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# Geological, petrographic and mineralogical study of the Bentonitic formations of Hammam Boughrara (North-West Algeria)

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**Key words:** Bentonite - Mineralogy - Geochemistry - Perlite - Miocene - NW Algeria.

## Abstract

Algeria's "bentonite" clay deposits are located in the north-west and have significant exploitable reserves. They are mainly found in the regions of Maghnia (Tlemcen) and M'zila (Mostaganem). The objective of this work is to undertake a geological, petrographic, mineralogical and geochemical study of the clay formations that characterize the region of Maghnia as they present specific properties for various applications. The study area, located north of the plain of Maghnia, is between two "atlasic" massifs; the mountains of Traras and Fillaoussène in the North and the mountains of Tlemcen in the South. From a geological point of view, the region consists mainly of Miocene and Pliocene formations. The petrographic examination indicates a very clear beginning of bentonitization process which develops from the pearlitic facies. Similarly, pyroclastites, pyroclastic tuffs and rhyolites are more or less vulnerable to bentonitization because of their microlithic matrix, clayey. Further XRD examination of the previous formations revealed that smectite is the main mineral phase, as well as kaolinite and illite with varying proportions. Non-clay minerals are also identified, such as quartz, cristobalite, plagioclases, potassium feldspars, dolomite and magnetite. Finally, results obtained from additional mineralogical analyzes (saturations with LiCl, KCl, and FTIR examination) revealed that the smectite is also of dioctahedral type and it is represented by montmorillonite.

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## Introduction

The geological characteristics of mineral resources in bentonitic clays in the world are mainly concentrated in areas related to strong magmatic activity, types of volcano-sedimentary deposits, altered and hydrothermal residues resulting from volcanic activity (El Miz *et al.*, 2017).

The bentonite clay deposit of Hammam Boughrara, is located southeast of the town of Maghnia with which it is connected by an asphalt road, the nearest locality is the village of Hammam Boughrara which is 4km east of the deposit. The estimated deposits of the Hammam Boughrara district in the Maghnia region total 11 million tonnes including 3.14 million tonnes for the Roussel site. These bentonite clays have been prospected to ensure the supply of raw materials for the Bental factory in Maghnia, whose annual production is 200 thousand tons per year. Economically, bentonite is exploited for its various applications such as drilling mud, cosmetics, bleaching earths, drugs, paints and the field of civil engineering (Churchman *et al.*, 2006).

Characterization, estimation and valorization of bentonite clay deposits from Hammam Boughrara in Maghnia region (Tlemcen, NW Algeria) are crucial and essentially needed. Especially as the dominant clay minerals of these natural resources are smectites, such as montmorillonite, beidellite, saponite, and hectorite (Grimm, 1968). In addition, important information from these deposit are still poorly constrained. In this paper we attempt a better understanding and to eventually shed light on the lithological, mineralogical and geochemical properties of the NW Algeria's "bentonite" clay deposits.

## Materials and methods

### Materials

*Geological characterization*

The study area is located in the eastern Tafna Valley, which is part of the Tafna Basin, as well as part of the Oranie in NW Algeria (Dakiche 2005, Yebdri *et al.*, 2005). This area, located near the village of Hammam Boughrara is limited to the North and West by the mountains of Fillaoussene and to the south by the plain of Maghnia and the mountains of Tlemcen (Fig. 01). The magmatic formations are represented by rhyolites, perlites, cinerites of the Miocene age and some basaltic dykes of plio-quaternary age (Fig. 02). The deposit is exploited through several quarries, the most important of which are Roussel (RS) and Dar Embarek (DM) (SIDAM, 1994). The bentonite deposit of Hammam Boughrara (HB) is very complex and shows vertical and lateral heterogeneity (Fig. 03 for the Roussel site (RS), Fig. 04 for the Dar Embarek site (DM)).It is located in the volcano-sedimentary base of the Miocene terminal phase.

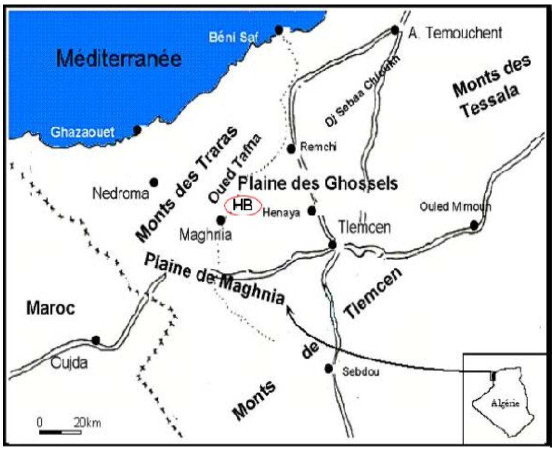



**Fig. 1.** Geographical location of the study area. HB: Hammam Boughrara.

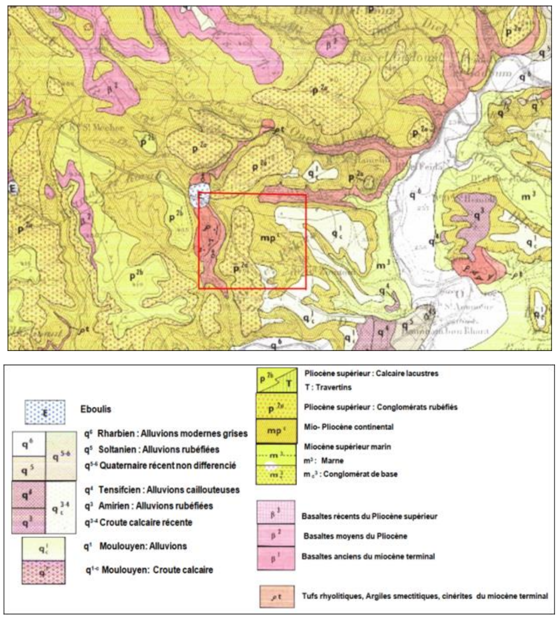



**Fig. 2.** Geological map of the Hammam Boughrara region (from the geological map of Nedrouma).

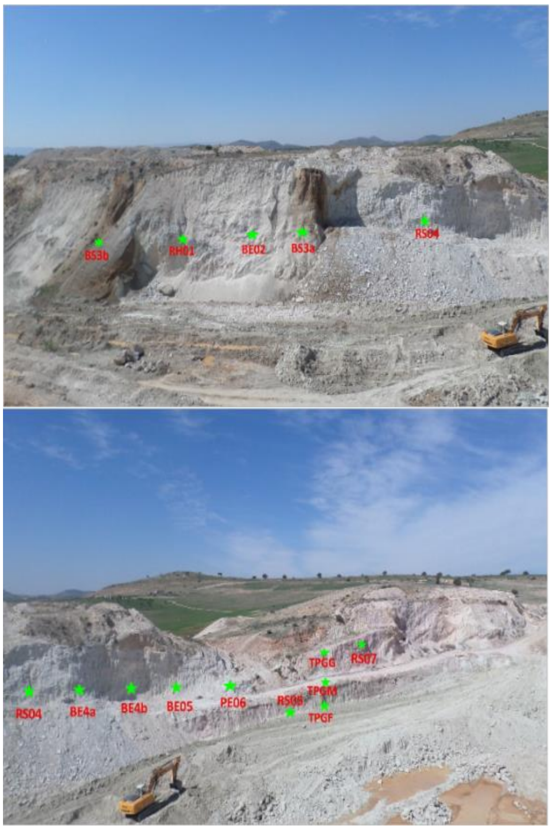



**Fig. 3.** Aspect of the Outcrop of the Roussel site with the sampling plan.

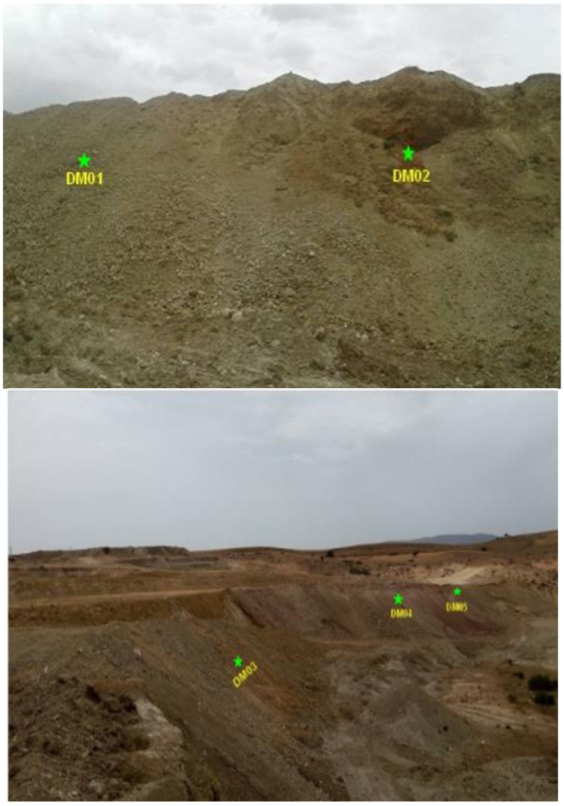



**Fig. 4.** Aspect of the outcrop of the Dar Embarek site with the sampling plan.

**Methods**

Thin-sections were made in the Department of Geology at Badji Mokhtar University in Annaba. The petrographic description was made using a polarizing microscope of "LEICA DMLP" type, installed with a camera of the same type, at the Department of Geology in the University of Liege and finalized at the Department of Geology at Badji Mokhtar University.

Mineralogical analyzes by "XRD" were made at the laboratory of Clays, Geochemistry and Sedimentary Environments (AGEs) at the University of Liège, using a diffractometer type "Brucker D8-Advance", using the $K\alpha 1$ radiation of copper ($\lambda$ = 1.5418A°). The acceleration voltage is 40Kv, the intensity is 30 mA, the measuring speed of the goniometer is 0.6s / step, rotational speed 2°2θ / min, inducing an analysis time of several minutes to cover a 2θ angular range from 2° to 45°, depending on the preparation:

- Disoriented powders: Powders: 2-45°2θ (without rotation).
- Oriented clay mounts: Normal spectrum "N", 2-30°2θ

Solvated Spectrum with ethylen glycol "EG", 2-23°2θ

Spectrum heated to 500°C. "CH", 2-15°2θ.

The estimation of minerals percentage is obtained by using the "Peak-height Ratio" method whose vertical distance between the apex of each characteristic peak and the baseline is measured and multiplied by a corrective factor, such as report by Cook and *et al.*, (1975) and modified by Boski and *et al.,* (1998). The semi-quantitative estimation of clay content was obtained by multiplying the intensity of specific reflections, generally measured on the solvated spectrum (Fagel and Boës, 2008, Fagel and *et al.,* 2007) by a corrective factor (Biscay, 1965). In order to differentiate the nature of smectites and to identify the relative abundance of montmorillonite and/or beidellite, oriented clay-aggregate mounts were prepared according to the saturation procedure with lithium chloride (LiCl2). This method is called "Hofmann and Klemen test", whose principle is to eliminate octahedric charges by lithium exchange in octahedral position (Holtzapffel, 1985); while the genetic origin of smectites (transformed or neoformed) was identified by K-saturation with potassium chloride (KCl) (Thorez, 1998).

Absorption spectra "FTIR" were realized on the clay fraction (<63µm) using a "Nicolet NEXUS" spectrometer at the laboratory of mineralogy and crystallochemistry at the Department of Geology in Liège University. The technique known as "DRIFTS" (Diffuse Reflectance Infrared Fourier Transform Spectroscopy) has been used in a field wave between $4000cm^{-1}$ and $400cm^{-1}$ with a resolution of $1cm^{-1}$.

Chemical analyzes (major elements) of samples collected from both sites (RS and DM) have been enquired using an "ARL" spectrometer of the "Perform X 9400" type in the chemistry laboratory at Liège University, according to a procedure that begins with a very fine grinding of a quantity of 1g then mixing with lithium tetra-borate in order to obtain pearls for geochemical analysis by XRF.

Granulometric measurements were carried out using a "Mastersizer 2000" laser granulometer, Malvern type, in the structural inorganic chemistry laboratory (LCIS) at the University of Liège. The data is treated by the software "Mastersizer 2000 v. 5.0" and the results obtained are given in the form of frequency curves (% volumic) according to the particle size. In order to characterize the clays according to their mechanical properties, it is evident to evaluate the Atterberg limits. Indeed, the liquidity limit (WL) is determined by the apparatus of Casagrande, while the plasticity limit (WP) is manually determined by the roll method.

## Results and discussion

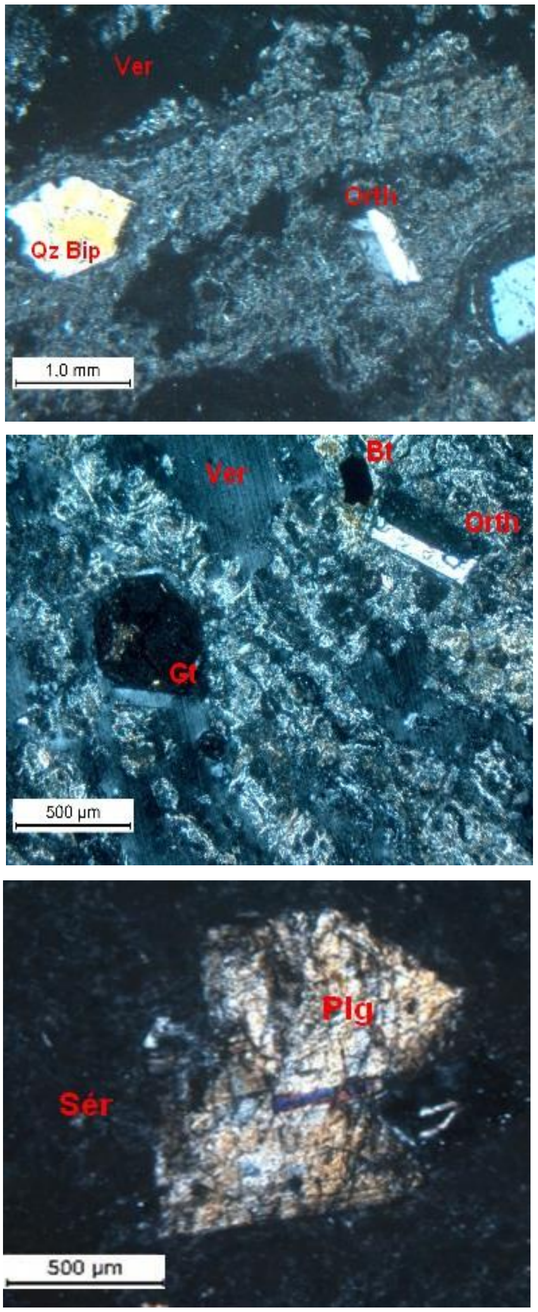
*Petrographic description*

The examination of thin-section under the polarizing microscope shows that the studied facies consist of:

a- rhyolite: quartz, plagioclase (oligoclase) little altered, biotite, volcanic glass, garnet and sericite.

b- b- perlite: quartz, highly altered plagioclase, orthoclase and biotite.

c-the bentonititic formation: quartz, highly altered plagioclase, altered orthoses, biotite, volcanic glass, garnet and sericite (Fig 05). The rhyolitic and pearlitic facies are the product of an explosive type in the volcanic eruption, usually deposited in a marine environment. They are formed by a volcanic glass dust more or less rich in feldspar and biotite and which favor the bentonitization in the area of Hammam Boughrara. This study shows a very clear beginning of bentonitization which develops mainly from pearlitic facies. Similarly, rhyolites are vulnerable to bentonitization by their microlitic matrix wich is clayey.



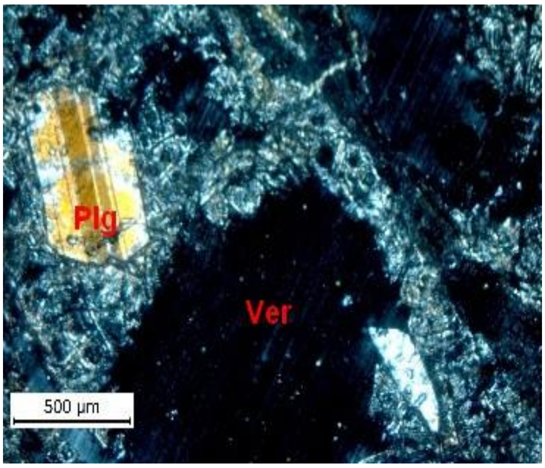



**Fig. 5.** Microscopic aspects of the bentonite facies of Hammam Boughrara.

*Mineralogical characterization*

*Total rock*

The mineralogical composition of the bentonite clay formations of Hammam Boughrara is as follows: The clay minerals are mainly smectite for both sites (Figs 06, 07 and 08). The non-clay minerals are composed of quartz, cristobalite, plagioclase, K feldspar, biotite, dolomite and magnetite.

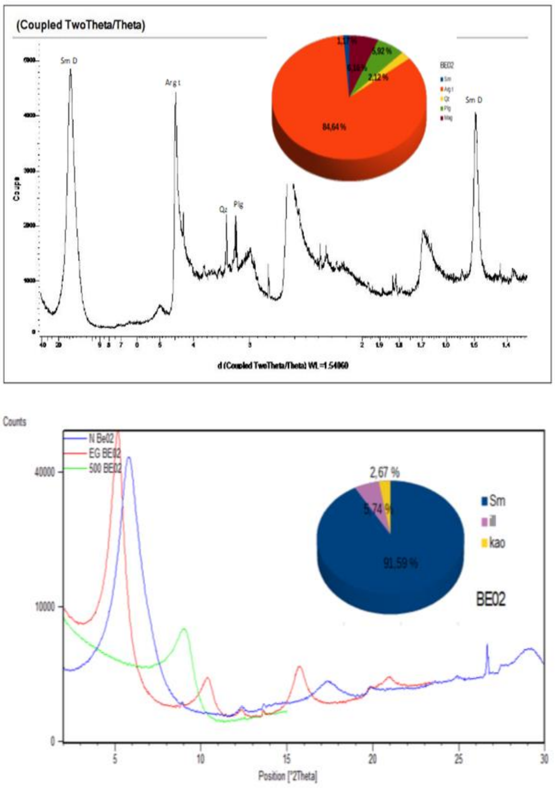



**Fig. 6.** Diffractograms of the BE02 sample (a- Whole rock; b-clay fraction).

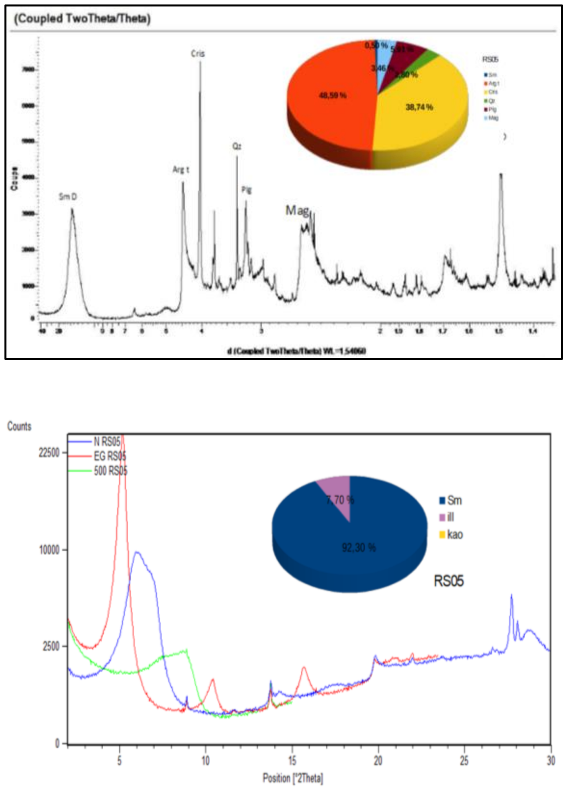



**Fig. 7.** Diffractograms of the RS05 sample (a- Whole rock; b- clay fraction).

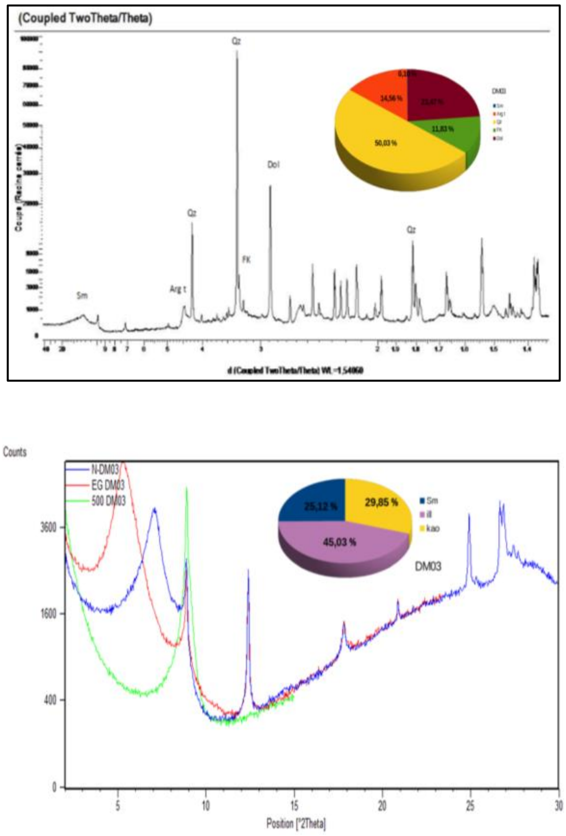



**Fig. 8.** Diffractograms of the sample DM03 (a- Whole rock; b- clay fraction).

*Clay fraction*

Bentonite clays show mineralogical similarities in both sites; for RS, the clay mineral assemblage is characterized by the dominance of smectite, illite (5 to 9%); kaolinite is present in a very small amount (Fig. 06-b- and Fig. 07-b-). For DM, predominance of illite and kaolinite is evidenced, while the smectite is only with minor quantity (Fig. 08-b).

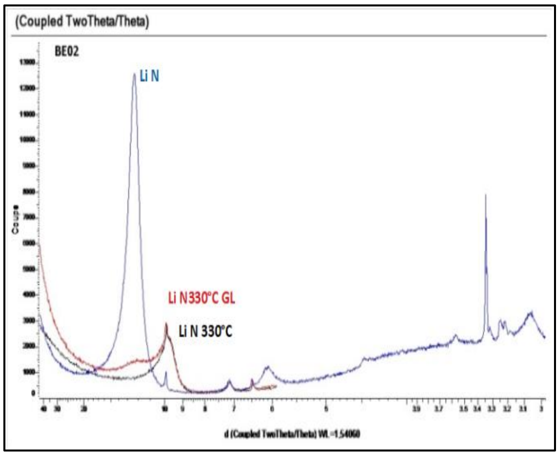
*Smectites*

a- The abundance of montmorillonite and beidellite was estimated by comparing the peak intensities to 10 A° on the three Li-saturated X-ray diagrams (Thorez, 1998) as follows:

(10-14Sm) Al = ILi300 - ILiN; montmorillonite = ILi300G1-ILiN; beidellite = Ili 300- Ili 300Gl.

The saturation methods with lithium chloride (LiCl) and the presence of the reflection (060) show that smectites of "HB" are typical of dioctahedral smectites, where the main component is montmorillonite (Fig. 09), the abundance of which depends on the location of the mine site.

b- The presence and relative abundance of neoformed smectites were estimated by comparing the peak intensities to 10 A° on X-ray diagrams saturated with K (K-N, K-110, K-110EG), as follows: neoformed (10 -14Sm) Al = IK110-IK110EG; transformed (10 -14Sm) Al if IK110 = IK110EG (Fagel and *et al.,* 2003). Potassium chloride (KCl) saturation methods indicate that the smectites are fully neoformed (Fig. 10); they crystallized *in situ* from the available ions present in the solutions, by the phenomenon of authigenesis.



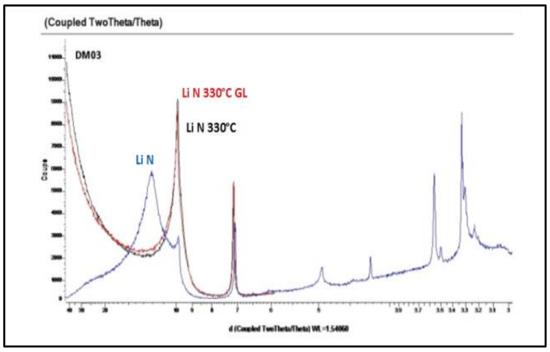




**Fig. 9.** Diffractograms of samples BE02 and DM03 after saturation with LiCl.

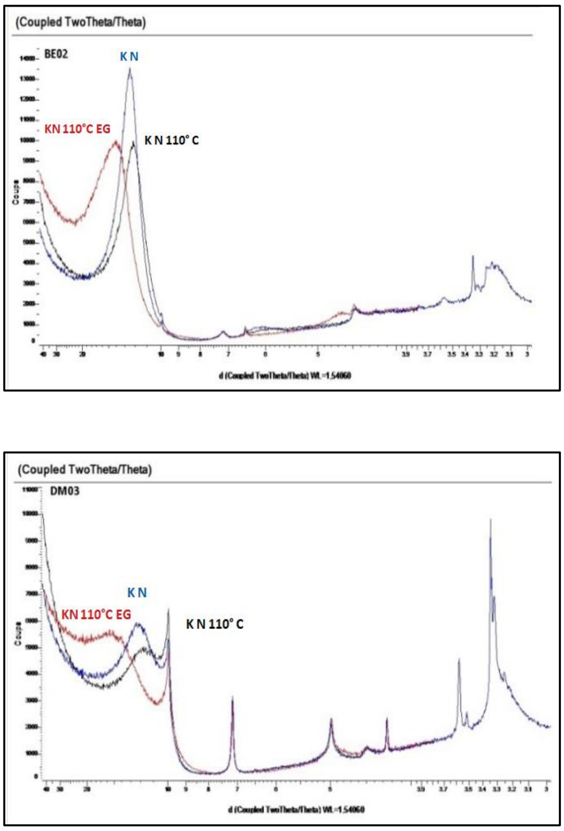



**Fig. 10.** Diffractograms of samples BE02 and DM03 after saturation with KCl.

*FTIR spectroscopy analyzes*

The "FTIR" data from the Hammam Boughrara deposit with the corresponding values of the different sites followed by an interpretation of each value are summarized in table 01. The inter-layer hydrogen bond in the clay is assigned by a characteristic band at 3620cm-1. The latter characterizes montmorillonite and corresponds to the groups stretching OH-coordinated vibrations to the octahedral layer at Al + Mg (3621cm$^{-1}$) (Farmer *et al.*, 1979), (Salerno *et al.*, 2001).

**Table 1.** Summary of FTIR data.

| Absorption band values (cm-1) | | Interpretation of the assignment |
|---|---|---|
| Roussel | Dar-Embarek | |
| 3620 | 3630 | OH vibration stretching |
| 3400 | 3400 | Stretching OH structural hydroxyl groups |
| 1640 | 1640 | H-O-H deformation of water |
| 1040 | 1040 | Si-O stretch of silica and quartz |
| 920 | | Deformation Al - Al - OH |
| | 800 | Vibrational deformation of Si-O-Al, Si-O-Mg and Si-O-Fe bonds |
| | 700 | Al-O-Si vibrations out of plan |
| 520 | 540 | Deformation Al - O - Si |
| 480 | 480 | Deformation Si - O - Si |

FTIR spectra counting, confirm the result of lithium chloride saturation tests and the identification of d (060) reflections on XRD diffractograms. These results show that the IR spectra of representative clay fractions of bentonites at both sites are typical for dioctahedral smectites (Figs 11 and 12). They allow so, to confirm the montmorillonitic character of the bentonite clay of Hammam Boughrara.

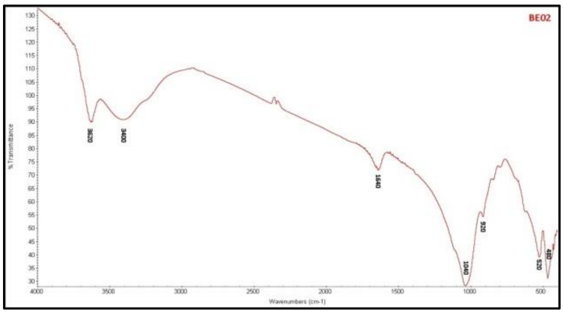



**Fig. 11.** "FTIR" spectrum of the BE02 sample from the Roussel site.

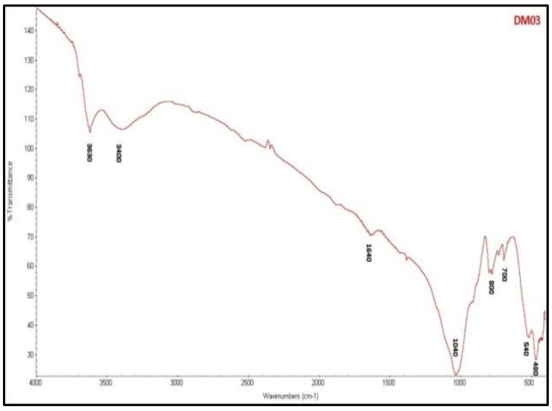



**Fig. 12.** "FTIR" spectrum of the DM03 sample from the Dar Embarek site.

*Physicochemical characterization*

*Chemical analysis*

The results of the chemical analysis by XRF (% wt) are reported in Table 2. The behavior of chemical elements during weathering is complex because of their redistribution and the type of the altered minerals (Düzgören, Aydin *et al.*, 2002).

The most significant variations in major elements controlling bentonitization at Hammam Boughrara are silicon, aluminum, calcium and magnesium. Indeed, there is a regular evolution in these elements, from the unweathered facies to the bentonitized facies.

The two diagrams used to better identify the variations in the chemical composition of the materials made it possible to accurately identify and separate the bentonitized facies of the unweathered facies.

**Table 2.** Chemical composition of magmatic and bentonite formations.

| Sample N ° | RH 01 | BE 02 | BE4A | RS07 | PE04 | PE06 | DM03 |
|---|---|---|---|---|---|---|---|
| SiO2 | 74,01 | 53,49 | 53,1 | 52,68 | 57,22 | 66,13 | 69,55 |
| TiO2 | 0,01 | 0,01 | 0,02 | 0,02 | 0,02 | 0,03 | 0,44 |
| Al2O3 | 13,96 | 17,98 | 18,03 | 19,51 | 18,26 | 15,06 | 5,47 |
| Fe2O3 | 0,79 | 1,22 | 1,58 | 1,49 | 1,33 | 1,14 | 1,84 |
| MgO | 0,18 | 3,99 | 3,87 | 3,31 | 4,24 | 1,27 | 3,07 |
| MnO | 0,09 | 0,03 | 0,08 | 0,06 | 0,03 | 0,1 | 0,01 |
| CaO | 0,44 | 1,1 | 1,07 | 0,7 | 1,13 | 0,54 | 3,59 |
| Na2O | 3,38 | 0,88 | 0,88 | 1,38 | 1,3 | 2,96 | 0,38 |
| K2O | 4,61 | 0,37 | 0,34 | 0,38 | 0,36 | 2,85 | 1,28 |
| P2O5 | 0,06 | 0,05 | 0,04 | 0,04 | 0,04 | 0,05 | 0,07 |
| LOI | 2,42 | 21,35 | 21,46 | 20,62 | 16,34 | 9,64 | 9,44 |
| Somme | 99,95 | 100,47 | 100,47 | 100,19 | 100,27 | 99,77 | 95,14 |

*The $Al_2O_3/SiO_2$ diagram*

This diagram differentiates between bentonitized facies and unweathered facies in the study area (Fig.13). Indeed, all the bentonitized facies have values between 14 and 27% in $Al_2O_3$ and less than 57% in $SiO_2$, whereas the unweathered facies have values lower than 18% in $Al_2O_3$ and values between 57 and 74% in $SiO_2$. So, this diagram shows a regular decrease in $SiO_2$ accompanied by an increase in $Al_2O_3$ and that since the unweathered facies to the bentonitized facies.

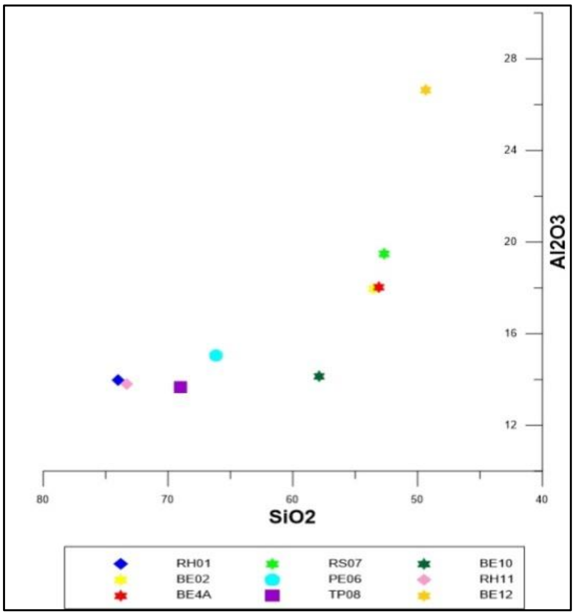



**Fig. 13.** $SiO_2$ / $Al_2O_3$ Diagram of unweathered and Bentonitized Facies of Hammam Boughrara.

*The MgO/CaO diagram*

This diagram allows to differentiate the bentonitized facies of Hammam Boughrara (Fig.14). Indeed, in this diagram, all the bentonitized facies have values between 1.77 and 4% in MgO and between 0.3 and 1.1% in CaO; whereas unweathered facies have values lower than 1.27% in MgO and 0.92% in CaO.

This diagram also shows a sharp increase in MgO then CaO from unweathered facies to bentonitized facies.

The comparison of the chemical analyzes of all the samples made on the perlitic and rhyolitic facies allows to report that they have been subject of an important secondary transformations that probably influence the occurrence of bentonite clay minerals.

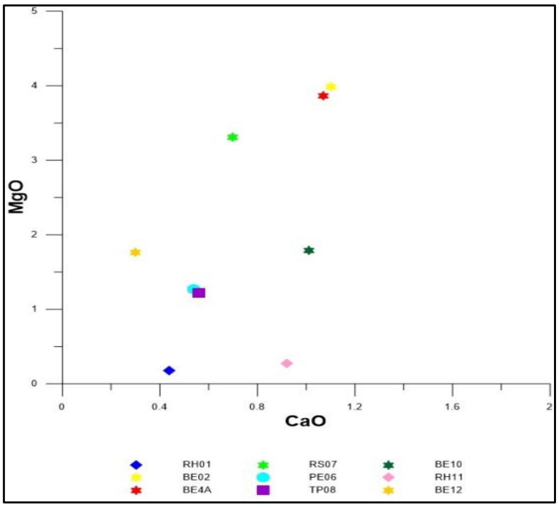



**Fig. 14.** Diagram CaO / MgO of unweathered and Bentonitized Facies of Hammam Boughrara.

*Particle size analysis*

"Laser" granulometric analysis allowed to know the distribution of the fine fraction (<2 µm) in each sample. From these results (Table 03), three grain size fractions were selected (<2 µm, between 2 and 63µm and> 63µm). Given that the size of the clay grains of these materials is essentially in the intermediate fraction between 2 and 63µm (Fig. 15) due to the high quantity of quartz and the presence of unaltered feldspars, which influences directly on the amount of the fine fraction (<2µm) in all the samples.

**Table 3.** Granulometric distribution of the different fractions (in%).

| | Sand | Silt | Clay |
|---|---|---|---|
| Samples | Ø > 63 µm | 2µm < Ø < 63 µm | Ø < 2 µm |
| BE02 | 7,679 | 83,713 | 8,608 |
| RS05 | 4,410 | 88,733 | 6,858 |
| DM03 | 31,803 | 63,773 | 4,424 |

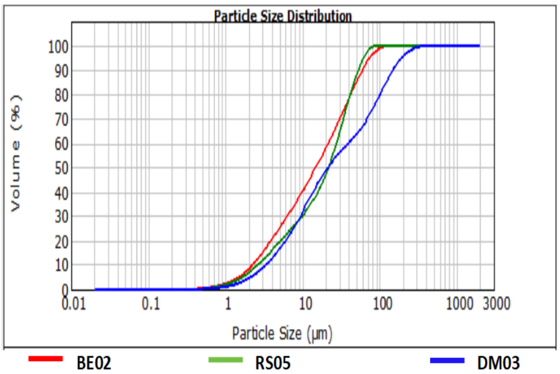



**Fig. 15.** Granulometric curves of the bentonitic clays.

*The limits of Atterberg*

The results are reported in Table 04; the plasticity index "Ip", which represents the difference between the liquidity limit and the plasticity limit, allows to divide the samples into three groups (Fig. 16):

- The first group concerns the treated bentonite formations (BE10 - BN -BR), which have a very high plasticity index (92-109), they are classified as very plastic bentonites.
- The second group, which concerns the bentonite clays of Roussel site (except the two samples BE05 and RS05 which have a slightly lower plasticity), they are classified as plastic silty clays (55-56).
- The third group contains bentonite BE05 and RS05 from the Roussel site and Dar-Embarek bentonites (DM 01 and DM 03) which have a very low plasticity index (8 - 10) compared to the other two sites, because of the very high percentage of illite and kaolinite in the deposit.

**Table 4.** Values of Atterberg limits.

| | Samples | $W_L$ | $W_P$ | Ip |
|---|---|---|---|---|
| Roussel Bentonite | BE4A | 119,20 | 63,15 | 56,05 |
| | BE4B | 95,60 | 40,60 | 55,00 |
| | BE05 | 45,80 | 30,88 | 14,92 |
| | RS05 | 47,75 | 33,53 | 14,22 |
| treated Bentonite | BN | 152,63 | 60,40 | 92,23 |
| | BR | 160,00 | 50,30 | 109,70 |
| Dar Embarek Bentonite | DM01 | 29,00 | 20,26 | 8,74 |
| | DM03 | 34,90 | 24,06 | 10,84 |

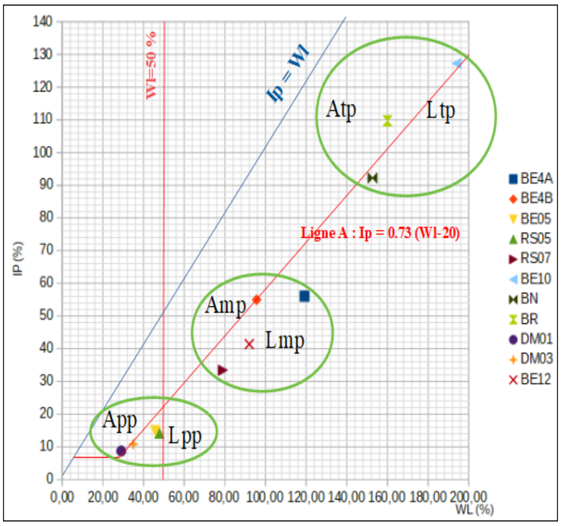



**Fig. 16.** Classification of bentonite clays of HB according to the Casagrande diagram.

## Conclusion

This study was carried out in order to provide necessary basic data on Hammam Boughrara bentonites as well as to evaluate their industrial use. Combination of optical microscopy, X-ray diffraction (XRD), infrared spectroscopy (FTIR) and chemical analysis (XRF) have shown that the main clay mineral phase of the studied formations is smectite.

It is associated to a lesser degree with kaolinite and illite. Non-clay minerals such as quartz, cristobalite, plagioclase of the albite and oligoclase type, potassium feldspars, dolomite and magnetite are also identified in varying amounts. Key information on the relationships between bentonite facies and nearby magmatic facies was obtained from the petrographic study as well as physicochemical characterization on samples taken from the main sites of the deposit.

The mineralogical and chemical compositions of the samples studied are typical of montmorillonite type dioctahedral smectite clay minerals. Finally, the preliminary obtained results can serve as a basis for any study of the Algeria's "bentonite" clay deposits, especially as they can be applied in numerous fields, such as drilling mud, cosmetics, bleaching earths, drugs, paints and the field of civil engineering.

## Abbreviations

Qz: quartz - Qz bip: bipyramid quartz - - Ver: volcanic glass - Plg: plagioclase; Olig: oligoclase-Orth: orthose ; Sér: sericite - - Bt : biotite -Gt: garnet - HB: Hammam Boughrara - DM: Dar-Embarek site - RS: Roussel site.

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