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Investigating the Suitability of Locally Sourced Kaolinite Clay and Silica Minerals for the Production of Bioceramics

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A B S T R A C T

The suitability of Nsu clay and Ndibe beach sand for the production of dental ceramics was studied. The phase structures and chemical compositions of these raw materials were investigated using the x-ray diffraction (XRD) and x-ray fluorescence (XRF), respectively. Dental ceramic test samples were fabricated and tested for linear shrinkage, apparent porosity and compressive strength properties in order to ascertain their compatibility and suitability. The XRD and XRF results showed that the Nsu clay is kaolinitic in nature consisting of 77.18 % SiO2, 16.64 % Al2O3 and 0.28 % Fe2O3 while the Ndibe beach sand contains high percentage of silica (40.95 %) with no trace of Fe2O3. The physical test results showed a lowest total linear shrinkage value of 10 % for both samples A and B, low apparent porosity values of 2.8 % for sample A, and 3 % for B. These results conform to the acceptable values for the production of dental ceramics. The samples showed moderate compressive strength with the highest value of 64.2 N/m2 for sample C and the lowest value (19.51 N/m2) for sample A.

Keywords: X-ray diffraction, x-fluorescence, dental ceramics, linear shrinkage, apparent porosity

1. Introduction

Bioceramics are electroceramic materials specially designed for use in medicine and dentistry. Though some traditional ceramics are also useful in dentistry. For example, crowns and other artificial dentures made of porcelain fused to metal [Iyasara et al, 2014]. Typical examples of bioceramics include alumina, zirconia, silica, bioactive glass, glass-ceramics, hydroxyapatite, titanium oxide and resorbable calcium phosphates [Nasim et al, 2016]. In orthopedics, bioceramics such as calcium phosphates ceramics, e.g., hydroxyapatite are used for bone replacements in surgeries [Oonishi et al, 2008], while bioceramics composed of SiO_2 and TiO_2 have shown potential for orthopedic regenerating capabilities [Chen et al, 2019]. In dentistry, bioceramics are used for prosthetic dentures, filling bone defects, root repair and for apical retro fills [Nasim et al, 2016].

Generally, bioceramics are desired in medicine and dentistry because of the following qualities [Knapp, 2003]:

a. Hardness, hence able to withstand fatigue when pulled.

b. Resistant to wear. They can withstand long-term wear, and do not shrink or swell with temperature or when in liquids.

c. Impermeability and corrosion resistant. Bioceramics are impermeable to fluids and must not corrode when in contact with body fluids.

d. Non-toxicity. Bioceramics do not cause allergic reactions, hence not toxic to the body.

Bioceramics cover a wide range of materials used in the construction of medical devices that interact with the biological environment. These materials are generally described as biomaterials. A biomaterial is a material exploited in contact with living tissues, organisms, or micro-organisms. It is engineered to interact with biological systems for a medical purpose - either a therapeutic (treat, augment, repair or replace a tissue function of the body) or a diagnostic one.

1.1 Dental Ceramics

Dental ceramics represent one of the fastest growing applications of Bioceramics. Thus, ceramics are used in a wide range of dental filling material such as glass filler extensively used in the construction of crowns for the restoration or replacement of missing teeth [6]. Millions of people seek dental treatment every year and the demands for aesthetic look is on the increase, and ceramics are well suited to meet this demand because of their raw materials' suitability for use in dental fabrication. Dental ceramics are mainly composed of crystalline minerals and glass modifiers. The crystalline raw materials are kaolin, feldspar and quartz (silica) while the glass modifiers include K, Na, or Ca oxides or basic oxides [Babu et al, 2015].

Kaolin ($Al_2O_3.2SiO_2.2H_2O$) is used as a binder in the matrix, and it increases the mouldability of the unfired dental ceramic as well as imparting opacity to the finished dental ceramic product. Feldspar [naturally occurring minerals composed of potash (K_2O), soda (Na_2O), alumina (Al_2O_3)] is an alkali flux

which lowers the sintering temperature and controls the density (and porosity) of the body. Silica (SiO_2) is the lowest fusing component, which melts first and flows during firing thereby initiating these components into a solid mass. SiO_2 also strengthens the fired porcelain (dental ceramic) restoration, it remains unchanged at the temperature normally used for sintering and thus contributes to the stability of the mass during heating by providing framework for the other ingredients. The glass modifiers interrupt the integrity of silica network and acts as flux while Color pigments or frits provide suitable shade to the restoration. The additives such as Zr/Ce/Sn oxides, and Uranium oxide help in developing the appropriate opacity.

The science of dental ceramics for any given application particularly in dentistry involves a study of the materials' compositions, properties and interactions with the applied environment. Therefore, the selection of the materials must be undertaken with confidence and sound judgment [McCabe, 1998]. Many dental ceramics are fixed permanently into the patient's month and removed only intermittently for cleaning. Such materials must have the capacity to withstand the effect of most hazardous environment, variations in temperature, acidity/alkalinity and high stresses.

1.1.1 Functionality and Properties of Dental Ceramics

The dental ceramics fabricated for use as replacement for natural tissues place a very high demand upon the chemical, physical and biological properties. For effective functionality as a dental aid, the dental ceramics must be [Iyasara et al, 2014; McCabe, 1998].

i. Compatible with biological tissues and without eliciting any adverse reactions.

ii. Capable of responding successfully to the stresses and strains.

iii. Able to withstand the corrosive or chemical environment.

iv. Capable of stimulating in most cases, the appearance of natural tissues in terms of both colour and translucency.

v. Capable of being reasonably easy to fabricate by traditional methods.

Dental ceramics exhibit excellent biocompatibility with the oral soft tissues, chemically inert in oral cavity and they possess excellent aesthetics. The structure of dental ceramic restoration is probably the most important mechanical property. The physical and mechanical properties of dental ceramics are summarized in Table.1. The structure of ceramics depends on their composition, surface integrity and presence of voids. The strength also depends on the presence of surface ingredients. The nature, amount, particle size and coefficient of thermal expansion of crystalline phases influence the mechanical and optical properties of the materials [Denry, 1996].

Dental ceramics possess very good resistance to the compressive stresses; however, they are very poor under tensile and shear stresses. This imparts brittle nature to the ceramics and tend to fracture under tensile stresses. Various modes of clinical fractures of ceramic structures include cracks initiating from the contact zone at the occlusal surface from the cementation surface beneath the contact and from the margins of crowns and connectors in fixed partial dentures. Structural defects lead to the failure in dental ceramic prostheses. Defects may arise in the form of micro-cracks of sub-millimeter scale; during fabrication of ceramic prostheses and also from application of masticatory forces in the oral cavity [Denry and Holloway, 2010].

Fatigue strength plays an important role in the durability and longevity of dental ceramic restorations. Fatigue can be accounted for by chemically-enhanced, rate-dependent crack growth in the presence of moisture and cyclic application of stresses [Datla et al, 2015]. Water enters incipient fissures and breaks down cohesive bonds holding the crack walls together and results in initiation of slow crack growth which progresses steadily over time, accelerating at higher stress levels and ultimately leading to failure [Iyasara et al, 2014].

Surface hardness of ceramics is very high; hence they can abrade the opposing natural or artificial teeth. Ceramics are good thermal insulators and their coefficient of thermal expansion is almost close to the natural tooth [Datla et al, 2015]. During firing any residual water is lost from the material accompanied by loss of any binders that results in volume shrinkage of about 30–40 %, due to elimination of voids during sintering. Therefore, a precise control of the condensation and firing technique is required to compensate for such shrinkage value during the construction of ceramics restoration. Adhesion of ceramic restoration to the natural tooth also plays a significant role in the durability of the restoration. The success of a fixed restoration depends on the use of the luting agent and cementation technique [Babu et al, 2015].

The major drawbacks associated with dental ceramics are brittleness, low fracture toughness and low tensile strength. However, there are two methods used to overcome these deficiencies. These methods are brittle materials strengthening and design components strategy to minimize stress concentration and tensile stress [Phillips, 2010].

1.2 Aim and Objectives of the Study

The overriding goal of dental ceramics is to improve the life quality of the dental patients with the aim of providing suitable dental structure replacement. Unfortunately, the cost of these dental structures such as artificial teeth in Nigeria is expensive and also imported. Evidence abound that Nigeria is blessed with huge solid mineral deposits, and if properly harnessed can be utilized in the production of artificial teeth and other structural body parts, and consequently reduce overdependence on imported ones.

Based on the background of the identified problems, this research aims at assessing the suitability of Nsu clay (as a source of kaolin) and Ndibe Beach Sand (as a source of silica) for the production of dental ceramics.

The objectives of this study are:

- To process and fabricate test samples of dental ceramics and carryout experimental tests on the trial samples with a view to producing affordable and compatible dental ceramics of acceptable standard using the local ceramic raw materials
- To analyse the phase structures of the local ceramic raw materials using x-ray diffraction
- To determine the chemical compositions of the ceramic raw materials using x-ray diffraction fluorescence.
- To subject the test samples (dental ceramics) to the following properties associated with dental ceramics: linear shrinkage, apparent porosity, compressive strength and biocompatibility tests.
- To promote local content utilization through the application of local raw materials in the development of ceramics and other products.

Table 1. Physical and Mechanical properties of Dental Ceramics

Compressive strength	330 MPa
Diametral tensile strength	34 MPa
Transverse strength	62 - 90 MPa
Shear strength	110 MPa
MOE	69 GPa
Surface hardness	460 KHN
Specific gravity	2.2-2.3
Thermal conductivity	0.0030 Cal/Sec/cm^2
Thermal diffusivity	0.64 mm^2/sec
Coefficient of Thermal expansion	12 10^{-6}/°C

2. Materials and Methods

2.1 Sourcing of Raw Materials

The ceramic raw materials used were:

i. Nsu clay (a source of kaolin) code-named NSU in this study and was sourced from Agbahara, Nsu in Ehime Mbano L.G.A of Imo State, South East Nigeria

ii. Ndibe beach sand (a source of silica) code-named NBIBE in this study and was sourced from Ndibe beach sand in Afikpo, Afikpo North L.G.A of Ebonyi State, South East Nigeria

iii. Feldspar was obtained from Ijero-Ekiti in Ekiti State, South West Nigeria.

iv. Other ceramic raw materials such as talc and alumina were purchased from a chemical and scientific store in Ariaria market, Aba, Abia State, South East Nigeria.

2.2 Processing and Analysis of Raw Materials

The sourced raw materials were washed, air dried to eliminate contained moisture, crushed, ground, and sieved using jaw crusher, ball mill and mesh 60 and 80, respectively. Thereafter they were stored in an air tight container for further use.

Prior to use, the processed Nsu clay, Ndibe beach sand and feldspar raw materials were subjected to phase structure and chemical composition analyses using x-ray diffraction and x-ray fluorescence, respectively. The analyses were carried out at Royal Steel Industry Kaduna state, Nigeria

2.3 Production of Test Samples

The production sequence adopted in producing the test samples and was communition, sieving, batching, mixing, forming/shaping, drying and firing. The stored raw powders were weighed using weighing balance and batched into A, B, C and D samples with a total batch weight of 1500 g and mixed thoroughly with water in a bowl to form a plastic paste. The calculated batch composition is presented in Table 2.

The paste was wrapped in a nylon and allowed to age for 24 hours in order to obtain a workable mix. Consequently, the formed clay body was wedged and kneaded properly to eliminate air pocket after which it was rolled to form a slab using a rolling machine. It was then measured and cut into dimensions with a length of 12 cm and width 4 cm each. Three pieces were produced for each batch sample and a shrinkage line (10 cm) was drawn at the middle of each of the test samples. The 10 cm line served as the original or green length of the test sample.

All wet test samples were allowed to dry in air for 48 h and thereafter dried for 3 days in a drying cabinet at a temperature of 110 °C. Consequently, the test samples were fired at 1200 °C for 8 h.

Table 2. Batch Composition of Test Samples

Composition	**Test Samples (g)**			
	A	**B**	**C**	**D**
Nsu	630	600	270	300
Ndibe	465	420	270	630
Feldspar	375	450	525	300
Talc	30	30	30	30
Alumina	-	-	300	240
Water (ml)	550	345	390	390

2.4 Physical and Mechanical Analyses of Test Samples

The following basic physical and mechanical properties associated with dental ceramics were determined and carried out in the Department of Ceramic and Glass Technology Workshop:

2.4.1 Linear Shrinkage

Linear shrinkage test was determined by measuring the dimensional changes that occurred in each of the test sample after drying and firing processes and were compared with the green length. Before drying and firing of the test samples, the length of each wet test sample was determined. It was marked as a shrinkage line of 10 cm and was noted as the original or green length.

The linear drying and firing shrinkages of the test samples were then calculated using the following equations:

$$S_d = \frac{L_o - L_d}{L_o} x\ 100 \quad \text{---------- (1)}$$

where, S_d is the linear drying shrinkage, L_o is the original length of test sample and L_d is the dry length of the test sample.

$$S_f = \frac{L_d - L_f}{L_d} x100 \quad \text{---------- (2)}$$

where; S_f is the linear firing shrinkage and L_f is the fired length of the test sample

The combined effect of the drying and firing processes is known as the total linear shrinkage S_t, was determined and calculated using equation 3.

$$S_t = \frac{L_o - L_f}{L_o} x100 \quad \text{---------- (3)}$$

2.4.2 Apparent Porosity

Porosity test is a measure of a fractional void volume of a fired body. In this research, a boiling method was used to determine the apparent porosity, AP. The fired test samples were weighed in air and recorded as original weight, W_1. The measured test samples were transferred into a beaker of water and boiled for 5 h. Upon cooling, the test samples were removed from the water, cleaned and reweighed. The new weight is known as the boiled weight, W_2 of the test sample. The apparent porosity was determined using the formula:

$$AP = \frac{W_2 - W_1}{W_1} x100 \quad \text{---------- (4)}$$

2.4.3 Compressive Strength

The dimensions (length and breadth) of the test samples were measured. The samples were firmly mounted one after the other on the saddle of a locally compressive strength testing machine. A force was applied at a uniform rate by operating the hydraulic pump handle in an up and down movement till

the sample failed. The compressive strength (cold crushing strength) of the sample is therefore the maximum load at failure, and it was determined using the formula;

$\delta = \frac{F}{A} = \frac{F}{lb}$ ---------- (5)

where δ is the cold crushing strength (KN/m^2), F is the Applied force (KN), A is area of sample (m^2), l is the length of sample (m) and b is the breath (width) of the sample (m).

3. Results and Discussion

3.1 Results

The results of the chemical and phase analyses of the raw materials (Nsu clay, Ndibe beach sand and feldspar) are shown in Table 3 and Figures 1, 2 and 3, respectively. Tables 4, 5, and 6 show the results of the linear shrinkage, apparent porosity, and compressive strength tests carried out on the test samples.

Table 3. Chemical Analysis of the Raw Materials

Nsu clay Composition wt.%		Feldspar Composition wt. %		Ndibe beach sand Composition wt. %	
SiO_2:	77.18	SiO_2:	48.37	SiO_2:	40.95
Al_2O_3:	16.64	Al_2O_3:	9.52	Al_2O_3:	3.19
K_2O:	0.59	K_2O:	21.85	K_2O:	11.71
Fe_2O_3:	0.28	Fe_2O_3:	5.21	Fe_2O_3:	0.00
CaO:	0.19	CaO:	3.56	CaO:	6.08
Cl:	1.27	Cl:	0.80	Cl:	1.84
TiO_2:	2.44	TiO_2:	0.70	TiO_2:	1.54
SO_3:	1.27	SO_3:	0.47	SO_3:	2.39
Na_2O:	0.10	Na_2O:	0.45	Na_2O:	0.82
MgO:	0.06	MgO:	0.28	MgO:	0.74
Ag_2O_5:	0.08	Ag_2O_5:	4.49	Ag_2O_5:	5.45
Y_2O_3:	0.07	Y_2O_3:	2.46	Y_2O_3:	4.00
Nb_2O_5:	0.00	Nb_2O_5:	1.90	Nb_2O_5:	4.95
P_2O_5:	0.00	P_2O_5:	0.00	P_2O_5:	9.81
CO_3:	0.00	CO_3:	0.00	CO_3:	7.15

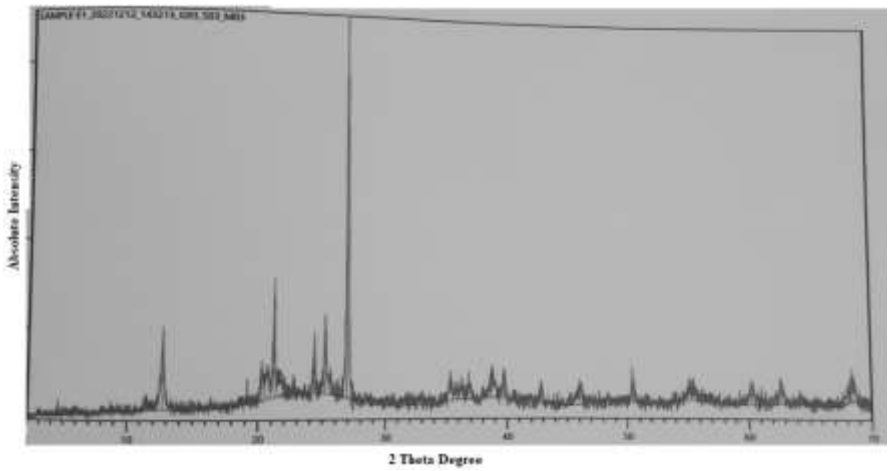

Figure 1. XRD patterns of Nsu Clay

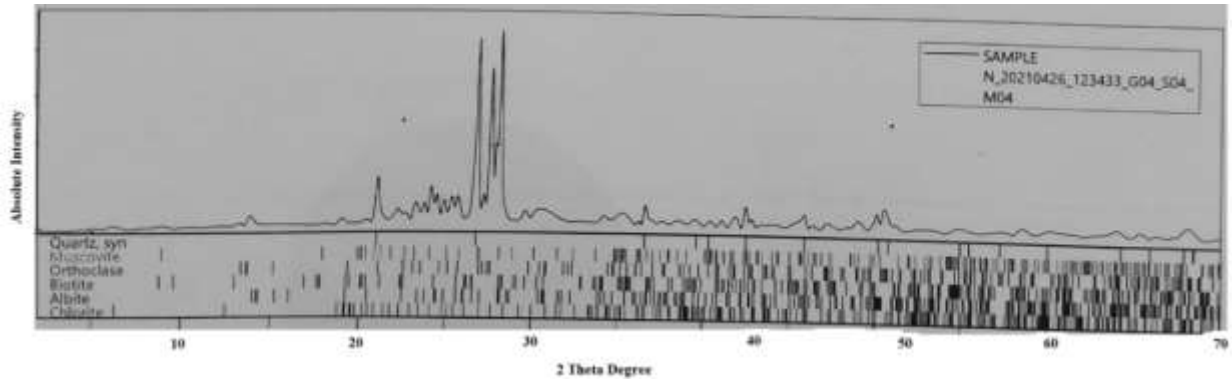

Figure 2. XRD patterns of Ndibe Beach Sand

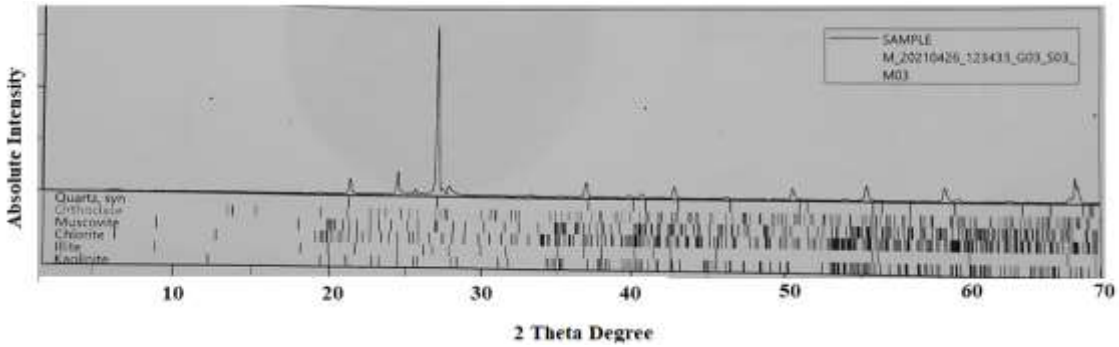

Figure 3. XRD patterns of Feldspar

Table 4. Linear Shrinkage Test Result

Sample	Original length (cm)	Dry length (cm)	Fired length (cm)	Linear shrinkage (%)		
				Dry	Fired	Total
A	10.0	9.6	9.0	4.0	10.0	10.0
B	10.0	9.5	9.0	5.0	5.26	10.0
C	10.0	9.9	9.1	1.0	8.42	9.0
D	10.0	9.8	8.8	2.0	10.20	12.0

Table 5. Apparent porosity test result

Sample	Fired weight W_1 (g)	Boiled weight W_2 (g)	Apparent Porosity (%)
A	338.8	348.3	2.80
B	340.9	351.2	3.02
C	339.2	374.0	10.25
D	311.9	348.4	11.70

Table 6. Compressive strength test results

Samples	Compressive strength (N/m^2)
Batch A	19.51
Batch B	36.0
Batch C	64.2
Batch D	31.5

3.2. Discussion

3.2.1 Chemical and Phase Composition

From the results of the chemical analysis (Table 3), Nsu clay has 77.18 % SiO_2 and 16.64 % Al_2O_3; Ndibe beach sand has 40.95 % SiO_2 and 3.19 % Al_2O_3 while feldspar has 48.37 % SiO_2 %, 9.52 % Al_2O_3 and 21.85 % K_2O. The chemical analysis showed that Nsu clay is a typical kaolinite clay with less than 45 wt. % Al_2O_3 [Iyasara et al, 2016] in conformity with the report that most clay deposits in Nigeria are kaolinite clays. The results of Ndibe beach sand showed values for a typical quartz.

The XRD patterns of the raw materials as shown in Figures 1-3 collaborated the results of the chemical analysis. The diffraction patterns of the Nsu clay exhibited ordered, narrow and intense peaks with the first peaks at 12.65^o (2θ). It has been established that in a poor crystalline clay, the first order spacing (d_{100}) occurs around 10^o (2θ) [Iyasara et al, 2016; Grim, 1971; Grim, 1968]. This therefore confirms that Nsu clay is a poor crystalline clay

3.2.2 Physical and Mechanical Properties

The results of the linear shrinkage are shown in Table 4. Since total linear shrinkage, St is the combined effect of drying and firing processes, emphasis will be on St only. All values except sample D (St = 12 %) of the total linear shrinkage conform to the standard linear shrinkage value range, 7-10 % for porcelain bodies [Chester, 1973; Ovri and Onuoha, 2015].

According to Della and Kelly (2008), dental ceramics must be compactible, aesthetic, and less porous with low shrinkage. Therefore, samples A-C are suitable for the development of dental ceramics.

Table 5 shows the apparent porosity (AP) properties of the dental ceramics. Low AP values of 2.8 % and 3 % were obtained for samples A and B, respectively. This low AP indicates that the dental ceramics depicts increase in durability and strength, and conformed to the acceptable standard [Iyasara et al, 2014; Della and Kelly, 2008]. On the contrary, samples C and D exhibited high porosity values (10.25 % and 11.70 %). These high values might be attributed to the introduction of alumina (Al_2O_3) into the matrix. The Al_2O_3 does not attain its maturity temperature during sintering at 1200 ^{o}C thereby creating additional pores into the lattice.

The compressive strength test results are presented in Table 6. The highest compressive strength (64.2 N/m^2) and the lowest (19.51 N/m^2) are observed in samples C and A, respectively. Although the strength showcased by the dental ceramics cannot withstand structural load, but has the ability to withstand abrasion. Consequently, the compressive strength values obtained are suitable for dental application.

4. Conclusion

Nsu clay and Ndibe beach sand as locally sourced materials were investigated to ascertain their suitability for the production of dental ceramics. The phase analysis showed that the Nsu clay is kaolinitic in nature while the Ndibe beach sand is rich in silica with no trace of iron II oxide.

The physical tests showed that samples A and B exhibited acceptable shrinkage (10 %) and porosity (2.8 – 3 %) levels. Samples C and D contain alumina and thus exhibited high porosity and compressive strength. It is suggested that sintering the dental ceramics at higher temperature (≥ 1500 ^{o}C) might cause the Al_2O_3 to attain its maturing temperature, hence becoming denser.

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