Physico-Chemical Characterization of Limestones and Sandstones in a Complex Geological Context, Example North-East Constantine: Preliminary Results

Benyamina Mounia, Bouchear Merzoug, Benabbas Chaouki, and Ait Abdelouahab Djaouza

***Abstract*—The geological context of North East Constantine is very complex. This region is characterized by a superposition of several thrust sheets, reflecting a geological and tectonic evolution during Mesozoic and cenozoic.**

In addition, there is a wide variety of sedimentary rocks, such as limestones, sandstones, clays and marls. A physico-chemical characterization of the lithological diversity would be a great contribution to the different users and managers at a time when the region is under major socio-economic development projects.

This study proposes a first phase, a physico-chemical characterization of limestones and sandstones (by optical microscopy, XRD, SEM/EDS and porosimetry), and a reflection on the development of mechanical properties of these materials in a second phase.

Preliminary results of this multidisciplinary study (obtained by various analytical techniques) show good agreement on the existing phases.

***Index Terms*—Limestone, porosity, sandstone, SEM /EDS.**

I. Introduction

There is an abundance of sandstone and carbonate formations in Constantine's region. Limestone and sandstone taken from the north-east of Constantine is distinguished by several features.

The establishment and evolution of these rocks took place in a complex geological context marked by changed paleogeographics, paleoclimatics and pd aleotectonics. These changes have influenced the quality and performance of these materials.

The use of these rocks in development projects, in particular in the construction of roads and highways (embankments, layer shapes, art works.....), requires a better knowledge and a thorough study of the physico-chemical characteristics properties of these rocks in the rough.

II. Experimental Part

A. Extraction and Preparation of Samples (Methodology)

The elaboration samples in this study: white limestone, gray limestone, and sandstone have been collected respectively at Jebel Kellal (mainly carbonated chain link Cenomanian-Turonian age and east-west direction) and at the southern massif of Jebel el Ouahch, their geographical positions are given in Table I.

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TABLE I: Positions of GPS Samples

	Latitude	Longitude
White limestone	36 °25 '27"	6 °38' 46"
Gray limestone	36 °25 '25"	6 °38' 45"
Sandstone	36 °23 '40"	6 °39' 59"

Rocks and sliced sugar using a chainsaw are glued to a glass slide, reduced by a grinder in thin film which is polished to a thickness micrometer will be used for optic observation (Laboratory of Geology and Environment, LGE, Constantine University 1, Algeria).

The same rocks are crushed, and sieved to a particle size ≤ 80 [µm]. This powder will be used for X-ray diffraction analysis (XRD) and scanning electron microscope (SEM) equipped by EDS analysis system.

The micrographs taken by polarizing microscope of prepared thin sections are observed in natural light using an optical microscope (Leitz brand).

The XRD analysis was performed by a diffractometer model PANalytical (Birine Nuclear Center, Ain Ouassera) with Cu anode, λ_{CU} = 1.5406 [A °], on uncompressed powders in order to collect the maximum of the diffraction lines and a better identification of the phases.

The pellets are prepared from powders of particle size ≤ 80 [µm] using a hydraulic press at a pressure of 75 [mPas] for 60 [s], and then dried in open air, for the environmental SEM analysis (ESEM), Quanta 200 model, equipped with a micro-energy dispersive analysis (EDS) (Quan Tax QX2, ROENTEC), (Process Engineering Laboratory, University of Bejaia, Algeria).

B. Optical Microscopy

The optical micrographs of white limestone are shown in Fig. 1 and Fig. 2, and those of gray limestone, in Figs. 3, 4 and 5, and finally Numidian sandstones are shown in Fig. 6.

The white limestone (Fig. 1) are formed by organism's debris, have worn bivalve shell which are perforated by bacteria on a background of $CaCO_3$ carbonates and $CaMg(CO_3)_2$ dolomite. The bivalves fragment are to be in micritic matrix (carbonated mud about 1 to 4 µm), formed by $CaCO_3$ and dolomite $CaMg(CO_3)_2$.

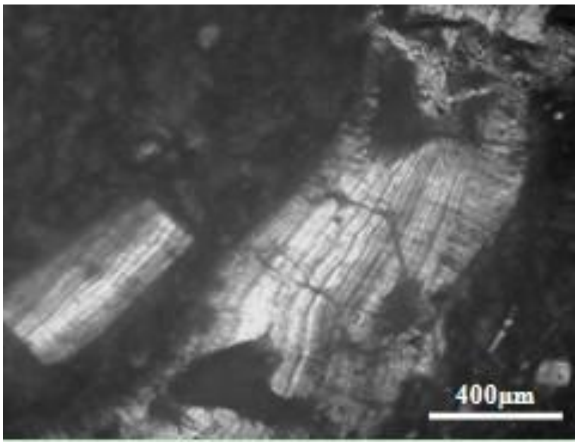

Fig. 1. Thin-section photomicrograph of white limestone, natural light.

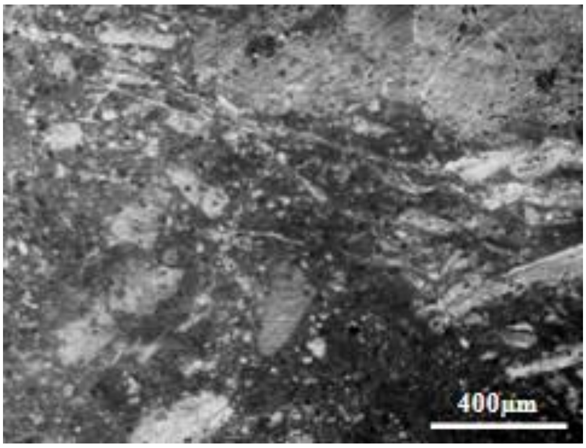

Fig. 2. Thin-section photomicrograph of white limestone, natural light.

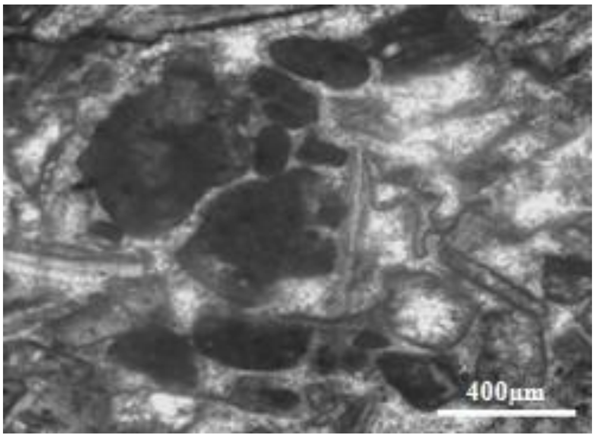

Fig. 3. Thin-section photomicrograph of gray limestone, natural light.

The Fig. 2 shows a finer organism debris and the fracturation porosity with calcitic filling (Fig. 2), [1]-[4].

The gray limestones (Fig. 3) are richer in organism debris, essentielly formed by bivalve fragments, a rare fragments of echinoderms and micritic microbreccias. the porosity type is vacuolar [10]-[11], the fracturation is very abundant in this facies and shows cimenting by calcite or iron oxides (Fig. 4), [1]. Show a significant amount of bivalve shell (marine fossils) and echinoderm, respectively (Fig. 3 and Fig. 4).

The Fig. 5 shows a void filled with zoned calcite, so the calcite can be zoned (Fig. 5), [1].

Sandstone, (Fig. 6) has quartz as a main phase with size grain varies from about 50 to 400 [µm] and can reach average 600 µm, with oxides and clays as phyllosilicates in intergranular contacts, [2]-[3]-[11].

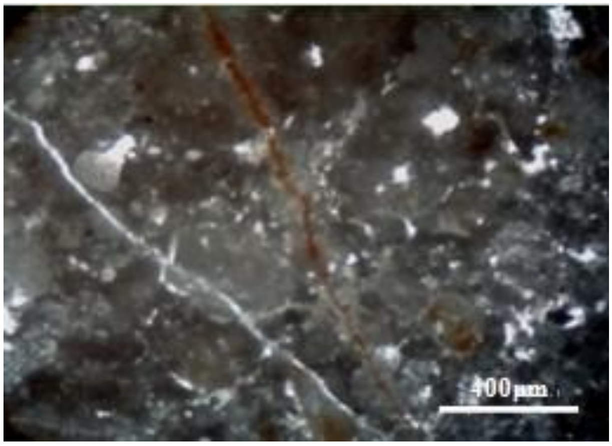

Fig. 4. Thin-section photomicrograph of gray limestone, natural light.

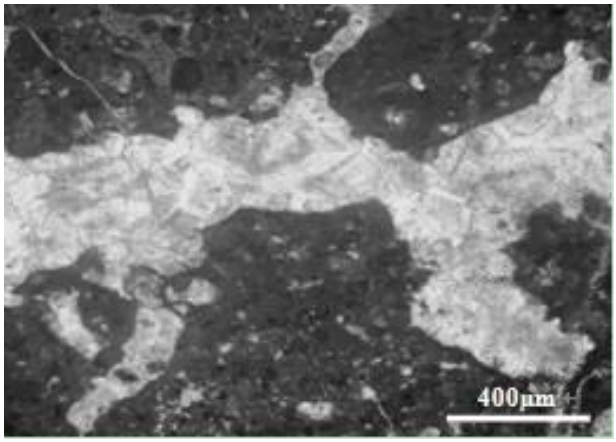

Fig. 5. Thin-section photomicrograph of gray limestone, natural light.

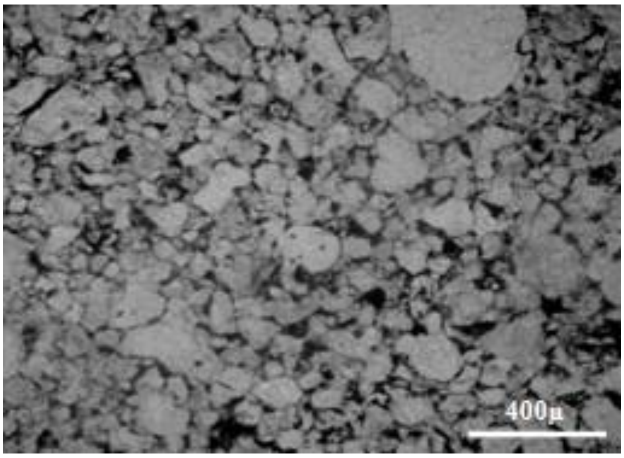

Fig. 6. Thin-section photomicrograph of sandstone, natural light.

C. *X-Ray Diffraction*

X-ray diffraction spectra of the three types of rocks: white limestone (CB), gray limestones (CG) and the Numidian sandstone (GN) are shown in Figs. 7-9 respectively.

The X ray diffraction pattern of sample CB1 and CG2 are presented in Figs. 7, 8: The Dolomite's pics are clearly appear (according to ASTM) on the pattern, they are not indexed, whilst the calcite pics and Aluminium ones with the Dolomite are indexed on the pattern.

One can note from the 3 spectrums:

For X-ray diffraction presented in Fig. 7, one can see presence of 2 important phases in the white limestone: the $CaCO_3$ carbonate and $CaMg(CO_3)_2$ dolomite, the minor elements are showing by the peaks of lower intensities.

For X-ray diffraction of Fig. 8, the gray limestones exhibit the same phases exist in the white ones, but there's a difference in their respective amounts, with other minor elements.

The X-ray diffraction of Fig. 9, shows that the predominant phase is quartz, calcite, minor elements: Mg_2C_3 and Al_20_3.

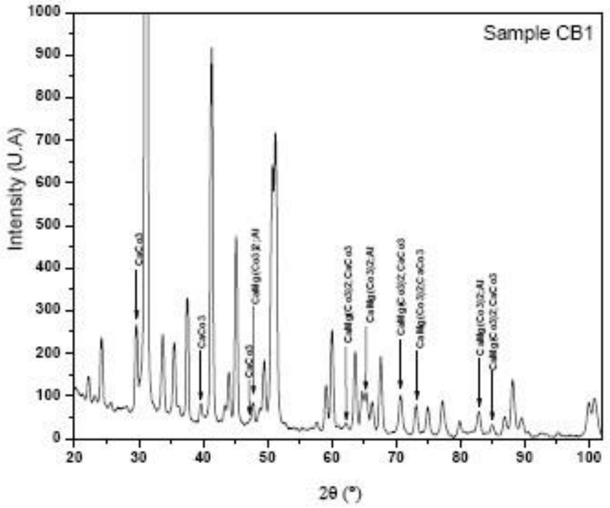

Fig. 7. XRD spectrum of white limestone.

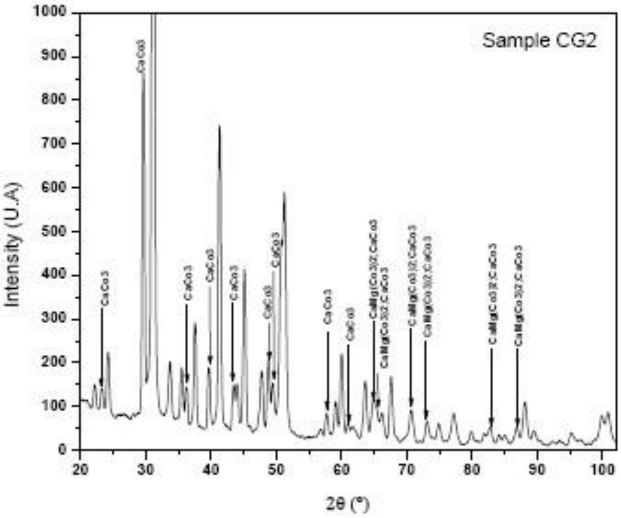

Fig. 8. XRD spectrum of grey limestone.

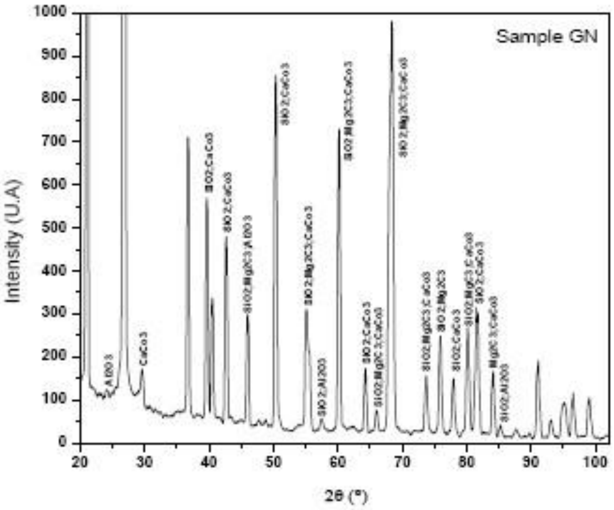

Fig. 9. XRD spectrum of numidian sandstone.

D. Analysis by Scanning Electron Microscopy SEM / EDS

The observation of the morphology using the scanning electron microscope of the white limestone, gray and Numidian sandstone is reported in Figs. 10-12 respectively. The three observed morphologies show that:

One can observed from the SEM morphology of white limestone (Fig. 10), that's the predominant phase in the carbonates with pores filled with organic fragments (fossil's fragment) and an open -porosity in some areas, [5].

For the gray limestones (Fig. 11), they have a more compact morphology, so the gray limestones have less porous then white one and the carbonates represent the main phase with calcite peaks [6].

For the Sandstones (Fig. 12), they have a morphology with angular grains of quartz size around:10 to 30 [µm] , covered with fine particles and the grains of calcite appear clearly.

This analysis of the morphology by SEM is associated by EDS analysis of the elements exist on choised line on area with maximum amount of informations as it is showing in: Fig. 13 and Table II, Fig. 14 and Table III, Fig. 15 and Table IV, respectively, for white limestone, gray limestone and numidian sandstone.

For white limestone (Fig. 13), the presence of Ca, O, C, Mg as major element confirms the importance of carbonate phases, $CaCO_3$ and $CaMg(CO_3)_2$, same remarks have been observed in gray limestones (Fig. 14), except, that the amount of carbonates are not the same.

The sandstones (Fig. 15) have a very important percentage of Si and O, with the oxides and the intergranular phyllosilicates such as: the Montmorillonite $(Na, Ca)_{0,3}(Al, Mg)_2Si_4O_{10}$ or the

Kaolinite $(Al_2Si_2O_5)$ as minor elements observed by optical microscopy.

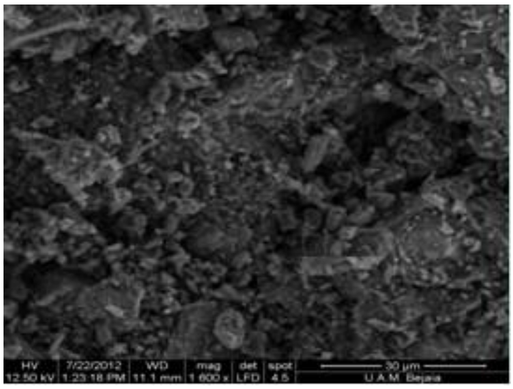

Fig. 10. SEM morphology of white limestone (X 1600)

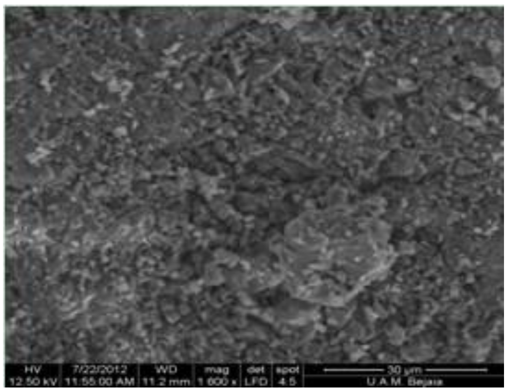

Fig. 11. SEM morphology of gray limestone (X 1600)

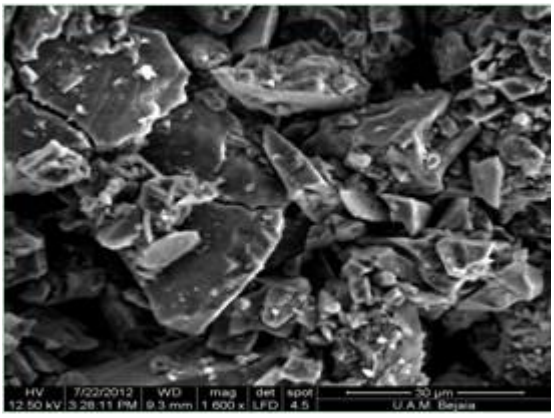

Fig. 12. SEM morphology of sandstone (X 1600)

Tables: Table II, Table III and Table IV represent the percentage of important elements and minor elements that represent the oxides and the intergranular phyllosilicates observed by optical microscopy.

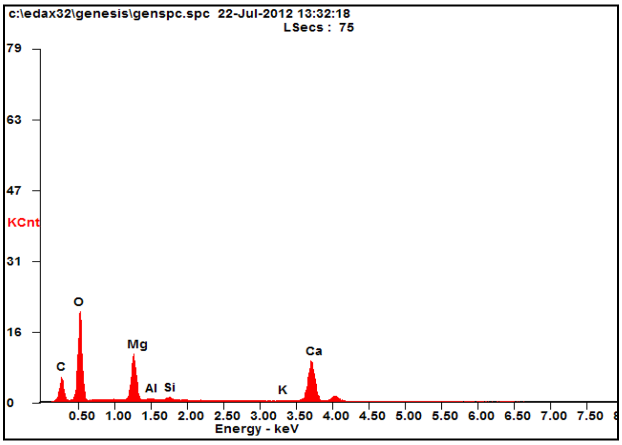

Fig. 13. Energy dispersive microanalysis EDS on the white limestone.

TABLE II: ELEMENTAL ANALYSIS OF WHITE LIMESTONE.

Element	Wt %	At %
C K	17.47	26.78
O K	46.15	53.10
MgK	10.71	08.11
AlK	00.37	00.25
SiK	00.75	00.49
K K	00.30	00.14
CaK	24.25	11.14

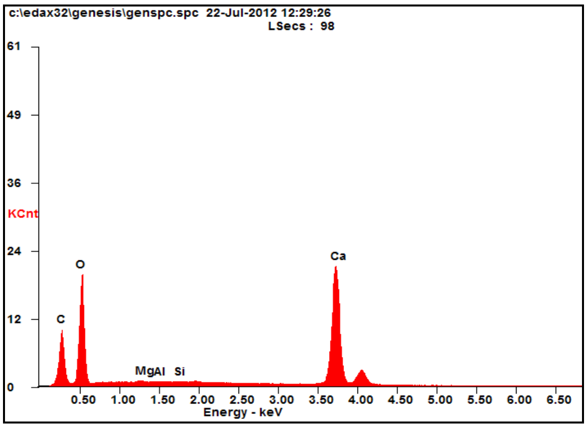

Fig. 14. Energy Dispersive Microanalysis EDS gray limestone

TABLE III: ELEMENTAL ANALYSIS OF GRAY LIMESTONE.

Element	Wt %	At %
C K	17.85	29.09
O K	41.72	51.05
MgK	00.22	00.18
AlK	00.09	00.06
SiK	00.12	00.08
CaK	40.01	19.54

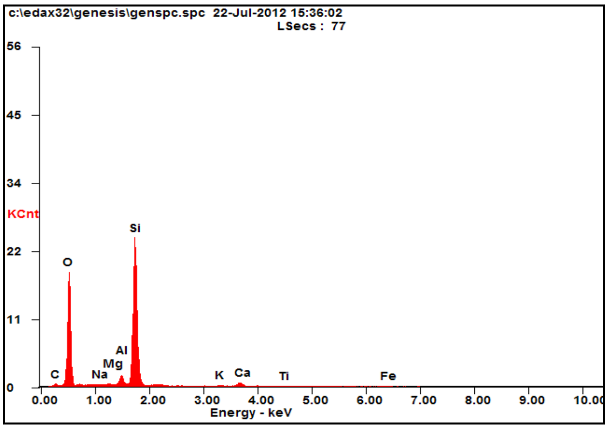

Fig. 15. Energy dispersive microanalysis EDS numidian sandstone.

TABLE IV: ELEMENTAL ANALYSIS OF NUMIDIAN SANDSTONE.

Element	*Wt %*	*At %*
C K	05.12	08.63
O K	45.05	57.02
NaK	00.08	00.07
MgK	00.22	00.18
AlK	02.37	01.78
SiK	41.18	29.69
K K	00.50	00.26
CaK	02.50	01.26
TiK	00.41	00.17
FeK	02.57	00.93

E. Porosimetry

The void ratio N (%) is calculated by hydrostatic weighing method that is based on the Archimeds methode, [7]-[9] and immersed in distilled water.

This method is applied on the three types of rocks and obtained a good approach of porosity ratio (N). The test is repeated 3 times on two samples for each rock.

The empirical formula [7]-[8], Refer to "(1)" , is applied for the calcul of the rate's porosity [%], measures are reported in Table 5.

$$N = [(M_{sat} - M_s)/(M_{sat} - M_{hyd})] \times 100 \qquad (1)$$

with: *Ms* = Mass of the dry rock.
Msat = Saturated mass.
Mhyd = Hydrostatic mass.

It is clear that the white limestones are more porous than the gray limestones, due essentially to the difference in texture (grain size) and the phenomena of dissolution and erosion, this does not prevent the growth of fossil and calcite in the pores or grains.

TABLE V: RATE OF POROSITY.

	white limestone		gray limestone		Numidian sandstone	
Samples	1	2	1	2	1	2
M_s [gr]	28.8155	43.6143	23.9864	30.5848	19.1125	28.6278
M_{sat} [gr]	29.4927	44.6714	24.2194	30.8764	19.4035	29.0271
M_{hyd} [gr]	17.3188	26.4816	15.1377	19.5013	11.4157	17.2556
N %	5.5627	5.8114	2.5655	2.5634	3.6430	3.3920
N_{moy}%	5.6870		2.5644		3.5175	
T_{Vacuum}[h]	1	1	1	1	1	1
T imbibition[h]	48	48	48	48	48	48

The sandstones represent a midium degree of porosity, because on one hand the size of the quartz grains is important and in the other hand, the intergranular voids are filled with oxide and phyllosilicates.

III. CONCLUSION

The obtained results by different analyses: optical, XRD, SEM / EDS and porosimetry, are in a good agreement and confirm the highlight of existing of the phases in our samples.

The phases observed by optical microscopy of three samples are found by X-ray diffraction, the SEM morphology coupled with EDS analysis shows what has been observed by previous techniques, confirms the existence of element's proportions.

A reasonable degree of porosity in the limestones confirm the work done by Katia BESNARD [6], it
is mainly due to the difference in the texture of the specimens.

One note that the degree of porosity in the gray limestone is lower than that of white limestone, which is observed before in the optical micrographs where the compactness in gray limestone is higher than that in whites, so more porous.

These preliminary analyses will be followed by the study of :

The effect of temperature on the physico-mechanical behaviour of these samples,

The improvement by additions of physico-mechanical properties of these elements, because there is a problem of economic overhead, when their using in the raw state, so there is a good normalization (American standard, standard AFNOR French and standard African which it's the lowest quality and cost).

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