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THE PROPERTIES OF FELDSPATHIC DENTAL PORCELAIN FROM TANZANIAN ALUMINOSILICATE MATERIALS

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ABSTRACT

Kaolin and quartz from Pugu and feldspar from Morogoro were used to produce feldspathic dental porcelain bodies. Mineralogical results from XRD revealed the presence of kaolinite, illite, microcline and clinochlore in the kaolin samples; albite, microcline, sanidine in the feldspar samples and only silica in the quartz samples. The chemical results studies by XRF revealed the presence of BaO, SiO_2, Al_2O_3, TiO_2, Cr_2O_3, MnO, K_2O, CaO, Cl, SO_3, MgO, Na_2O, and Fe_2O_3 in varying proportions. Particle size distribution of all the raw materials were determined and particles <63 µm were used for preparation of batches. The batches of different compositions with ranges: quartz (12-22%), feldspar (75-88%) and kaolin (3-5%) were fired at 1100, 1200 and 1250 °C, and the developed properties were tested. The physico-mechanical properties studied exhibited volumetric shrinkage between 2 and 55.17%, open porosity between 6.67 and 40.00%, water of absorption between 7.1 and 40%, bulk density of 1.43 – 5.68 g/cm^3, and compressive strength at 0.13 - 32.25 MPa. The optimal firing temperature for the best results was found to be 1250 °C. The results obtained indicate that kaolin and quartz from Pugu and feldspar from Morogoro are potential raw materials for the production of dental porcelain.

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INTRODUCTION

Dental ceramics are the preferred materials for oral restorations capitalising on its favourable characteristics, such as; adequate aesthetics, high fracture strength and chemical stability (Rosenblum and Schulman 1997). Dental ceramics may also be used to create crowns, veneers, inlays, onlays and denture teeth. A desirable feature of ceramics is that their appearance can be customised to simulate the colour, translucency and fluorescence of natural teeth. They are biologically and chemically inert, inherently brittle with a good resistance to abrasion. When ceramic powder is comprised of quartz, kaolin, pigments, opacifiers, and a suitable flux with distilled water, the product is traditionally known as dental porcelain. A more acceptable terminology is dental ceramics, however, these two terms are always used interchangeably (Jablonski 2001). The classification of dental porcelain based on composition include, feldspathic porcelains, leucite reinforced feldspathic porcelain, aluminous ceramics, glass infiltrated composites, alumina polycrystals, glass ceramics, leucite reinforced glass ceramics, lithium disilicate reinforced glass ceramics and zirconium oxide ceramics (Martinez 2007). The classification can also be based on the residence temperature during the firing circle, resulting into three categories namely high (1300 – 1400 °C), medium (1100 – 1300 °C), and low (850 – 1100 °C) fusing dental porcelains (Moffa 1988, Carrado 2001) High fusing dental porcelains are used for denture teeth, medium for pontics, whereas the low fusing porcelain are used for metal-ceramic restorations. The composition of typical feldspathic dental porcelain by weight is; feldspar (75-85%), quartz (12-22%), kaolin (3-5%) and metallic pigments which increase opacity of dental porcelain (<1 %) (Craig 1993). Scientific investigations have been carried out on the applications of Tanzanian kaolin, quartz and feldspar in refractories and other industrial uses (Lynne et al.1980, Lobitzer *et al.*, 1982, Mwakarukwa 1988, Akwilapo 1999, Akwilapo and Wiik 2003, Lugwisha 2006 a, b) but up to this moment there are no literature explaining their applications in dental industry. This research therefore explored the suitability of some selected Tanzanian non-metallic silicate minerals in the dental application. The study in particular dealt with feldspathic porcelains in low and medium firing temperatures utilizing kaolin, quartz and

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feldspar. The properties of dental ceramics which were determined in this study included chemical composition, mineralogy, density, porosity, volumetric shrinkage and compressive strength. Testing results from each property were used to assess the suitability of the fired samples.

MATERIALS AND METHODS

Raw materials

The raw materials were kaolin clays and quartz from Pugu deposits and feldspar from Morogoro Tanzania. The chemical composition of the raw materials was determined by XRF using the Semi Quantitative XRF Analysis technique (Siemens SRS 3000 X-ray Fluorescence (Rhodium anode, 8 analyzer crystals with beryllium windows 125 μm). The semi quant program was used throughout. The mineral characterization of the raw materials were performed on Siemens D-5005 X-ray Diffractometer using Cu Kα1 radiation (λ = 1.54056 Å). The samples underwent diffraction in the measuring range 2θ ranging between $2^o - 65^o$.

Particle size distribution of the raw materials and Batch composition

The grain size distribution was determined from weight distribution in screen residues combined with sub-sieve analysis carried out using Andreasen pipette sedimentation method. In kaolin, the non-clay fraction was 37.57% while the clay fraction in the range 0 – 63 μm was 62.43%. In feldspar and quartz, grain particle size in the range > 63 μm were 24.82, 22.16 and in the range between 0 – 63 μm were 75.18, 77.84, respectively. The 0-63 μm particles size were selected for all raw materials for batch preparation in order to improve the physico-mechanical properties of the test specimens (Ili'na 2007). The use of raw materials of heterogeneous composition such as 0-63 μm ensures reduced void spaces as a result of an increase in the inter-particle contacts of small particles (Fraunhofer 2009).

Sample preparation

Kaolin purification was done by means of wet beneficiation followed by homogenization to minimize errors attributable to sample heterogeneity. Then clay fraction separation (by means of deflocculating agent and peristaltic pumps) was done to obtain fractions needed for XRF and XRD Analysis. Batch compositions typical of the ranges used in dental porcelain were chosen. Thirteen different body compositions were prepared, these are indicated in Table 1.

Table 1. Batch compositions (mass %) of the raw mixes used in dental porcelain

Batch → / Componcnt ↓ (mass %)	S_1	S_2	S3	S_4	S_5	S_6	S_7	S_8	S_9	S_{10}	S_{11}	S_{12}	S_{13}
Feldspar	75	76	77	78	79	87	81	82	83	84	85	80	77
Quartz	20	19	19	19	19	12	18	15	14	14	13	19	22
Kaolin	5	5	4	3	2	1	1	3	3	2	2	1	1

The body formulations were mixed with a given volume of water so that the resulting slip contained 35% water. Sodium silicate (3%) and sodium hexametaphosphate were added in order to act as deflocculant. The mixtures were ball milled and left to equilibrate for 36 hours to allow particles in the slips to undergo sorption reactions with electrolytes and surfactants in the liquid while altering their viscosity and casting characteristics. The slips were then casted into truncated cones in plaster of Paris moulds.

Drying and Firing

The cones were dried in an open air for seven days and then in an oven at 110 °C for 48 hours. The prolonged drying was considered suitable for the purpose of avoiding surface cracking which would have occurred during firing due to excessive surface drying. Advanced firing was done at 1100, 1200 and 1250 °C, at a holding time of 8 hours at a heating rate of 300 °C per hour. The fired samples were then allowed to cool slowly at room temperature by natural convention for 24 hours.

Volumetric shrinkage

The volume of specimens with truncated cone shapes were determined by the formula $V= (\pi/3)h(Rr + R^2 + r^2)$ before and after firing. Where R and r are the radii of (base circle and top circle, respectively) the ends of the cone; h is the height of the cone. R, r and h were measured by using veneer caliper. The difference in volume before and after firing was used to calculate % shrinkage by using the formula $(\Delta V/V_1) \times 100$; where ΔV is the volume difference and V_1 is the volume before firing.

Open Porosity

Weights of dry specimens were determined by using Sartorius analytical balance. True and bulk volume was determined by using measuring cylinder and the formula $\rho = W/V$ was used to determine the true density and the bulk density; where ρ=density, w= weight and v= volume. Porosity (Q) was then determined by using the formula: $1-\rho_{bulk}/\rho_{particle}$.

Water of absorption

The specimens were soaked in water at room temperature for 48 hours. The mass before and after saturation was determined and thereafter used to calculate % water absorption by using the formula:

% water absorption = $M_a - M_b/M_b \times 100\%$. Where, M_a = mass after saturation (saturated specimen), M_b= mass before saturation (dry specimen).

Bulk Density

The bulk density, B (g/cm^3), of the specimen is the quotient of its dry mass divided by the exterior volume, including pores, is

given by: $B = D/V$. Where D is the mass of dry specimen and V is the exterior volume. The exterior volume, V is given by $V = M - S$, where M is the saturated mass and S is the suspended mass.

Compressive strengths

For load determination, each specimen was compressed by compression tester UTM of type UPD 40 with energy 400 V, 50 Hz, 6A and maximum load 400 KN. Compressive strength was then determined from the formula: C_s = L/A. Where, C_s= compressive strength, L= load in Newton, and A= area in mm^2.

RESULTS AND DISCUSSION

The chemical composition

The chemical composition and the loss on ignition (LOI) are presented in Table 2. The dominant oxides for kaolin (63.64%, 31.43%) and feldspar (62.3%, 14%) were SiO_2 and Al_2O_3, respectively, the typical proportion percentage constituents needed for dental feldspathic glass.

Table 2. Chemical compositions (% mass) of the raw materials

Sample → / Parameter (%) ↓	Kaolin	Feldspar	Quartz
SiO_2	63.64	62.30	94.93
Al_2O_3	31.43	14.00	0.38
Na_2O	0.43	0.81	-
MgO	0.16	0.04	0.04
SO_3	0.25	0.98	1.00
Cl	0.76	0.19	0.19
K_2O	1.39	12.20	0.09
CaO	0.06	7.65	0.98
TiO_2	0.80	0.04	0.11
Cr_2O_3	0.03	0.14	0.21
MnO	0.01	0.04	0.03
Fe_2O_3	1.01	0.85	1.95
BaO	-	0.46	-
LOI	0.03	0.3	0.09
Total	100	100	100

As can be seen, there is a significant amount of alumina in both kaolin and feldspar. Alumina in dental porcelains is known to increase the strength of the dental ceramic materials (Chu *et al.*, 2004) and is useful in maintaining opacity properties (McLean and Hughes 1965; Southan 1968). Alkali oxides whose role is to improve the coefficient of the thermal expansion of the ceramic articles (Southan 1968) were found in significant amount (13%) in feldspar in which only BaO was also detected. The levels of coloured oxides TiO_2 which imparts yellow brown colour, Cr_2O_3 green colour, MnO for lavender pigment and Fe_2O_3, were in acceptable amounts. Fe_2O_3 is useful for colouring dental porcelain (Southan 1968) but when colouring is not needed this oxide becomes undesirable. The quartz used was seen to contain considerable amount of silica needed in the batches.

The Mineralogical Composition

Clay minerals and non clay minerals identified in kaolin and feldspar are given in Figure 1. XRD results revealed that the major content in kaolin samples was kaolinite $[Al_2Si_2O_5(OH)4]$ at d = 7.16, 4.41, and 3.56 Å around 14° and 29° 2θ, respectively, surbordinated with illite $[KAl_2(Si_3AlO_{10})(OH)_2]$ at d = 3.33 Å around 31° 2θ; while the minor minerals observed in kaolin were quartz (SiO_2) at d = 4.25 (24° 2θ) and 3.34 Å (31° 2θ) and microcline $[KAlSi_3O_8]$ at d = 4.22 (25° 2θ). In the feldspar sample, the prominent minerals were Albite $[NaAlSi_3O_8]$ at d = 4.02 (26° 2θ) and 3.19 Å (33° 2θ) and Microcline $[KAlSi_3O_8]$ at d = 4.22 (24° 2θ) and 3.24 Å (32° 2θ). The minor peaks were observed in the mineral sanidine $[K(Si_3Al)O_8]$ identified by the presence of peaks around 27°, 35°, and 54° 2θ and oligoclase $[(Na,Ca)Al(Al,Si)Si_2O_8]$ identified by the presence of peaks around 38°, 42°, 45° and 49° 2θ. In quartz, the mineral silica (SiO_2) was the only peak at d = 4.25 and 3.34 Å.

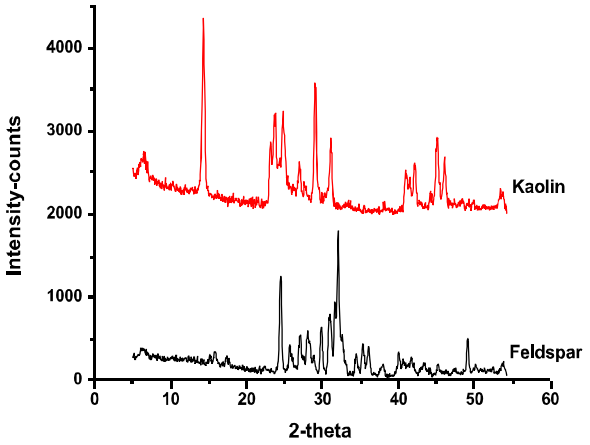


Fig. 1. XRD patterns of (a) Kaolin from Pugu and (b) feldspar from Morogoro

Volumetric Shrinkage

The variation of percentage shrinkage with firing temperatures 1100, 1200 and 1250 °C and composition (Figure 2) shows that shrinkage is a function of batch composition and temperature. The non- uniform shrinkage pattern has been obtained in different composition fired at the same temperature. However, the onset of shrinkage above 1000 °C, which is known to depend on the formation of liquid phase, depends on the composition of the batches (Akwilapo 1999). Bodies with high silica content normally experience a low shrinkage during firing while bodies with high feldspar start to densify at as low as 1000 °C thus cause higher contraction (Kobayashi et al. 1991). For example the highest shrinkage at 1250 °C is recorded for $S6_{(K1\text{-}F87\text{-}Q12)}$ batch. It is reasoned that the low quartz content in the raw porcelain mixes, which decreased even further after firing, caused the higher shrinkage of this sample. Associated with this was the higher content of feldspar which also participates in the melt formation. From Figure 2, it is revealed that firing temperature and holding time affect shrinkage more than does composition of the fired specimen. The effect of composition was seen to be substantial upon dealing with specimen of different composition fired at the same temperature but upon raising firing temperature to about 50 °C, the effect of composition was outweighed by the changes brought in through rise in temperature. It was noticed

that shrinkage increased to about twice when firing temperature rose from 1100 to 1250 °C. The reason behind this is that, the distribution and size of particles involved in solid state reactions significantly affected the firing shrinkage of porcelain materials because at high temperature, fusion of particles and their ability to penetrate inter-particle spaces is maximized (Fraunhofer 2009). There is a decrease in void spaces and an increase in densification and compaction with raise in firing temperature. Results obtained from this study are in line with the results described by Southan (1968).

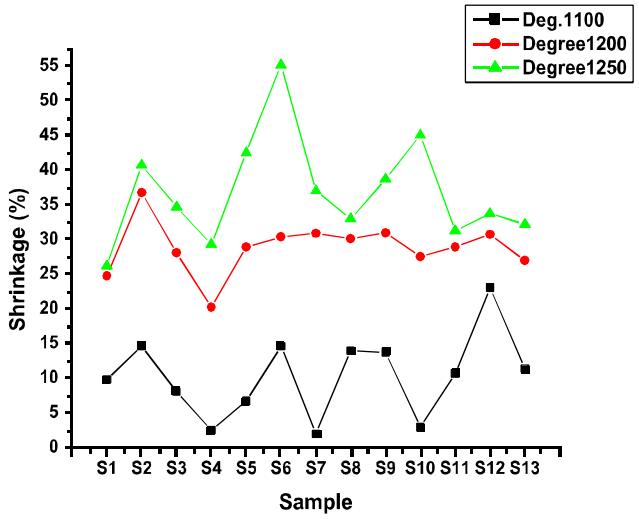


Fig. 2. Variation of volumetric shrinkage with firing temperature of the 13 batches

Porosity

The general trend observed (Figure 3) revealed that there was a decrease in porosity with an increase in firing temperature from 1100 to 1250 °C, hence porosity was seen to be dependent on firing temperature.

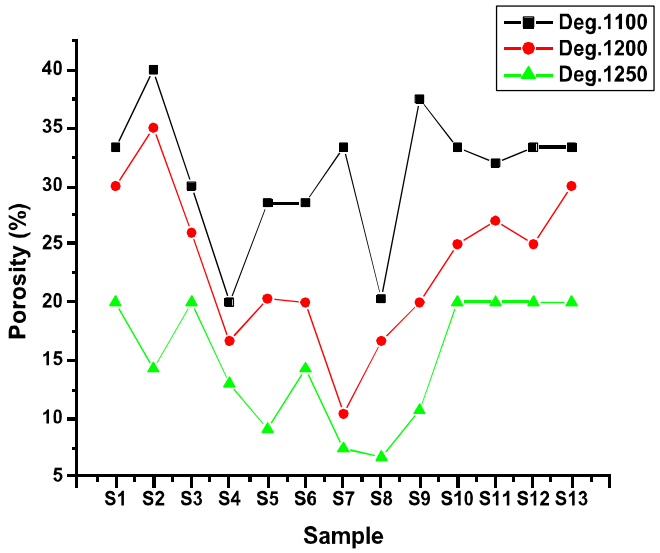


Fig. 3. Variation of porosity with firing temperature of the 13 batches

All compositions prepared in this work exhibited very low porosity at 1250 °C. The decrease of open porosity at higher temperature is associated with the further formation of melt/liquid phase which blocks the open porosity of the body and reduces them on cooling (Akwilapo and Wiik 2003). This observation is reported by Viruthagiri *et al.*, (2009) who also noted that the amount of liquid phase increases with firing temperature. However, in some cases porosity may increase at higher firing temperature (>1200) than expected as observed in batches S1,S3, S10-S13. Such increase in porosity at higher firing temperature is thought to be caused by bloating which takes place as gases are expelled from the matrixes and cause an increase in porosity (Akwilapo 1999, Cheung and Darvell 2002).

Water of Absorption

It is clearly seen (Figure 4) that percentage water of absorption was found to decrease substantially when firing temperature was raised from 1100 to 1250 °C. Water absorption decreases with increase in temperature due to the formation of the liquid phase at high firing temperatures. The liquid phase formed is the one which fills the pores and decreases the porosity. Similar trend was obtained by Khabas *et al.*, (2003), where he found that upon firing dental porcelain specimen at 900 °C, 1000 °C and 1100 °C the percentage water of absorption kept on decreasing. The difference with Khabas *et al.*, (2003) results and our results is that he obtained very low percentage water of absorption of 1.10% 0.97% and 0.78% at 900, 1000, and 1100 °C respectively. His results might have been influenced by the undercoat layer on the samples which acted as a mask. Water absorption is directly related to open porosity According to Khabas *et al.*, (2003) high water of absorption signifies an increased open porosity.

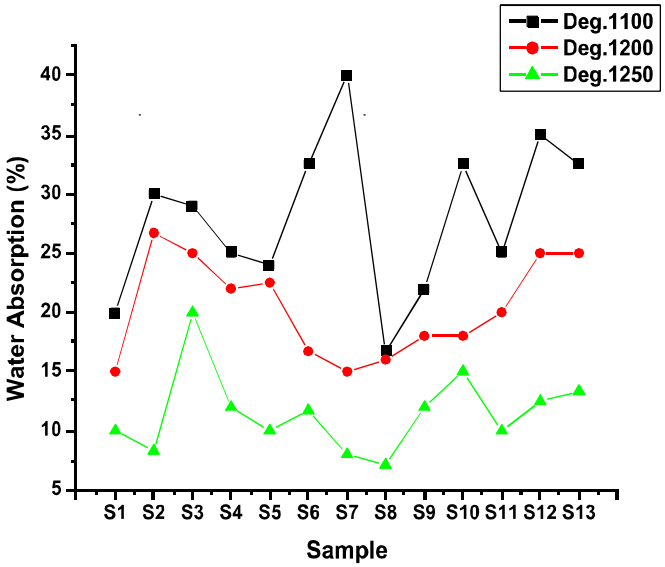


Fig. 4. Variation of water of absorption with firing temperature of the 13 batches

Bulk Density

The variation in bulk density of the tested samples (Figure 5) shows that bulk density increased with increase in firing temperature. This is because at higher temperature samples will form higher amount of melt phase, the more melt is formed the higher will the density of the material be, this is due to the filling of the pores by melt (Akwilapo and Wiik 2003). The bulk density also shows dependence on the amount

of kaolin mixed in the batches. Batches with relatively higher kaolin content e.g. S1 and S2 showed a higher value of bulk density. However, the bulk densities obtained from all compositions and temperatures in this study were found to be low for the production of dental materials with compressive strength comparable to natural teeth strength (Williams 1979).

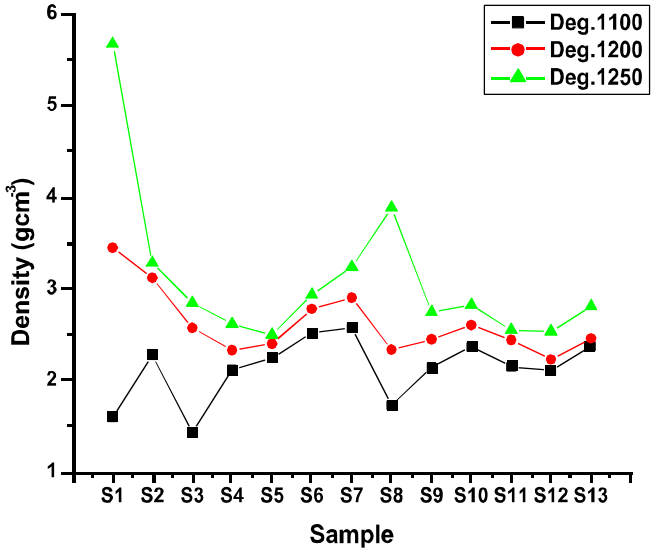


Fig. 5. Variations in bulk density with firing temperature of the 13 batches

Compressive Strength

Compressive strength was observed to increase with increasing firing temperature from 1100 to 1250 °C. Indeed, it was clearly seen that all specimen used showed a decreasing porosity when firing temperature was raised from 1100 to 1250 °C (Figure 3) which is also related to the increase of bulk density. This might be one of the factors leading to the patterns observed in Figure 6.

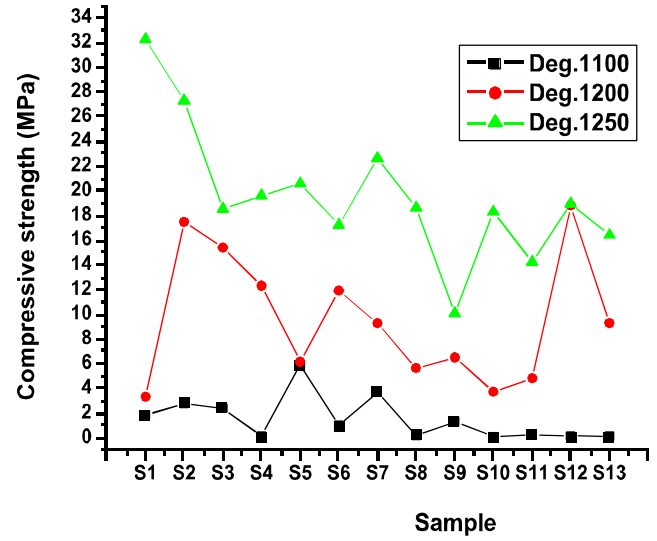


Fig. 6. Variation in compressive strength with firing temperature of the 13 batches

Similar observation was made by Yan et al., (2011) whose results revealed that the decrease in porosity, increase in bulk density and well distribution of pore sizes are factors which can be ascribed for increased compressive strength. At 1250 °C, the highest compressive strength observed was 32.25 MPa for S1 composition. However, the compressive strength results obtained in this study are compared below with standard samples reported in the literature. Khabas *et al.*, (2003) report the compressive strength of four samples as: 31.97 MPa, 39.30 MPa, 39.48 MPa and 48.00 MPa for GFSn (abrupt cooled), GFSn (slow cooled), GF + 10% SnO_2 and natural tooth dentine, respectively. On the other hand, Williams (1979) in his work reported the compressive strength of ten samples on dental stone materials in the range 51.71 – 78.6 MPa.

Conclusion

The present study has revealed the potentiality of Tanzania aluminosilicate minerals: kaolin, feldspar and quartz for use in medical applications. However, there is a need to improve compressive strength and to decrease porosity of the test specimen prepared in this work due to the fact that the prepared dental porcelain were found to have an average low compressive strength and high porosity as compared to that in natural teeth. This might have probably been attributed by the preparation methods and the size of the particles used. More work must be done to investigate other mechanical properties which were not tested in this work due to shortage of funds. The foundation made from this pioneered study will open up the doors for more researches in order to improve the findings obtained by applying the techniques used in this study or more advanced techniques. Thus, further endeavours into these Tanzania aluminosilicate minerals in dental applications will increase their value both for industrial and medicinal applications.

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