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EFFECT OF FELDSPAR AND SILICA VARIATION ON THE PROPERTIES OF DENTAL PORCELAIN

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Abstract
Dental porcelain was produced by mixing feldspar, silica, kaolin and bone ash by varying the contents of feldspar and silica. The processing steps include milling, sieving, pressing/shaping, drying, and sintering while the characterisation techniques were Hardness, Compressive strength, X-ray diffraction, Scanning electron microscopy and Fourier Transform Infrared (FTIR). The mixture was subjected to temperatures of 1100 and 1200 ^oC in a sintering furnace. The chemical composition was determined using X-ray fluorescence and they confirm that SiO_2 and Al_2O_3 are the two major constituents in feldspar and kaolin while CaO is the major constituent in bone ash. For samples sintered at 1200^oC, the X-ray diffraction showed that some glass phase possibly consisting of hedenbergite, ilmenite and silica were formed while crystalline phases namely microcline and sanidine were obtained for samples sintered at 1100°C. The morphology of the grains revealed that samples sintered at 1200^oC had some hexagonal silica crystals while flakes of different sizes were obtained for samples sintered at 1100^oC. Hardness values between 262 and 536 BHN, compressive modulus values ranging from 219 MPa to 324 MPa and linear shrinkage values between 6.34 and 7.6% were obtained. The batches of different compositions with ranges: quartz (silica) (15-25%), feldspar (70-80%), kaolin (Edda/Bauchi) (4%) and bone ash (1%) were fired at 1100, 1200^oC, and the developed properties were tested. The sample with 70 wt.% of feldspar, 25 wt.% silica, 4 wt.% of Bauchi clay, and 1 wt.% bone ash sintered at 1200^oC gave the best properties and has the potential to be used in dental restoration.

Keywords: Dental porcelain, Feldspar, Silica, Bone ash, Kaolin, Characterisation.

1.0 INTRODUCTION

Porcelain is an inorganic ceramic material obtained by sintering Kaolin ($Al_2O_32SiO)_22H_2O$, Silica (SiO_2) and Feldspar ($K_2O.Al_2O_3.6SiO_2$) such that they react with one another at appropriately high temperature [1, 2]. Kaolin serves as the plastic material while silica and feldspar are the non-plastic materials. Feldspar acts as an alkali flux with the objective of lowering the sintering temperature as well as controlling both density and porosity of the ceramics. Silica controls the rate of shrinkage and gives both rigidity and support to the porcelain body [2-4]. Porcelain as a clay body when fired becomes very hard, strong and translucent. It is mostly unreactive and must contain as little iron impurities as possible. Different types of porcelain are used for different applications and are classified into dental, electrical, translucent and hard porcelain [5]. The composition of the starting raw materials and the temperature of fusion determine the type of porcelain. Dental porcelain is usually produced as a dental prosthesis to replace either missing or damaged dental structures [1, 6]. Tooth loss can be very traumatic, upsetting and may be regarded as a serious life event that needs to be solved to avoid social and psychological problems [7, 8]. To improve facial appearance, ability to masticate, ease of speech and maintain oral tissues, it is necessary to replace missing tooth [9]. Other types of dental ceramic restorations like Bioactive Glass Composites[10], ZrO_2 ceramics [11], ZrO_2-dental porcelain [12, 13], Al_2O_3 [3] etc.

There are reports in the literature concerning researches on dental porcelain. Locally available materials have been used to produce dental porcelain [14-16]. The results show that the clay is kaolinitic in nature and the properties are acceptable for standard dental porcelain. Direct Ceramic Machining (DCM) method has been used to investigate the reliability and strength of all-ceramic dental restorations [17]. The DCM method allowed rapid production of all-ceramic dental restorations with high mechanical strength and good biocompatibility. The findings from the study of the significance of glazing and finishing of dental porcelain showed that glazed porcelain produced smooth and dense surfaces with better mechanical properties [18]. An evaluation of the composition, structure and properties of dental ceramics reported that there is need to find a balance between aesthetics and biomechanical strength [7, 19]. A study on the insights of ceramics as dental materials showed that higher crystalline phase content improved mechanical strength [11, 20].

Nigeria has all the raw materials required to produce dental porcelain. According to the Raw Materials Research and Development Council of Nigeria, numerous deposits of these materials exist in different parts of the country [21]. Most of the raw materials are left untapped and there is very little research on the suitability of these materials for the production of dental porcelain. The objective of this work therefore is to investigate the use of these raw materials in the production of dental porcelain and also to study the effect of varying the amounts of silica and feldspar on the properties.

2.0 MATERIALS AND METHODS

2.1 Sample Preparation

The raw materials used in this study include feldspar, silica, kaolin and bone ash. The feldspar and silica were collected from a glass factory (Oluwa Glass Company in Igbokoda, Ondo State, Nigeria) while the kaolin samples were obtained from Edda in Ebonyi State Nigeria. The bone ash was obtained from a cow bone collected from a local abattoir in Lagos Nigeria. The bone was dried, ground and heated in a controlled manner at 1000^{o}C for 5 hours using a chamber furnace. Three different compositions of the dental porcelain were produced. The initial amount (wt. %) of feldspar, silica, kaolin and bone ash are 70, 25, 4 and 1 respectively. In the other 2 compositions, the amounts of kaolin and bone ash were kept constant while that of feldspar was increased by 5 wt.% and silica decreased by 5 wt.% [24].

The raw materials were weighed and ball-milled for 8 h for reduction in average particle size and homogenisation purposes. To prepare the powders for pressing, they were mixed with starch as binder. A hydraulic press (P100EH, model No 38280, Weber Hydraulik GmbH, Germany) operating at 130 MPa was used to produce samples with dimensions 23 mm Ø by 25 mm length. Thermal de-binding of the samples was done in a muffle furnace at a temperature of 400 ^{o}C for 2 h. Two sintering temperatures, 1100 ^{o}C and 1200 ^{o}C were used to sinter the samples at a heating rate of 5 ^{o}C/min and held for 2 h. The samples were allowed to slowly cool in the furnace.

2.2 Sample Characterisation

The chemical composition of the raw materials were determined using an X-ray Fluorescence (XRF) analyser (SKYRAY Instrument: EDX 3600B). X-Ray Diffraction (XRD) was used to characterize the phases in the raw synthesized samples. The XRD measurements were carried out with an Empyrean diffractometer having Ni-filter CuKα radiation. The counting statistics of the XRD study are: step size 0.0260° 2θ, start angle 4.01°, end angle: 75°; scan speed: 0.01° 2θ/sec. Fourier Transform Infra-red (FTIR) spectroscopy (Shimadzu Spectrometer Spectrum 1000, City Japan) was used to analyse the samples in transmittance mode. The samples were prepared using KBr while the resolution of the measurement is 4 cm^{-1}. The linear shrinkage was calculated by measuring the initial and final dimensions of the samples and using the formula as reported by Viruthagiri et al., [22].

The microstructural features of the samples were examined with a scanning electron microscope (Phenom Prox Scanning Electron Microscope Model No: 800-07334). The compressive strength (CS) was determined by placing the samples in a Universal Testing Machine (Instron Series 3369, Load Cell Capacity: 50 KN U.S.A.) under compression mode. The hardness value of the samples was determined using a Brinell hardness tester (Tensometer Type W, Serial number 10055). The diameter of the spherical indenter is 10 mm while the maximum applied load is 200N.

3.0 RESULTS AND DISCUSSION

The chemical compositions of the starting raw powders are presented in Table 1. The major constituents in kaolin are SiO_2 and Al_2O_3 in addition to other minor compounds as impurities. The SiO_2

contents in the kaolin are 51.38 wt. % and 46.53 % while the Al_2O_3 contents are 42.05 and 39.19 % respectively. The feldspar contains 69.12 wt.% SiO_2 while the Al_2O_3 amount is 18.2 wt.%. The amount of the alkali oxides present (Na_2O and K_2O) are 6.24 wt.% and 2.83 wt.% respectively. The amount of the SiO_2 present in the silica sand is 93.49% wt. % while the bone ash contains 51.30 wt. % and 32.17 wt. % of CaO and P_2O_5 respectively.

Table 1: A table showing the chemical composition (weight %) of the raw materials used in producing the dental porcelain

Sample →	Kaolin		Feldspar	Silica sand	Bone ash
Parameter (%)	Edda	Darazo			
SiO_2	46.53	51..38	69.12	93.49	2.31
Al_2O_3	39.19	42.05	18.20	0.014	2.04
Fe_2O_3	0.62	1.39	0.074	0.690	0.16
CaO	0.31	0.16	0.210	0.001	51.30
MgO	0.001	0.02	0.170	0.000	1.55
Na_2O	0.22	1.73	6.24	0.051	1.06
K_2O	0.19	0.43	2.83	0.001	0.13
TiO_2	3.08	2.37	0.173	0.023	1.87
MnO	0.23	0.007	0.00	0.070	0.00
BaO	0.001	0.001	0.011	0.039	0.11
P_2O_5	0.00	0.00	0.05	0.120	32.17
PbO	0.00	0.00	0.00	0.032	0.00
ZnO	0.001	0.005	0.000	0.001	0.00
L.O.I.	9.627	0.457	2.924	5.243	7.30

The Al_2O_3 in dental porcelain is believed to increase its strength and also helps to maintain opacity [23-25]. The coefficient of thermal expansion in the ceramics is improved by the alkali oxides [25] and was present in significant amount (9%) in feldspar. The reported amount of Fe_2O_3 in the kaolin sample is low and may not contribute much to lowering the properties of the dental porcelain. The amount of SiO_2 in the silica is adequate and contains considerable amounts needed in the batches.

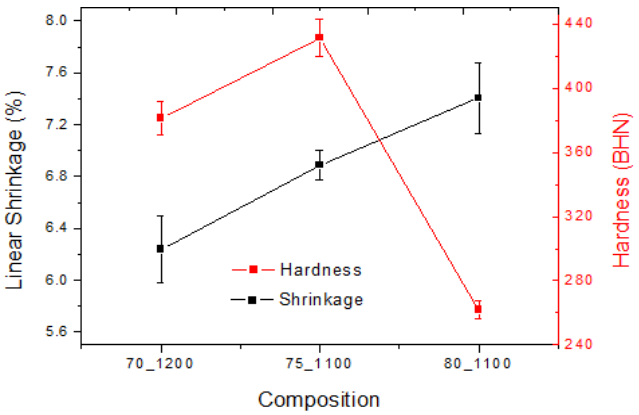

Figure 1: A graph of linear shrinkage and Brinell Hardness (BHN) as a function of sample composition at room temperature.

The graph of linear shrinkage and hardness of the samples as a function of composition is shown in figure 1. As the amount of feldspar in the porcelain increases, the shrinkage rate also increases. The percentage shrinkage is ~6 % for samples with feldspar contents of 70 wt. % sintered at 1200°C and increases to 7 and 7.5 % for samples with feldspar contents of 75 wt. % and 80 wt. % respectively. Sintering of different compositions at the same temperature resulted in a non-uniform shrinkage pattern. High Al_2O_3 content usually leads to a low shrinkage during sintering while high feldspar content results in densification at temperatures as low as 1000°C [26]. Samples with higher SiO_2 contents led to higher shrinkage when sintered at 1100°C. Similar results have been reported in the literature [25].

The hardness values are observed to increase with increasing sintering temperature from 1100 to 1200°C except for the sample with feldspar content of 70 wt. %. Similar observation has been reported by Yan et al., [27] where a decrease in density and poor distribution of pore sizes resulted in lower hardness values.

The X-ray diffraction (XRD) patterns for the samples in the present investigation are presented in Figure 2. Dental porcelain materials can have both crystalline and amorphous phases depending on the sintering temperature used. For the samples sintered at 1100°C, sharp diffraction peaks are observed indicating their crystalline nature while samples sintered at 1200°C showed a broad peak only at Bragg angle of 27°. The other angles where broad peaks are observed at 1100°C disappeared with peak broadening which is an indication of the increase in the amorphous phase. An analysis carried out on the patterns show that for samples sintered at 1200°C, the major phase present is the silicate phase while for samples sintered at 1100°C

with 75 and 80 wt. % of feldspar, a mixture of microcline and sanidine is observed.

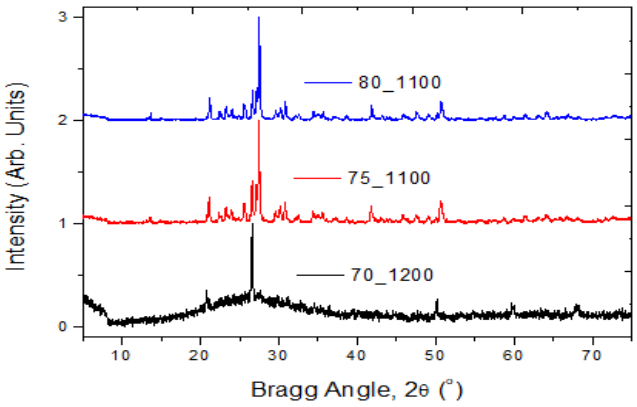

Figure 2: X-ray diffraction patterns of samples 70-1200, 75-1100 and 80-1100. The first two digits indicate the amount of feldspar in the porcelain while the last four digits indicate the sintering temperature.

The morphology of the samples as observed using a scanning electron microscope is shown in Figure 3. The images labelled Figure 3 a-c represent samples 70-1200, 75-1100 and 80-1100 respectively. For the sample with composition 70-1200, a mixture of glassy matrix and very little crystalline phases can be observed. The analysis with X-ray diffraction revealed the presence of SiO_2 phase which is confirmed by the hexagonal structures on the surface of the SEM image. The crystalline phase in the sample is believed to be related to the ilmenite structure. The microstructural images of the samples sintered at 1100°C gives an indication of the presence of flake-like grains. The degree of flakiness however varies with the composition of the samples. The amounts of feldspar in the starting materials also have an effect on the size of the flakes. The average particle size of the sample with feldspar content of 80 wt. % is higher than that with feldspar content of 75 wt. % and lower. Based on the X-ray diffraction, the crystalline phases are believed to be those of Microcline and Sanidine.

The Fourier Transform Infra-red (FTIR) spectra for the samples are shown in Figure 4. The bands observed at wavenumbers 3000 – 3500 cm^{-1} are due to the conversion from kaolin to metakaolin which is an amorphous phase. In the FTIR spectra, broad bands of the amorphous silicate matrix were clearly observed at 490.28 cm^{-1}, 648.10 cm^{-1}, 1000.55 cm^{-1} and assigned to the Si-O-Si bending. The bands at 3500-3750 cm^{-1} are assigned to Si-O stretching vibrations while those at 600 cm^{-1} and 750 cm^{-1}, are attributed to the vibration motion of the bridging oxygen in a direction perpendicular to the Si-Si axis [28]. The sample sintered at 1200°C showed much higher transmittance values compared to samples sintered at 1100°C.

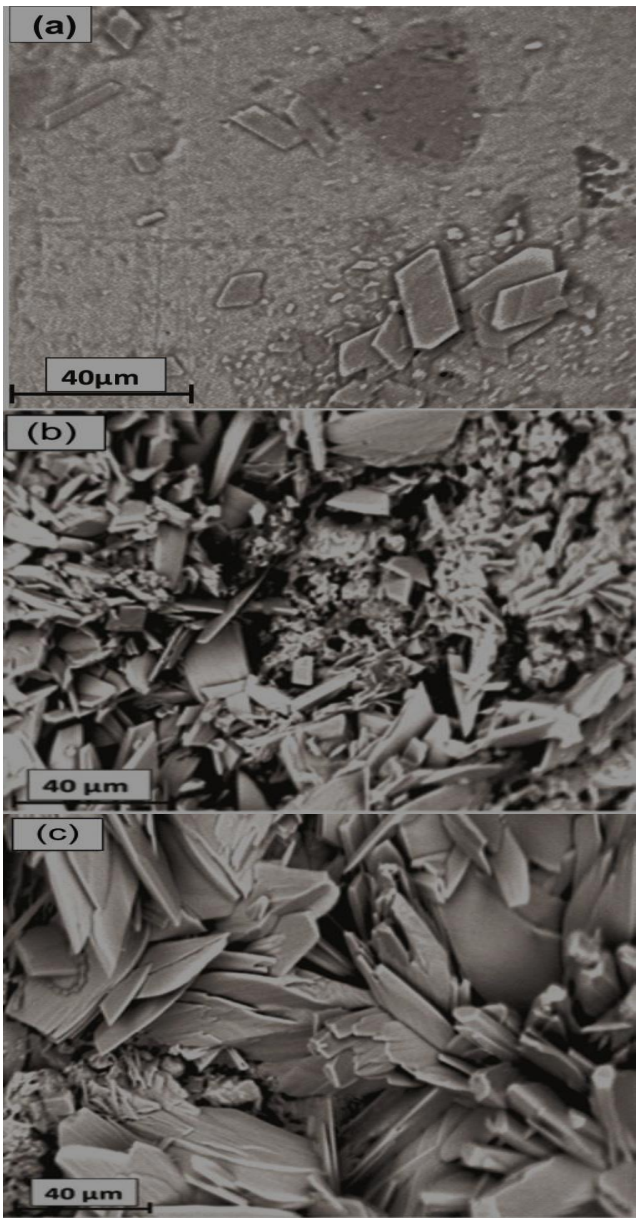

Figure 3: Scanning electron micrographs of (a) 70-1200, (b) 75-1100 and (c) 80-1100 samples

The graphs of compressive stress as a function of compressive strain are shown in Figure 5. For all the samples, as the value of the compressive strain increases, the compressive stress also increases. It is expected that the compressive strength of the samples would increase with increasing sintering temperature possibly due to decrease in sample porosity, increase in bulk density [29]. The formation of glassy (amorphous) phases is also believed to contribute to the higher compressive strength of the samples

sintered at 1200°C. The lower value of the compressive strain for the sample sintered at 1200°C may be due to bloating which increases the porosity thereby lowering the bulk density and compressive strength values [30]. The compressive modulus values for the dental porcelain samples are 228 MPa, 254 MPa and 219 MPa for 70-1200, 75-1100 and 80-1100 respectively.

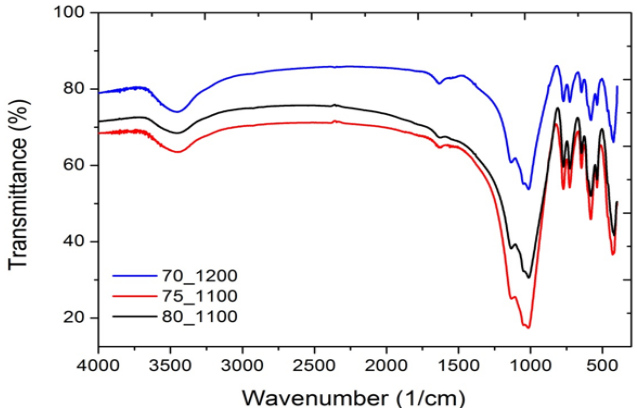

Figure 4: Fourier Transform Infra-red plots of 70-1200, 75-1100 and 80-1100 porcelain samples showing the percentage transmittance as a function of the wavenumber

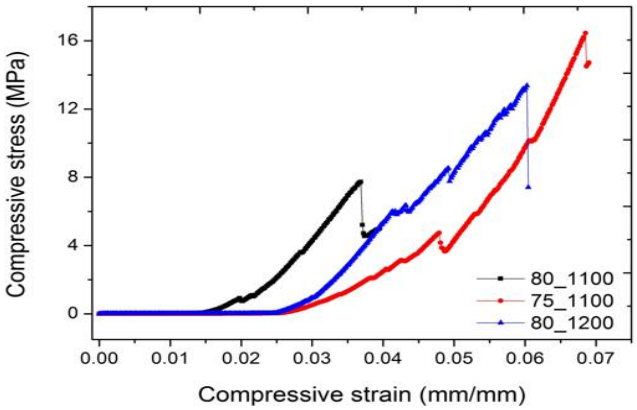

Figure 5: Compressive stress as a function of compressive strain for the porcelain samples

As the amount of feldspar present in the samples increases, the compressive strengths initially increase without a corresponding increase in the compressive strain before failure of the sample. The possible explanation for this is that feldspar which contains high amount of SiO_2 melts at relatively lower temperature and therefore acts as a flux. The molten phases which are formed tend to fill the pores, and decrease the porosity which causes the increase in bulk density of the porcelain body leading to the initial increase in compressive strength [29, 31]. The sample made from the composition with 80 wt. % feldspar is believed to easily form a liquid phase during sintering which blocked the pores resulting in an increased density and compressive strength. The lower compressive strength indicates that the dental porcelain produced were highly porous and less dense.

4.0 CONCLUSION

Dental porcelain has been produced using the conventional ceramics synthesis method from a mixture of feldspar, kaolin, silica sand and bone ash. The dependence of the composition of the raw materials and sintering temperature on the properties of the dental porcelain was clearly observed. Linear shrinkage values ranging from 6.34 - 7.60 %, compressive strength values between 346 - 964.2 N/mm^2 and hardness values between 262-536 BHN were obtained depending on the composition and sintering temperature. The samples sintered at 1200°C consist of a mixture of glassy matrix and crystalline phases such as silica, Ilmenite and Hedenbergite, while those sintered at 1100°C consist of crystalline phases Microcline and Sanidine. The FTIR spectra confirmed the presence of crystalline as well as amorphous phases in the samples. For applications in dental porcelains, the sample with composition 70 wt.% feldspar, 25 wt.% silica, 4 wt.% kaolin and 1 wt.% bone ash and sintered at 1200°C appears to be the most suitable.

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