Abidi, R., Slim-Shimi, N., Somarain, A.K., 2015. Potentiality of clay raw materials from Gram area (Northern Tunisia) in the ceramic industry. Appl. Clay Sci. 112 (113), 1–9.
Islam, R.A., Chao, Y.C., Islam, M.F., 2004. Structure-property relationship in high-tension ceramic insulator fired at high temperature. Mater. Sci. Eng. B: Solid State Mater. Adv. Technol. 106 (2), 132–140.
Kasrani, S., Harabi, A., Barana, S.E., Foughali, L., Benhassine, M.T., Alhdayan, D.M., 2016. Sintering and dielectric properties of a technical porcelain prepared from economical natural raw materials. Ceramica 62 (364), 405–412.
Krupa, P., Malinari, S., 2015. Thermal properties of green alumina porcelain. Ceram. Int. 41 (2), 3254–3258.
Lee, W.E., Iqbal, Y., 2001. Influence of mixing on mullite formation in porcelain. J. Eur. Ceram. Soc. 21 (4), 2583–2586.
Lee, W.E., Souza, G.P., McConville, C.J., Tarvormapanich, T., Iqbal, Y., 2008. Mullite formation in clays and clay-derived vitreous ceramics. J. Eur. Ceram. Soc. 28 (2), 465–471.
Mahmoudi, S., Bennouar, A., Serna, E., Zargouni, F., 2017. Characterization, firing behavior and ceramic application of clays from the Gabes region in South Tunisia. Appl. Clay Sci. 135, 215–225.
Martín-Márquez, J., De La Torre, A.G., Aranda, M.A.G., Rincón, J.M., Romero, M., 2009. Evolution with temperature of crystalline and amorphous phases in porcelain stoneware. J. Am. Ceram. Soc. 92 (1), 229–234.
Martín-Márquez, J., Rincón, J.M., Romero, M., 2010. Mullite development on firing in porcelain stoneware bodies. J. Eur. Ceram. Soc. 30 (7), 1599–1607.
Meng, Y., Gong, G., Wu, Z., Yin, Z., Xie, Y., Liu, S., 2012. Fabrication and microstructure investigation of ultra-high-strength porcelain insulator. J. Eur. Ceram. Soc. 32 (12), 3043–3049.
Meng, Y., Gong, G., Wei, D., Xie, Y., Yin, Z., 2014. Comparative microstructure study of high strength alumina and bauxite insulator. Ceram. Int. 40 (7), 10677–10684.
Meng, Y., Gong, G., Wei, D., Xie, Y., 2016. In situ high temperature X-ray diffraction study on high strength alumina porcelain insulator with the AI2O3-SiO2-K2O-Na2O system. Appl. Clay Sci. 132–133, 760–767.
Merga, A., Murthy, H.C.A., Amare, E., Ahmed, K., Bekele, E., 2019. Fabrication of electrical porcelain insulator from ceramic raw materials of Oromia region, Ethiopia. Heliyon 5 (8), e02327.
Moyo, M.G., Park, E., 2014. Ceramic raw materials in Tanzania - structure and properties for electrical insulation application. Int. J. Eng. Res. Technol. 3 (10), 1015–1020.
Nzuokwu Nzeugang, A., Ouahabi, M. El, Aziwo, B., Mache, J.R., Mefire Mounton, H.S., Fagel, N., 2018. Characterization of kaolin from Mankon, northwest Cameroon. Clay Miner. 53 (4), 563–577.
Olugunusa, T.P., 2020. Process and development of porcelain electrical insulator using Edo state, Nigerian raw materials. Int. J. Eng. Manuf. 10 (3), 43–55.
Olupot, P.W., Jonsson, S., Byaruhanga, J.K., 2010. Development and characterisation of triaxial electrical porcelain from Ugandan ceramic minerals. Ceram. Int. 36 (4), 1455–1461.
Senoussi, H., Osmali, H., Courtois, C., Bouraib, M.E.H., 2016. Mineralogical and chemical characterization of D33 kaolin from the east of Algeria. Bol. Soc. Española Ceram. Vitr. 55 (3), 121–126.
Singh, N., Kumar, P., Tripathi, P., Pyare, R., Majhi, M.R., 2017. Influence of Alumina and Silica Addition on the Physico-Mechanical and Dielectric Behavior of Ceramic Porcelain Insulator at High Sintering Temperature, 7, pp. 151–159.
Wang, H., Li, C., Peng, Z., Zhang, S., 2011. Characterization and thermal behavior of kaolin. J. Therm. Anal. Calorim. 105 (1), 157–160.