Jean - Eudes BOULINGUI, et. al. International Journal of Engineering Research and Applications
www.ijera.com
ISSN: 2248-9622, Vol. 13, Issue 1, January 2023, pp. 07-14

RESEARCH ARTICLE OPEN ACCESS

Use of Sibang clays in raw and terracotta for housing in Gabon

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ABSTRACT
The clay mineral materials of the Sibang district find use in the raw and terracotta of the building. This work consists in characterizing Sibang clays for use in the formulation of raw and baked bricks for buildings. The clay powders of the Sibang series of Turonian age, in the Libreville region, were the subject of detailed analyzes using laser granulometry, chemical analyzes of the major elements in whole rock, X-ray diffractometry on whole rock and oriented blades, infrared spectroscopy, scanning electron microscopy, cation exchange capacities. Thermogravimetric analyzes (ATD-ATG), firing tests at 1100°C were also carried out on the clay mineral materials of the Sibang series to observe the behavior of melting clays and the transformations of quartz. The main clay phases of Sibang show a dominance of illite and kaolinite associated with quartz. Sibang clays comply with the French standard for compressed earth bricks. The clay mineral materials of Sibang constitute an ecological material and an alternative to the use of conventional concrete blocks in Gabon.
Keywords: Clays, raw and fired bricks, Sibang, Turonian, Uses.

Date of Submission: 20-12-2022 Date of Acceptance: 02-01-2023

I. Introduction

Libreville, a coastal city on the Atlantic Ocean located at latitude 0°23'24" North and longitude 9°27'15" East [1], is located in the sedimentary basin of Gabon [2]. The coastal basin characterized by the Phanerozoic (540 Ma to the present) includes a set of continental and marine deposits ranging from the Permian to the lower Cretaceous [3, 4]. The Sibang geological formations of Turonian age, 500 to 600 m thick, follow the Cape Lopez group. The most abundant facies are marine limestones testifying to a transgression, but punctuated by a sandstone interval attributed to a regressive phase [2]. Towards the edge and the southern Atlantic Basin (South Basin), [2] attribute to the Sibang group occurrences of continental sediments (clays, clayey or siliceous sandstones, ferruginous crusts, kaolinic sandstones with rare conglomerate beds) reputed to be Turonian. In this group near the city of Libreville, samples were taken from uninhabited areas (green spaces) (Fig.1). In the manuscript of [5], the Sibang group could contain clay mineral materials listed in the series of Cap Lopez and that of the Madiéla group. The Sibang group resting on that of Cape Lopez would be the scene of an exchange of mineral materials. Sibang series is a series which has red and gray color clay materials. In Gabon, the shortage of building materials is severely disrupting the national housing construction policy. All clay-based products currently on the national market are imported (bricks, tiles, tiles, crockery, medicines, cosmetics, etc.). To promote the use of local clays under conditions of sustainable development, a prospection of useful resources has identified clay indices in the Sibang series that can contribute to the reduction of the housing deficit problem posed by the Gabonese populations.

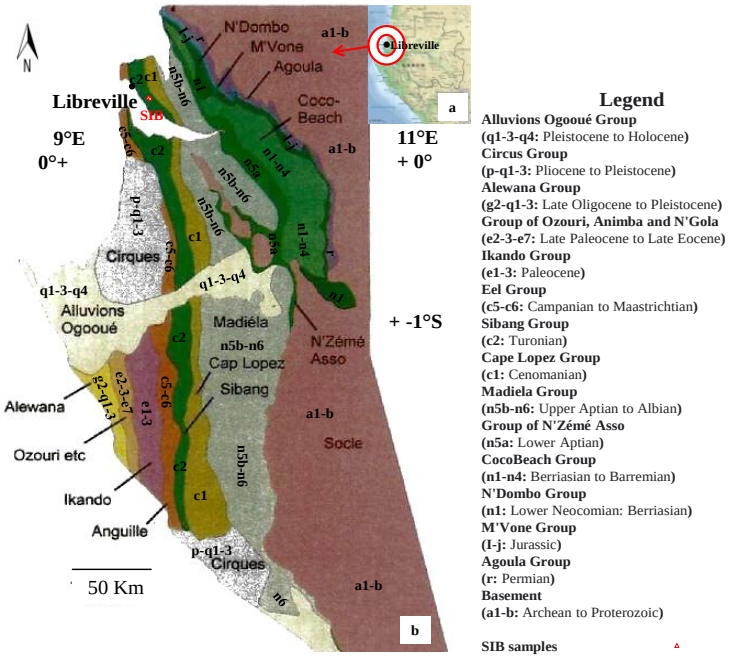

Figure 1: Topographic map of Gabon (a) and location of the Sibang clays (b)

II. Materials and Methods

In each study site, a number, a denomination of choice of the sample (SIB) based on location criteria of the sampling area, GPS coordinates, and one observation per sample were here our basic criteria for the description macroscopic field summary. These sites are made up of fine clay materials of various colors: red and gray. The sampling method used is that of an acquisition of data on the ground according to the information of the literature on the region in terms of topography and geology by the acquisition of maps. The information acquisition method is the one chosen in the field as a sample collection system (SIB 1 and 2) which obeys a protocol defined by the decision of the number of sites to prospect, the determination of the observation locations, carrying out field measurements and collecting samples for laboratory analysis based on the manuscripts of [6, 7, 8].

Sibang clays have been characterized by conventional methods [5]. The particle size distribution was measured by the Fraunhofer laser light scattering method using a "Malvern Mastersizer X" size analyzer. The measurements were made on dilute aqueous suspensions with an obscuration rate of 10 to 12% after ultrasonication. Ultrasound was used to disaggregate the flocculated particles. The chemical compositions of the rock formations of the Sibang series were determined by ICP-AES for the major elements, after fusion with LiBO2 and dissolution in HNO3 using an ICP-AES device. X-ray diffraction was performed by reflection on both random and oriented powders, with a Bruker D8 Advance apparatus, using Co Kα1 radiation (λ = 1.789 Å), under operational conditions of 35 kV and 45 mA.

Data were recorded between 3° and 70° (2θ) for whole rock samples and between 2.5° and 40° (2θ) for fine particles (< 2 µm), stepwise analysis 0.036°, step 3.0 sec. Fourier Transform Infrared Spectroscopy (DRFTIS) was performed in a wavenumber range between 4000 and 600 cm-1 with a Bruker IFS 55 spectrometer. The powdered sample was diluted in KBr (50 mg of sample in 270 mg of KBr). The spectrum of each sample was recorded by accumulating 200 scans at a resolution of 2.0 cm-1. The cation exchange capacity was determined by two different exchange methods using either ammonium acetate or cobalt hexammonia chloride. Particle size, morphology and mineral assemblages were studied using a high-resolution (1 nm) HITACHI S-4800 scanning electron microscope. The samples were coated with carbon. Thermal analyzes (DTA and TG) were carried out on the mineral materials of the Sibang series. The study was conducted for a temperature varying from 20 to 1100 °C under a self-generated atmosphere, with continuous recording. The heating rate was 10°C/min. Al_2O_3 preheated to 1500°C was used as a reference. The apparatus used is of the SETARAM Couplings / Gas Analysis - SETSYS Evolution TGA-DTA/DSC brand. The analyzes made it possible to follow the reactions affecting the material according to the temperatures (endothermic, exothermic, loss of mass). In

particular, the curve reveals levels allowing the different types of water to be quantified [9]. The modal proportions were determined according to the multilinear methods developed by [10, 11] whose unconstrained base expresses:

$$T(a) = \sum_{1}^{n} Mi \times Pi(a)$$

With:
T(a) = Content (%) of element « a » in the material,
Mi = content (%) of mineral « i » in the material and
Pi(a) = proportion of element « a » in mineral « i ».

III. Results and discussions

The clay outcrop is a high hill about 20 meters east of Libreville. A series that presents a stratification with deposits in the form of wrinkles of various red-white colors. These wrinkles have a more or less rounded appearance, steep furrows on the surface of the clayey bed and conchoidal breaks (Fig.2).

Figure 2: Red (a) and gray to white (b) clays from Sibang

The granularity of the sediments of the materials of the Sibang series is one of the mineralogical properties of the clayey phases. Laser granulometry reveals grain sizes (diameter D) in microns related to percentages (%) 10; 16; 50; 84; 90 and 99. This distribution reveals coarse particles in the arenite and pelite boundary of the SIB1 and SIB2 materials with 216.695µm for an average population of 99% grain diameter. According to the classification of granular scales and denominations [12], these materials belong to the group of arenites and pelites with a fine to very fine sandy fraction on the arenite side for coarse to fine fraction on the pelite side in the whole series.

In the classification defined by asymmetry, the material of the Sibang series is classified in the zone of strong asymmetry towards small sizes between 1.6 and 7 µm for the average population [5]. [13], clearly mentions that a cation exchange capacity (CEC) of between 10 and 40 meq/100g is typical of a material dominated by illite.

The cation exchange capacity of the Sibang series material is 15.88 meq/100g justified by the high percentage of illite at 40.10% for 11.68% kaolinite. We confirm that the Sibang clays are mainly made up of illite.

In the X-ray diffractograms (Fig. 3 a), on whole rock, the minerals present are illite at 9.98 Å; 4.46 Å; 2.56 Å, kaolinite at 7.15 Å; 4.46 Å; 2.57Å; 2.35 Å, quartz at 3.35 Å; 4.26Å; 1.82 Å; 2.46 Å; 2.13 Å; 2.28 Å; 1.98 Å and hematite at 2.70 Å; 2.51 Å. In order to find other fine fractions, a treatment of clays from the Sibang series in oriented blades (untreated, glycolated, heated to 550°C) of each sample was carried out. The diffractometry analysis of these oriented blades of clays of the Sibang series, gives results showing mineral phases represented in the X-ray diffractograms (Fig. 3 b) mainly of illite at 10.12 Å; 5.03 Å, kaolinite at 7.19 Å; 3.58 Å, muscovite at 10.01 Å, quartz at 3.34 Å, rutile at 3.25 Å and hematite identified at 2.71 Å. The clay material of the Sibang series has as main clay phases after treatment with ethylene glycol and heated to 550°C: Illite and Kaolinite [14]. Treatment with ethylene glycol does not reveal bulking agents. The presence of rutile in the series may mean that the clays come from the weathering of crystalline rocks [15]. Hydrolysis is sufficient to cause the partial disintegration or transformation of primary

minerals into secondary minerals during weathering. In this series we have a disordered kaolinite. The crystallinity and fineness increase with the disorder justifying the high degree of hydration of the mineral material of the Sibang series.

The material of the Sibang series, through the infrared and the spectra associated with the main bands show the characterization of the associated clay phases. The clay minerals are illite and kaolinite (Fig. 3 c). The recognition is done according to the models for the identification of the main clay minerals mentioned in the manuscript of [16].

Illite is defined by the presence of the main bands at 3622 Cm^{-1} and 3624 Cm^{-1} and the elongation of OH with binding of $2Al^{3+}$. At 3580 Cm^{-1}, wave number, OH elongation associated with the Al^{3+} and Fe^{3+} bond. At 3437 $Cm^{-1}$and 3439 Cm^{-1}, hydration then elongation of OH. Band gap at 1400 Cm^{-1} signifying the absence of traces of carbonates. From 1025 Cm^{-1} to 1035 Cm^{-1}, the elongation of SiO. At 914 Cm^{-1} deformations of OH and binding of $2Al^{3+}$. Gap of the 880 Cm^{-1} band and that at 830 Cm^{-1}, deformation of OH with bond of Al^{3+} and Fe^{3+}, deformation of OH followed by Al^{3+} and Mg^{2+} bond.

Kaolinite, similar to the 3698 Cm^{-1} band, gap in the 3668 Cm^{-1}, 3653 Cm^{-1} and 3654 Cm^{-1} band. These main bands are the OH elongation characteristic for kaolinite clay and the 3622 Cm^{-1} and 3624 Cm^{-1} bands. At 1631 Cm^{-1} and 1639 Cm^{-1} hydration and deformation of HOH. For the band at 1631 Cm^{-1}, it is liquid water associated with the sheet, a sign of small-sized particles. The 1114 Cm^{-1} band, that of out-of-plane SiO elongation. Bands 798 Cm^{-1}, 797 Cm^{-1}, 790 Cm^{-1}, 759 Cm^{-1}, 757 Cm^{-1}, 754 Cm^{-1}, 696 Cm^{-1}, 645 Cm^{-1} and 635 Cm^{-1} are characteristic bands impurities for this material of the Sibang series.

The Sibang series presents disordered kaolinite and illite [17]. The disorder and order parameters are the consequence of faulty stacking of sheets.

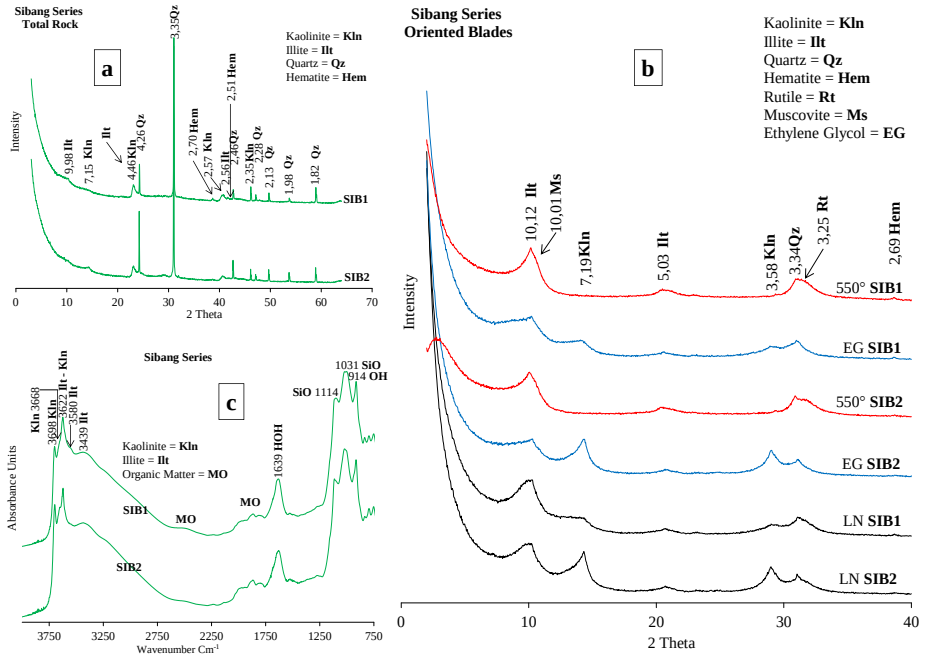

Figure 3: X-ray diffractograms on whole rocks (a), on oriented blades (b) and infrared spectrum (c) of clays of the Sibang series

Fig. 4 shows the morphology of illite spangle. In the prospected range, in addition to silicon and aluminum, potassium, iron and titanium are identified. The presence of sodium and magnesium is recorded. The associated EDS reports a non-pure material composed of trace atomic elements such as magnesium and iron oxide in the form of hematite present in a low percentage in the series.

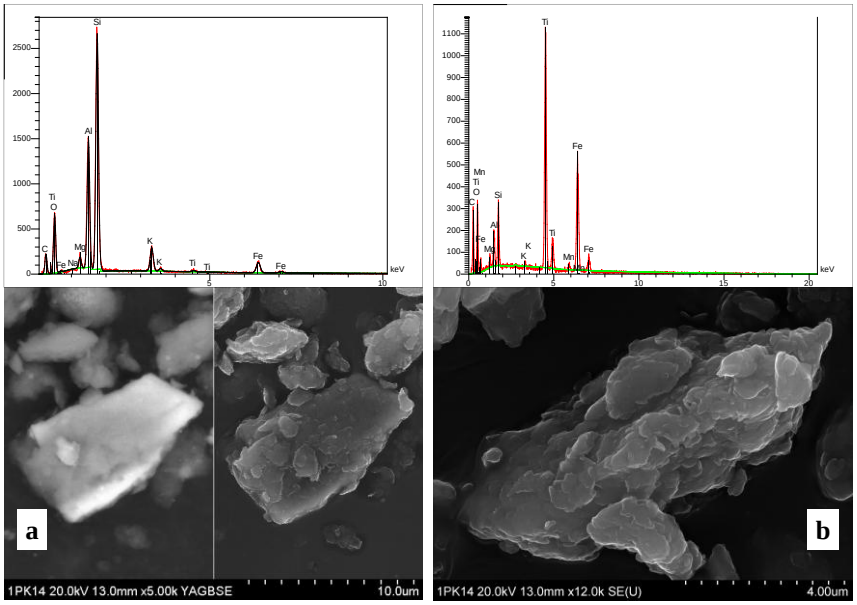

Figure 4: Scanning electron microscope of clay materials of the Sibang series on whole rock

The chemistry is dominated by the aluminum and silicon contents, which is compatible with the mineralogical analysis. We note the very low alkaline and alkaline-earth contents, which testifies to a strong leaching of the bases and indicates that the materials constitute the final stages of alteration (Table 1a). In total rock (Table 1a), the percentages of silicon and aluminum oxide exceed the other oxides in agreement with the presence of dioctahedral phyllosilicates, on average, the materials are around 17.24%, in oxide aluminum and 63.14% in silicon oxide, well above the values quoted by Clarke and Goldschmidt [18].

The proportions of the essential mineral materials that make up this series in average percentages of the mass contents (Table 1b) are Illite (31.3%), Kaolinite (14.99%), Quartz (44.51%), Hematite (6.82%), Rutile (1.46%), Iron oxides (5.67%), Others (1.27%).

Table 1: (a) Chemical analysis of majors and (b) Mass contents of minerals in clay materials of the Sibang series

(a) Chemical analysis of majors

Samples	Major oxides (%)									
	Na_2O	K_2O	CaO	MgO	Fe_2O_3	Al_2O_3	SiO_2	MnO	$P.A.F$	$Total$
SIB1	0,04	2,95	<0,01	1,64	6,82	17,24	63,14	0,03	6,59	98,46
SIB2	0,03	1,63	<0,01	0,94	6,48	14,02	69,00	0,03	6,18	98,31

(b) Mass contents of minerals in clay materials

Samples	Mineral	Chemical formula	Mass content (%)	Total (%)
SIB1	Kaolinite	$Al_2Si_2O_5(OH)_4$	11,69	**99,00**
	Illite	$K_{0,65}Al_2Al_{0,65}Si_{3,35}O_{10}(OH)_2$	40,10	
	Quartz	SiO_2	38,77	
	Hematite	Fe_2O_3	6,82	
	Rutile	TiO_2	1,61	
	Others			**1,00**
SIB2	Kaolinite	$Al_2Si_2O_5(OH)_4$	18,29	**98,46**
	Illite	$K_{0,65}Al_2Al_{0,65}Si_{3,35}O_{10}(OH)_2$	22,50	
	Quartz	SiO_2	50,24	
	Muscovite	Si_3AlO_{10} $Al_2(OH)_2K$	0,45	
	Rutile	TiO_2	1,31	
	Iron oxide	$Fe(OH)_3$	5,67	
	Others			**1,54**

Materials subjected to heat treatments undergo modifications of their physico-chemical properties and of their dimensions by transformation of the initial phases and of the texture [19]. These transformations are studied by thermal analyzes [20, 21] (ATD-ATG coupling, Fig. 5 a and b). In general, under the action of temperature, at the rate of 5°C/min, these materials dehydrate up to 700°C then recrystallize around 1000°C to 1100°C according to the equation:

$$3\,(2\,SiO_2.Al_2O_3.2H_2O) \rightarrow 3\,Al_2O_3.SiO_2 + 4\,SiO_2 + 6\,H_2O.$$

Kaolinite *Mullite* *Cristobalite*

The firing temperature at 1100°C is chosen with reference to the firing test according to the PRE R 28, AFNOR B 40-375 and ISO 51010-1 [22] standards. The 1100°C value is a realistic

estimate of the firing temperature for transforming non-adjuvanted clay products of the Sibang series into terracotta.

Baked products are characterized by X-ray diffraction to determine their mineralogy, and by scanning electron microscopy to observe textural changes. The diffractograms of the materials of the Sibang series, fired at 1100°C (Fig. 5 c) reveal mainly quartz and do not show the main lines characteristic of mullite nor those of cristobalite. After firing at 1100°C, the diffractograms in Figure 5c show iron oxides such as hematite. It is necessary in passing to point out the probable phases: corundum, ilmenite, rutile, spinel. All these phases are identified on the X-ray diffractogram (Fig. 5c).

The investigation at 100 microns (Figure 5 d), clearly shows an overall surface of very smooth quartz grains with a pleasant flank texture aspect on which the iron oxide (hematite) rests on the north-western part. of the beach, titanium oxide in its southwestern part. The entire beach is crowded with aluminum characteristic of the formation of promullite products at the base (Fig. 5d).

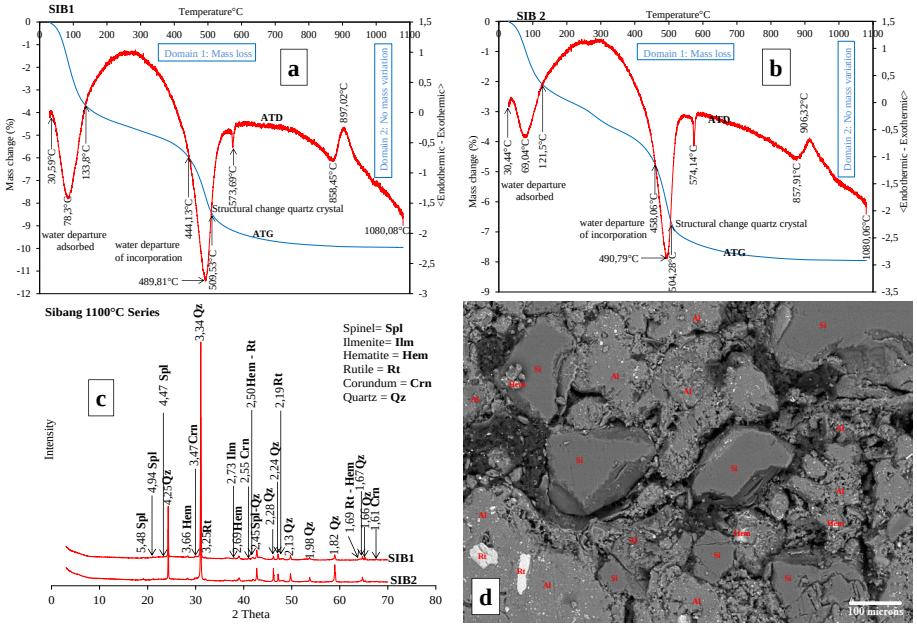

Figure 5: Thermograms (ATD-ATG) of SIB1 material (a), SIB2 material (b), diffractogram of SIB1 and SIB2 materials (c) and SEM on polished face (d) of clay mineral materials fired at 1100°C from Sibang clays

IV. Closing

The clay mineral materials of the Sibang series are conducive to the development of raw and fired bricks for the building with average modal proportions of 31.3% of illite as a natural melting element. All of the results propose to define potential applications and support existing structures that would use unprocessed soil to meet the demand for goods and services [23, 24]. In the classification of variables [25], the mineral raw materials of the Sibang series composed of Illite (31.3%), Kaolinite (14.99%), Quartz (44.51%), Hematite (6.82%), Rutile (1.46%), Iron oxides (5.67%), traces of swelling agents and impurities at (1.27%), have properties quite similar to mixtures intended for the manufacture of mud and fired bricks for the building. As shown by the fine fraction extraction tests, it may be possible to eliminate a large fraction of quartz and iron oxides, which would open up applications in refractories, such as for lining ovens intended to fire tiles and bricks.

Acknowledgments

The author would like to thank the Ecole Normale Supérieure de Libreville (Gabon), the University of Lorraine [Environment and Mineralurgy Laboratory (LEM) / Interdisciplinary Laboratory of Continental Environments (LIEC) and the Rocks and Minerals Analysis Service: SARM - Petrographic and Geochemical Research Center: CRPG] Nancy (France) and the Heterogeneous Materials Study Group of the National School of Industrial Ceramics (Limoges, France) for supporting this work.

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