POZZOLANICITY OF KIMBUNGU BASALT AS A SUBSTITUTE FOR CLINKER TO PRODUCE AN ECOLOGICAL CEMENT IN KONGO CENTRAL

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Abstract: On Earth, there are several natural disasters, many ask the question of what is happening. The Earth is suffering from the harms of global climate change. The DRC, in its Southwestern part, is vulnerable to this phenomenon. The main cause is the presence of cement plants that produce Portland cement, by heating the limestone to about 1450°C. At this process temperature, limestone releases CO_2 which is among the very harmful greenhouse gases. It is for this reason that a study of an ecological cement with 75% clinker and 25% basalt was conducted. The resistance of this composite cement is well.

Keywords: pozzolanicity, basalt, ecological cement, clinker

1. INTRODUCTION

The Democratic Republic of Congo (DRC) hasn't only its large surface area, but it is also known for its great diversity of natural resources and geological formations. All its provinces, including the province of Kongo Central, are rich in several riches and require more in-depth studies to better characterize and develop them economically. Among these resources, Central Kongo Province has several carbonate rocks that can be used for cement production. Thus, there are several cement plants following the presence of these limestone rocks. These rocks consist in large proportion (more than 90 %) of calcite which is calcium carbonate ($CaCO_3$) and a little of dolomite [(Ca, Mg) $(CO_3)_2$]. Calcite releases significant amounts of CO_2 during the production of cement called Portland, this gas is among the Greenhouse Gases (GHG) that contribute enormously to global warming.

The greenhouse effect is that natural phenomenon that causes an increase in the temperature at the Earth's surface. Human activities produce a lot of CO_2 which is a greenhouse gas that is polluting after methane. These gases affect the chemical composition of the atmosphere and lead to the appearance of an additional greenhouse effect which is largely responsible for the climate change currently occurring in the Earth. Today, everyone is aware of the need

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to significantly reduce emissions of these gases. The United Nations has established Conferences of the Parties (COP) every year to consider how to reduce emissions of these gases. Scientists are thinking and working for solutions on this issue. The DRC is not spared from this global phenomenon, its southwestern part towards this province of Kongo Central is more vulnerable and very affected by these effects of this phenomenon possibly following this multitude of cement plants These release significant amounts of carbon dioxide. Normally the CO_2 produced should be captured by floras for photosynthesis. We can observe the evolution of the disappearance of the primary forest (Figure 1) following the cutting of trees, the manufacture of embers (charcoal) and consequently these gases rise in the atmosphere and create harmful effects including excessive heat (heat wave), flooding of the lower parts following torrential rains (Figure 2), ravines in mountainous areas (Figure 3) [1-3].

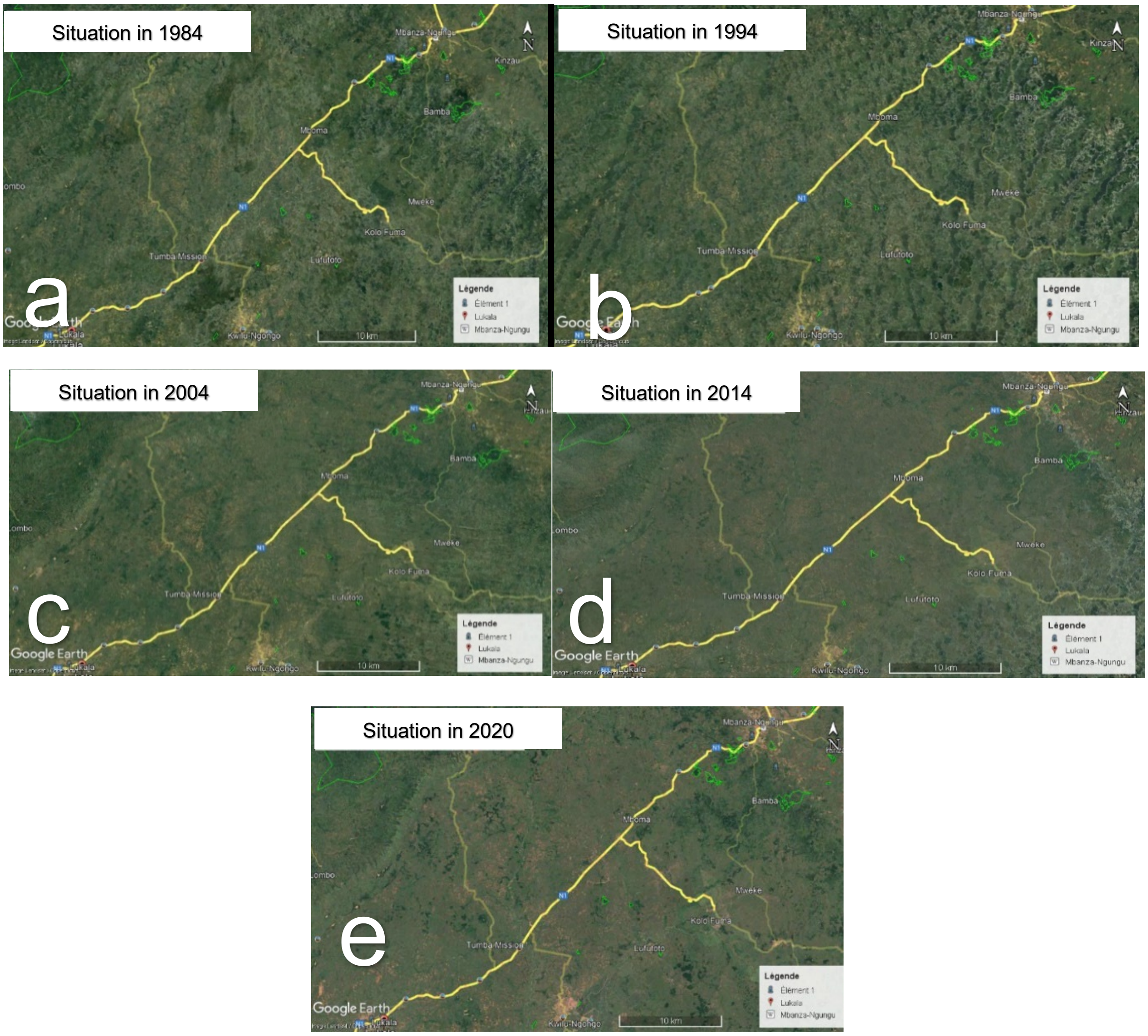

Fig. 1. Map of vegetation change: a) 1984 b) 1994 c) 2004 d) 2014 e) 2020.

It should be noted that while the production of Portland cement produced in the province of Kongo Central, based on limestone which is the main input, mixed with clay, laterite and other components heated to 1450 °C gives clinker, CO_2 and others gasses, this carbon dioxide pollutes, it presents a real danger for the balance of the globe [4] shows that:

- For 100 % (1000 kg) of limestone involved, its decarbonation gives 44 % (440 kg) of CO_2 (CO_2 -process) and 56 % (560 kg) of CaO;
- The combustion of coal to produce clinker also requires for 1000 kg of limestone, 130 kg of coal, this generates 465 kg of carbon dioxide (CO_2 -fuel);
- At the end, it has a total amount of 905 kg of carbon dioxide for one ton of limestone invested.

Fig. 2. Images from the affected zone: a) a bus is driving on a flooded road; b) collapse of a building following a torrential rain.

Fig. 3. Images with the affected road infrastructure: a) National road no 1 cut by a ravine; b) University of Kinshasa-Kimwenza road cut by the ravine.

Following this huge amount of CO_2 generated just for 1000 kg of limestone, it is therefore necessary to find a rock playing the role of a more reactive natural pozzolana to substitute clinker. The test begins with 25 % of basalt for preserving the physico-chemical and mechanical properties of ordinary cement. We were obliged to look for this pozzolanic rock by substituting it for clinker to produce an ecological cement type 32.5R, qualified as a CEM II/B. This cement must comply with standard NF EN 196-5 [1], to overcome the dilemma of the growing demand for cement and the reduction of CO_2 emissions which is currently a global requirement.

In Kongo Central, the province where these cement plants are located, there are not only sedimentary rocks, including limestone, which is an essential input to produce Portland cement, there are also magmatic plutonic, hypovolcanic or even volcanic rocks. When heated to 1450 °C, the inputs (limestone, clay, laterite, sand) form the clinker that is cooled abruptly from 1200 °C to around 200 °C. Volcanic rocks also are formed after a sudden cooling of magma when it reaches the earth surface. This subjects these rocks to the same setting mechanism as clinker (called quenching). It is also necessary that the content of calcium oxide and magnesium favor the use of this natural pozzolanic rock to be clinker substituent and at the same time alkaline oxides (Na and K) must be less abundant in these rocks [4-9]. This is the case of basalt, metabasalt, dolerite, shale. For this phase, we studied and used Mukimbungu basalt to reduce these carbon dioxide emissions (Figure 4).

The reactivity we are looking for in this clinker substitute rock is referred to as pozzolanicity. It is verified by the sum of the major oxides and the difference of silica and lime and are calculated by the following formulas:

$$SiO_2 + Al_2O_3 + Fe_2O_3 > 70\% \quad (1)$$

$$SiO_2 – CaO > 34\% \quad (2)$$

The pouzzolanicity index or pouzzolanic activity index (IP) must also be in the range of 67 % to 100 % i.e. $0.67 \leq IP < 1$ (or $67\ \% \leq IP < 100\ \%$) according to ASTM C-18 [9-12]. Mineralogy should also be tested with reactive silicates and aluminates [6]. According to [2], this pozzolanicity should improve the compactness and reduce the permeability of cementitious materials. The physical, chemical, or even mineralogical properties of cement produced from industrial clinker (75 %) and the substitution of 25 % basalt must lead to the production of a CPJ or EMC II/B cement whose mechanical properties remain preserved. Polymorphs show different physical and chemical properties [7]. The aim of this study is to reduce or to decrease the huge amount of CO_2 in the production of composite cement (cement low carbon or green cement).

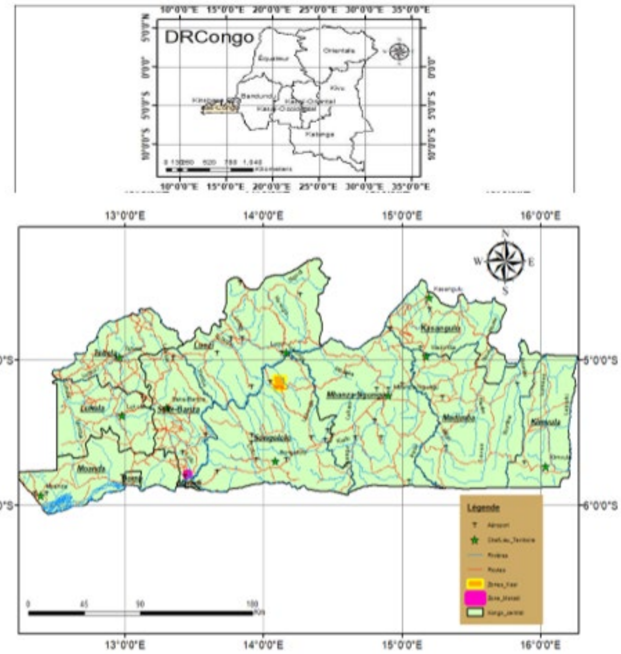

Fig. 4. Geographical location of the Mukimbungu site in Kasi.

2. EXPERIMENTAL SETUP

The basalt of Kimbungu a few kilometers from the river Congo and some 70 km from the national road n°. 1 was used in the present study from several different sites as shown in Figure 5 below. The geographical coordinates for the geolocation were taken by the Garmin 60 GPS map and the mass of 5 kg was used to take the samples to be analyzed in the laboratory. These samples were taken from natural outcrops of basalt flows.

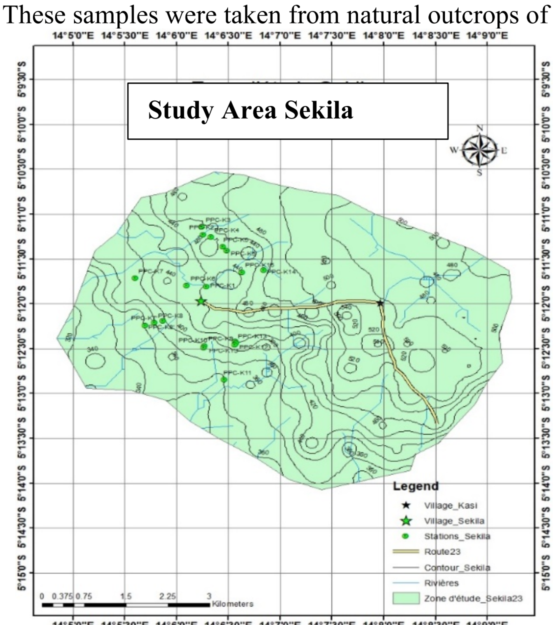

Fig. 5. Sekila/Mukimbungu sampling sites.

This Kimbungu massif is also described by many as the Mukimbungu massif. It is in the district of Cataractes, territory of Songololo and sector of Wombo a few kilometers from the town of Luozi and the Congo River. To get there you must take the road to luozi leaving the site of PPC, road that we leave to the left towards Kasi. There are many outcrops of basalt flows in Sekila village.

From the geological point of view, this massif is made of an anticlinal structure and affects the sedimentary rocks of Western Congo. These West Congolian formations consist of the various sedimentary rocks dated to the Upper Precambrian, precisely the Neoproterozoic (also called Upper Proterozoic). It should be noted that magmatic rocks, of a basic or even neutral nature, are found towards the base of these formations in the form of lava poured into the lower diamictite, or in the form of sills injected into the underlying rocks. These sills have intruded either in the part of the Lower Diamictite, which is older than that where the flows appear, or also in the Sansikua which is older than the Lower Diamictite of quartzo-schist composition. These basalt flows or dolerite are affected by the folding of western Congo, which is characterized in the region that concerns us by vigorous tectonics and by low-degree metamorphism [5].

After the descent of the ground, the next step was that of the expedition of the samples to the laboratories for the characterization not only petrographic, chemical, mineralogical but also the mechanical characterization of the cement that will be produced. We did the analyses: petrographic with a polarizing microscope, chemical with the X-ray fluorescence spectrometer (XRF), X-ray diffractometry (XRD). The basalt samples transformed into melted pearls and then they were mixed with a suitable fondant whose composition is according to [8], 66 % Li_2 B_4O_{7-32} % LiB0 2-0.5 % LiBr. Its principle consists in the dispersion analysis of wavelengths on the pearl. The spectrometer crucible is made of Au/Pt alloy in a ratio of 5%/95% to resist heat [4]. The Lime Saturation Factor (LSF) controls the clinker ratio by the following formula and it is reported to give the maximum amount of silicates [3]:

$$LSF = [\%CaO / (2.8 \times \%SiO_2 + 1.2 \times \%Al_2O_3 + 0.65 \times \%Fe_2O_3)] \text{ x } 100 \quad (3)$$

The lime saturation factor controls the alite/belite ratio in the clinker. A clinker with a very high lime saturation factor has a higher proportion of tri-calcium silicate C_3S compared to bicalcium silicate C_2S than a clinker with a low LSF. Typical values of this factor in modern clinkers are in the range of 0.92 to 0.98 (or 92 % to 98 %). Values above 1.0 indicate that free lime is present in the clinker. Indeed, at LSF equal to 1.0; all the free lime should have been combined with belite to form more alite. If the LSF is greater than 1.0; Excess free lime has nothing to combine and will remain free.

The oxides obtained by chemical analysis make it possible to calculate pouzzolanicity index or pouzzolanic activity index IP. It is the ratio between the compressive strength value of the substitute mortar which contains the proportion of natural pozzolana of 28 days and a control mortar of ordinary quality clinker of 28 days. This pozzolanicity index is calculated by the relation [13]:

$$IP = \text{resistance of substitution mortar/resistance of control mortar} \quad (4)$$

Chemical analyses also make it possible to use the Bogue calculation to verify the compositions of alite and belite [14]. The following formulas make it possible to calculate the 4 anhydrous mineral phases of clinker:

$$C_3S = 4.071CaO – 7.6024SiO_2 – 1.4297Fe_2O_3 – 6.7187Al_2O_3 \quad (5)$$

$$C_2S = 8.6024SiO_2 + 1.0785Fe_2O_3 + 5.0683Al_2O_3 - 3.071CaO \quad (6)$$

$$C_3A = 2.6504Al_2O_3 – 1.6920Fe_2O_3 \quad (7)$$

$$C_4AF = 3.0432Fe_2O_3 \quad (8)$$

The Bogue calculation makes it possible to calculate the approximative proportions of the four main mining phases of Portland cement clinker. Bogue's standard calculation refers to cement clinker rather than cement.

The mineralogical analyses were carried out by XRD by the Bruker brand diffractometer with 16 channels from the Heidelberg group cement/Cilu laboratory. The XRD made it possible to have the idea on the overly reactive polymorphs of the belite found in the clinker. According to [11], there are therefore two polymorphs which are α-

C2S and β-C2S. This analysis highlights the alitic nature of clinker. The equipments used on this study are shown on the Figures 6-9.

Fig. 6. Images with crusher machines:
a) Crusher of the model ZBSX-92A Standard Sieve Shake; b) Ball mine crusher.

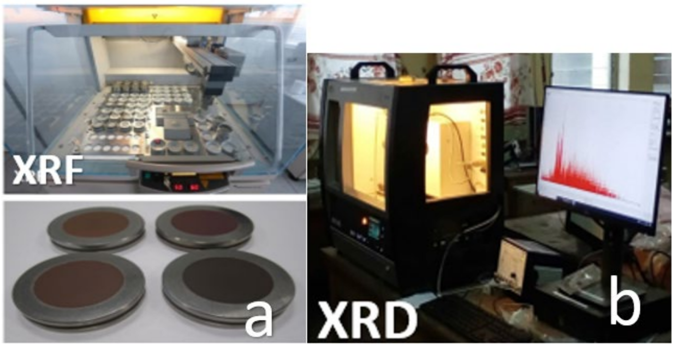

Fig. 7. Images with evaluation procedure:
a) XRF of model Bruker Tiger S8; b) XRD of model Bruker.

Fig. 8. Images with mixing machines:
a) Mixing machine which the model is on the side; b) Mixing of model UTEST.

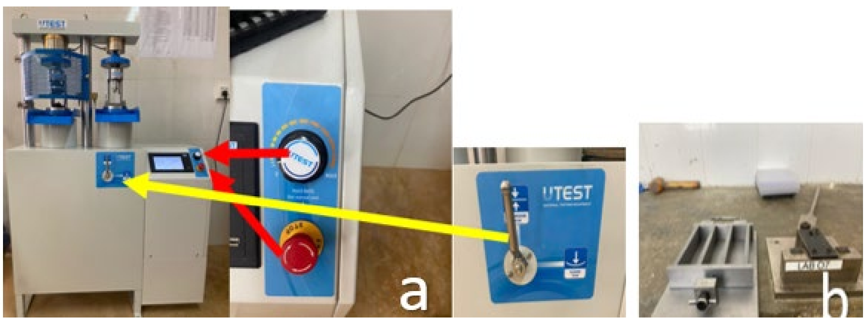

Fig. 9. Images with test equipments:
a) presse which the model is UTEST; b) the Moules.

3. RESULTS AND DISCUSSION

With regard to petrographic analyses, it should be noted that a few years ago a concise petrographic study made on some samples of greenstones taken in this Kimbungu massif had shown the existence of basaltic flows of spilitic character which included two differents facies following the research of [5]. We observed and took samples of two facies, on one side of the aphanitic lava with too fine grains not correctly identifiable to the naked eye, these lavas are also massive and dark in color. On the other side of the lava are very heterogeneous, they have a dark green coloration following the abundance of chlorite. The association of these two types of lithology, in the form of fragments of the first type coated in a paste of the second type, is characteristic of a hyaloclastite or a pillow breccia. However, we must point out that the real cushion lava has not been observed in the West Congo, these basalts are underwater. But nevertheless, we found that by breaking a sample of unaltered and large rock, we observe very characteristic cushions, purplish-gray, massive with fine crystallinity (Figure 10).

Fig. 10. Images from the field with the samples: a) Kimbungu basalt outcrop; b) Sample with pillow.

This outcrop has been described as "weathered, coarse-pebbled blocks of green-gray lava", with angular pieces of tonsillar lava, rich in calcite. The paste of the breccia is itself made up of elements also finer, chloritous, hyaloclastic. It is a hyaloclastic breach with aphanitic lava elements. In some places there are no pebbles and note that this term is used to designate angular elements. In many samples there is the presence of sulfide whose pyrite is dominant, it is sometimes altered. Note that we sent three samples (KM01 KM02 and KM03) to the petrography laboratory for thin slides with a polarizing microscope.

3.1. Macroscopic description of sample KM01

There is a rock of dark gray or grayish coloration and very massive. The minerals are fine and there is a trace of iron sulfide such as arsenopyrite. The weathering of the rock gives a reddish coloration. The rock outcrops on a river called Kunku having ENE-WSW flow direction, it is partially altered leaving boxworks or vugs after the alteration of sulfides (Figure 11).

Fig. 11. Image with samples places:
a) outcrop at station KM01 b) sample taken.

3.2. Microscopic description of sample KM01

The rock has a microlithic texture highlighted by a mesostasis in which the plagioclase rods are dispersed. The latter have a white tint in LPA and colorless in LPNA as shown in the Figure 12.

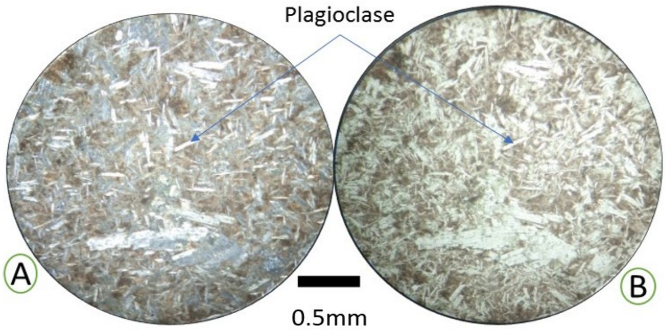

Fig. 12. Thin basalt blade (KM 01): A) LPA; B) LPNA.

3.3. Macroscopic description of sample KM03

Intra-stratified rock (diamictite + basalt) that contain pebbles included within the mass (clasts). The minerals are fine, the greenish coloration and its alteration gives a yellowish coloration. We can observe the boxworks and the vugs due to the alteration of sulfides here pyrite because in some places we can see the dissemination of pyrite (Figure 13).

3.4. Microscopic description of sample KM03

The rock has a microlithic texture evidenced by a mesostasis that encompasses white (LPA) and colorless (LPNA) plagioclase rods. The alignment of these plagioclases one behind the other gives the rock a fluid texture. Parallel to the latter, filaments of iron oxides are observed as shown in the Figure 14.

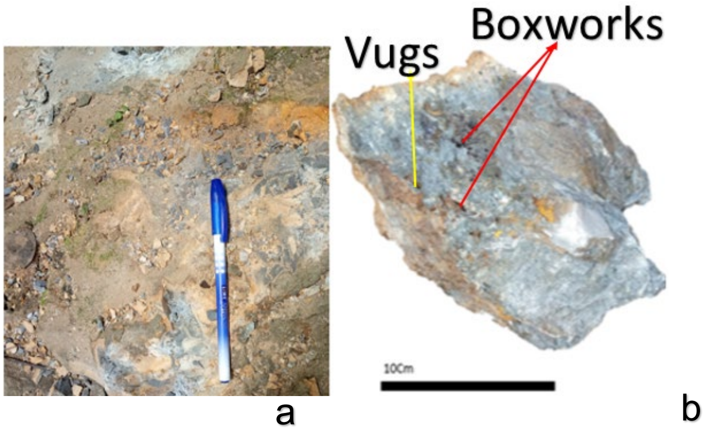

Fig. 13. Image with samples: a) outcrop at station KM03; b) sample taken.

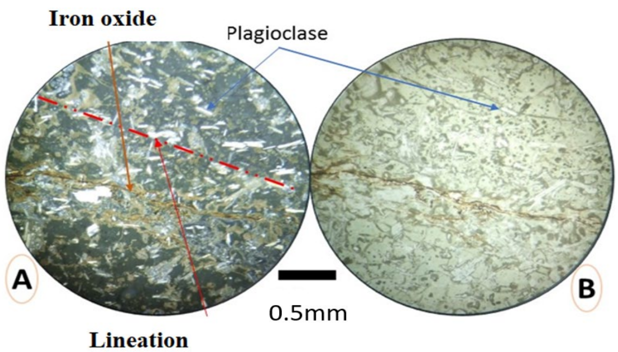

Fig. 14. Thin basalt blade (KM 03): A) LPA; B) LPNA.

The results of oxides displayed in the following table are based on chemical analyses with X-ray fluorescence spectrometry (Table 1).

Table 1. Results of chemical analysis by XRF.

Oxide (%)	KM01	KM02	KM03	CLINKER
SiO_2	45.24	41.24	40.85	22.08
Al_2O_3	14.45	13.46	13.59	6.09
Fe_2O_3	17.58	21.47	24.86	4.08
CaO	5.98	5.92	6.09	64.49
MnO	0.18	0.20	0.24	0.03
Ti_2O	2.06	2.92	1.03	0.00
MgO	5.41	5.60	5.52	2.11
SO_3	0.24	0.20	0.13	0.81
P_2O_5	0.26	0.24	0.14	0.09
Na_2O	3.90	3.35	3.44	0.35
K_2O	0.32	0.28	0.38	0.56
LOI	4.49	5.24	4.02	-
Total	100.11	100.22	100.29	100.69

The oxide diagrams of the KM01, KM02, KM03 and clinker samples are shown in the Figures 15 and 16. From these results, we first evaluated the degree of alteration (Alteration Index AI) which expresses the relative mobility of the elements in the sampled rock. Note that the initial chemical composition of a rock is generally affected to varying degrees by the action of one or more processes including the movement of meteoric waters, metamorphism, the circulation of hydrothermal fluids. The alteration index provides a good indication of the intensity of alteration that has affected igneous rocks.

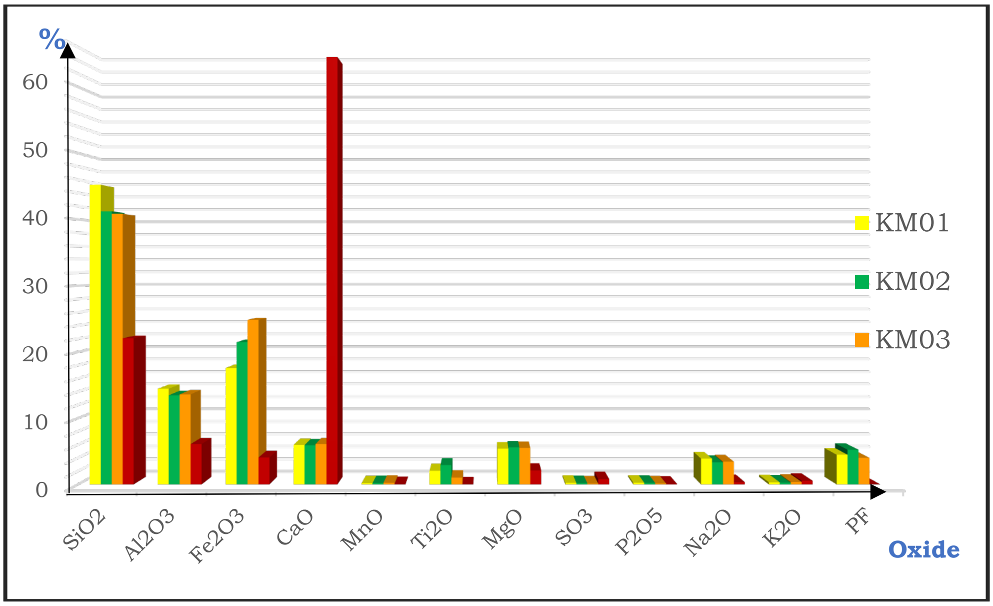

Fig. 15. Diagram of major oxides.

[2] noted that chloritization and sericitization of ferromagnesian rocks lead to values of A.I > 50% while albitization shows the decrease of this index. The chemical analyzes allowed us to determine the alteration index (A.I) (Figure 17) by the formula below and the results are in Table 2.

$$AI = [(MgO + K_2O) / (MgO + K_2O + CaO)] \times 100 \quad (9)$$

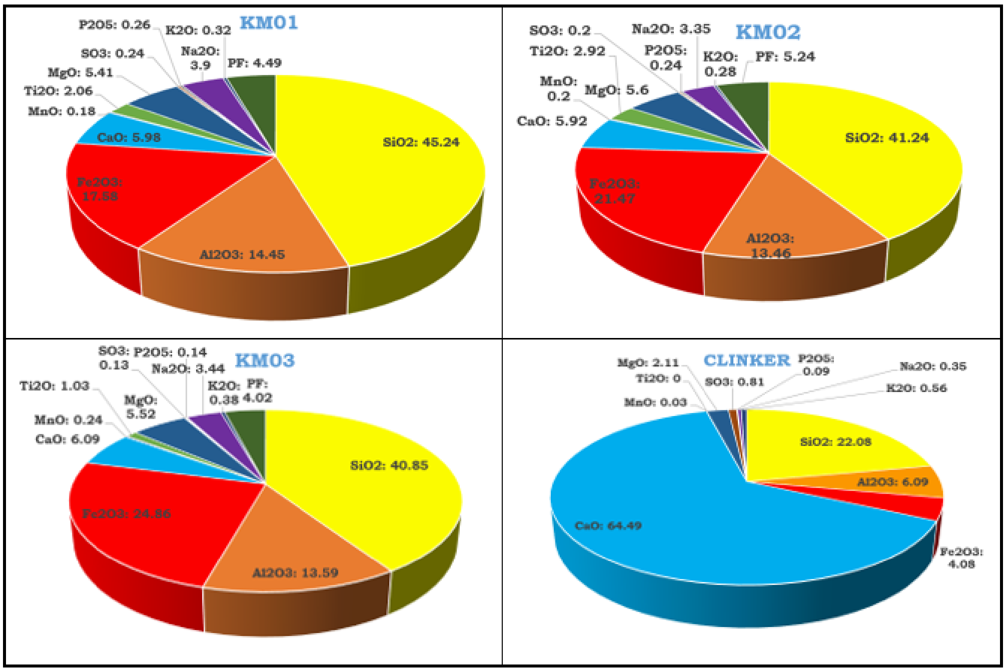

Fig. 16. Diagrams of major oxides for samples KM01, KM02, KM03 and Clinker.

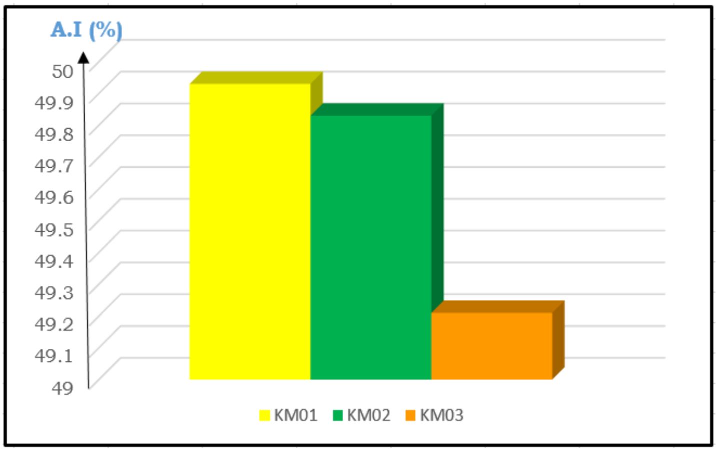

Fig. 17. Diagram of the AI.

Table 2. Chemical index of alteration results of samples KM01, KM02, and KM03.

Samples	AI (%)
KM01	49.93
KM02	49.83
KM03	49.21

According to the results obtained, our three basalt samples underwent albitization according to [15]. These rocks are also very rich in extremely small plagioclase to the naked eye, almost invisible, a little visible with a magnifying glass. Under the microscope, an abundance of plagioclase is observed in the form of small microlithic rods.

With regard to the lime saturation factor (LSF): LSF = 64.49 / [2.8 (0.2208) + 1.2 (0.0609) + 0.65 (0.0408)] = 64.49 / 0.71784 = 90%.

The following values (Table 3) were found after using Bogue's formula (the Bogue calculations) (3), (4), (5), and (6) as mentioned above.

Table 3. Major mineral phases of clinker calculated by Bogue's formula.

	C_3S (%)	C_2S (%)	C_3A (%)	C_4AF (%)
The Bogue Calculation	48	27	9	12

According to [15], a lime saturation factor greater than 89% shows albitic clinker and the opposite will show belitic clinker. After the chemical analysis of the clinker, the formulas of Bogue lead us to a lime saturation factor of 90% which is beyond 85%. Our clinker is therefore alitic (Figure 18).

[12] recently showed the positive influence of basalt on the grindability of our clinker used for the test. The blaine required by the clinker grindability standard must be around 4000 $cm^2.g^{-1}$ [13]. For this study, we reached our target already at 30 minutes of grinding, so we reached 3960 $cm^2.g^{-1}$ for clinker test (Figures 19÷20).

The mineralogical composition of clinker using XRD has generated different polymorphic ore grades of four mineral phases (Table 4).

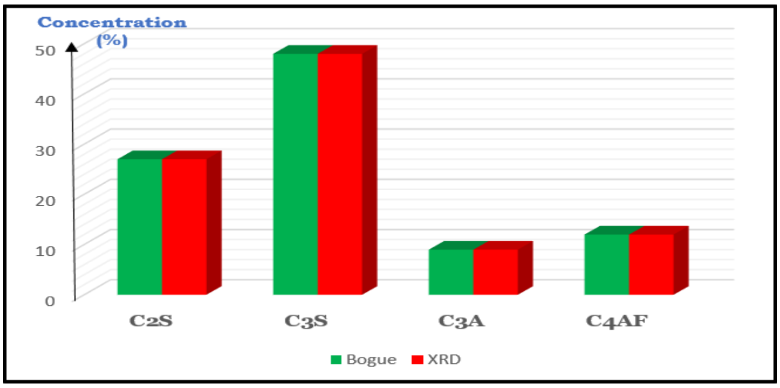

Fig. 18. Major mineral phases of clinker by Bogue and XRD.

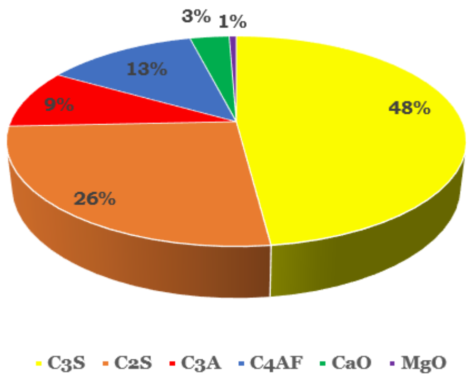

Fig. 19. Diagram of major mineral phases of clinker.

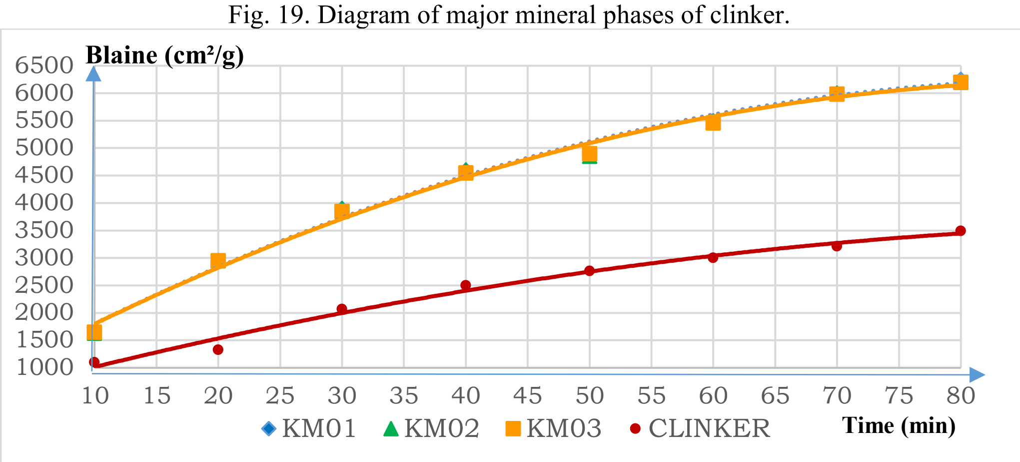

Fig. 20. Variations for KM01, KM02, KM03 and Clinker.

A good way to characterize the pozzolanicity of a rock or a material is not only to evaluate the decrease of the free lime that remains in the system, but also the mobility of the reaction [9]. Factors that commonly affect the pozzolanicinty are the nature of active phases and their percentage [10].

Table 4. Ore grades of differents polymorphs of clinker mineral phases.

Phases	Polymorphs	Ore grades (%)
Alite	Monoclinic-1 Monoclinic-3 Orthorhombic α‘-C_2S	18.9 15.2 13.9
Belite	Monoclinic β-C_2S	26.3
Cerite	Cubic Orthorhombic	3.1 6.0
Tetracalcium Alumino ferrite	Orthorhombic	12.7
Free lime	Cubic	3.3
Periclase	Cubic	0.6

When we look at the proportions of diverse mineral phases in the previous table, we notice that those ore grades support those found, using Bogue's formula.

Three (3) requirements to infer the pozzolanicity of a material are as follows:

$$SiO_2 + Al_2O_3 + Fe2O3 > 70\% \quad (10)$$

$$SiO_2 – CaO > 34\% \quad (11)$$

$$0.67 \leq IP < 1 \text{ or } 67\% \leq IP < 100\% \quad (12)$$

The Table 5 shows results of calculations of the 1st and 2nd pozzolanicity requirements.

Table 5. Results of the calculation about the pozzolanicity requirements.

Pozzolanicity requirements	**KM01 (%)**	**KM02 (%)**	**KM03 (%)**	**Average**
$SiO_2 + Al_2O_3 + Fe_2O_3 > 70\%$	77.27	76.17	79.30	77.58
$SiO_2 – CaO > 34\%$	39.26	35.32	34.76	36.45

We can clearly tell that the first two pozzolanicity requirements are obviously met as for our three samples of basalt. We conducted physical testing of produced cement based on 75% clinker and 25% of basalt substitution. After 2 days and 28 days of drying treatment and pozzolanicity index IP, resistances are revealed below in Table 6. We can say first that pozzolanicity is the ability of some rocks or materials that contain less or none of hydraulic properties owned within themselves, to produce binding substances like those of Portland cement, either into contact with water, or lime and at ambient temperature.

However, mixing pozzolan and cement shows such technical qualities, but also economic ones [11] as pozzolanic cements are industrially produced nowadays, in such way that mixing of pozzolan into cements is referred to many standards, in this instance, NF EN 197-1 and ASTM-C-618 standards that we have used [12].

Table 6. Result of the pozzolanicity index.

ASTM 0.67 ≤ IP < 1				67 % ≤ IP < 100%
ID-Cement	Blaine (cm²/g)	Basalt Substitution (%)	R (28 days) MPa	IP (%)
KM01	4030	25.0	37.59	74
KM02	3970	25.0	38.33	76
KM03	4000	25.0	37.01	73
Clinker	3900	0.00	50.50	100
Average/ KM01, KM02 et KM03	4000			74

Pozzolanicity index testing of three samples generates good values above the standards (Figures 21-22). The KM01 sample produces respectively a resistance of 15.64 MPa for 2 days and 37.58 MPa for 28 days. The second sample KM02 has produced respectively a resistance of 15.83 MPa for 2 days and 38.33 MPa for 28 days. The last sample KM03 produces a resistance of 14.99MPa for 2 days and 37.01MPa for 28 days.

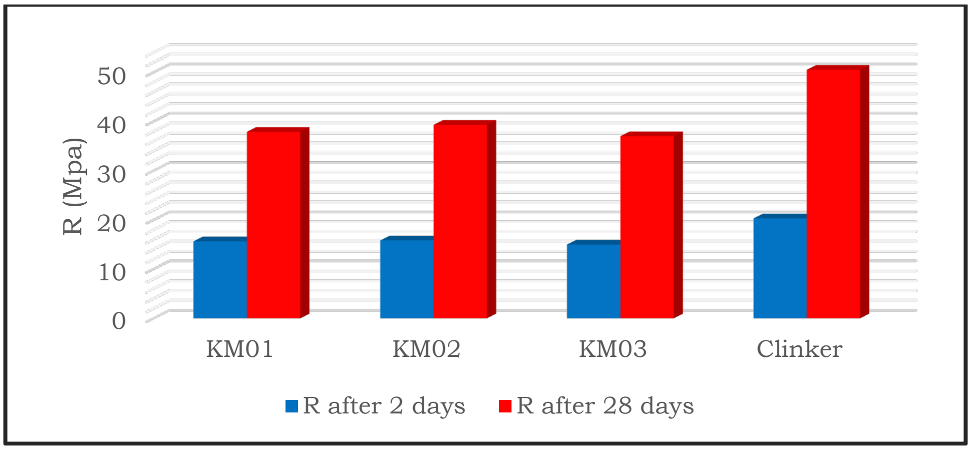

Fig. 21. KM01, KM02, KM03 and clinker diagram of resistances for 2 and 28 days.

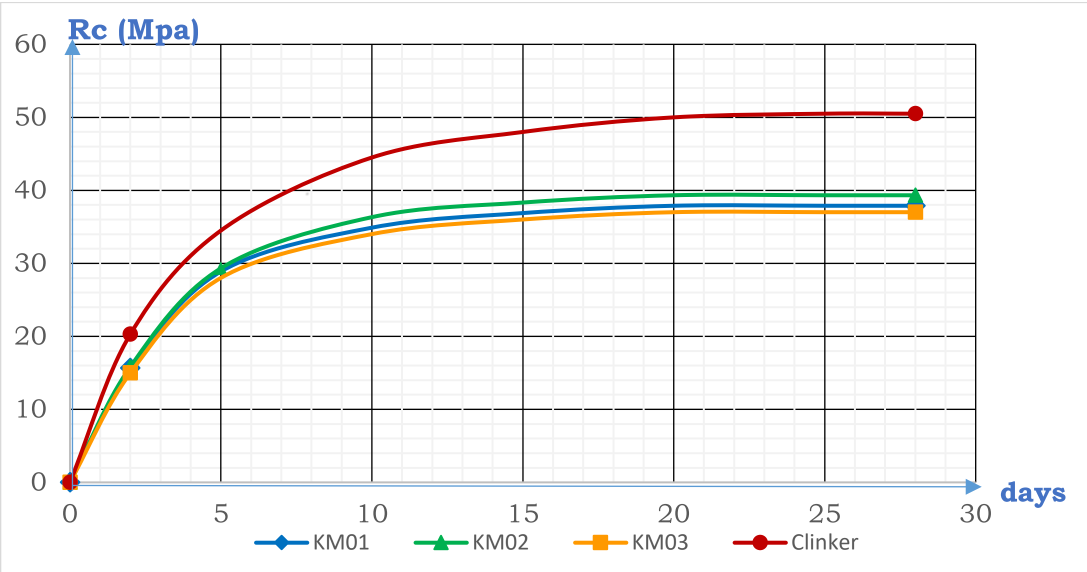

Fig. 22. KM01, KM02, KM03 and clinker resistance curves for 2 and 28 days.

It is clearly shown that the sum of major oxides is above 70 %, their average is 77.58 %, it complies with the standard requirements. The pozzolanicity index (IP) is also greater than 67 %, the average of our three samples is 74 %, which is greater than 67 %, it complies with ASTM C-618 standard requirement. This being said, it can be state that the studied basalt is really pozzolanic and can be used as substitution up to 25% into clinker (75 %) to produce an eco-friendly cement or green cement, low-carbon providing in the same time, the mechanical strength in compliance with the standard.

4. CONCLUSIONS

The Kimbungu basalt at Kasi area has undergone albitization because its weathering index is indeed less than 50 % for all our three samples. It can be clearly observed the presence of several sticks of plagioclase which form an isomorphic series starting with albite and ending with anorthite. As for the LSF of the clinker whose value 90 % shows an alitic clinker, the formulas of Bogue prove it as well. XRD of the clinker used shows that alitic and belitic polymers (orthorhombic α-C_2S and monoclinic β-C_2S) are thermodynamically unstable and highly reactive at room temperature. The sum of the major oxides leading to the pozzolanicity of the rocks is greater than 70 % according to the standard, our average then being 77.58 %. The test carried out on the natural composite cement produced with 75 % industrial clinker and 25 % Kimbungu basalt substitution gave resistances between 14.99 - 15.83 MPa with an average of 15.49MPa at 2 days from the cure. These resistances are between 37.01 - 39.33 MPa, their average is 38.08 MPa at 28 days of the cure. The pozzolanicity indices of our three samples are between 73 % and 76 %. The pozzolanic conditions of this basalt are well verified for the production of an ecological cement qualified as green or low carbon cement. It should be noted above all that this cement retains the cementitious requirements in this area.

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