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Physicochemical And Geotechnical Characterization And Application Of Berriasian Clays Of Djebel Oust (North Of Tunisia)

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ABSTRACT

The study of Djebel Oust Berriasian clays shows an abundance of illite (66%), the presence of kaolinite (16%), a fair amount of interstratified illite/smectite (10%) and chlorite (8%). The geochemical analysis proves that these clays are siliceous, Ca-rich and aluminous. The iron rate is 4.7%. The geotechnical study indicates that these clays are little to fairly plastic, and adapt fast to drying. Also, the study shows that firing shrinkage is little. In the light of these results, these clays could be used in manufacturing of the ceramic pieces.

KEYWORDS

Berriasian;
Clays;
Mineralogy;
Geochemistry;
Geotechnical;
Ceramic tests.

INTRODUCTION

In this study, we are focusing on the physical-chemical and geotechnical characterization of Berriasian clays which occupy east, west and northern west sides of Djebel Oust. The latter is situated 35 km West-South of Tunis and 20 km West North of Zaghouan. The series of the lower-Cretaceous of J. Oust and namely those of Berriasian clays have risen a great amount of discussion and interest among geologists since a long time. They were the subject of various studies: age, depositional environment and tectonic settings. Indeed, they studied the geological series of northern Tunisia and in particular those of J.Oust[1], tried to explain Jurassic extrusions and their relationships to the lower-Cretaceous series[2-3]. They produced the geological map of Bir M'echerga to the 1/50000 which covers all the series of J.Oust[4-5] and carried out a biostratigraphic study of the series of the lower-Cretaceous of J.Oust [6-8]. They established a correlation between the series of the lower-Cretaceous of the Eastern Atlas while referring to biostratigraphic data[9] and established a detailed zonation of the series of the lower-Cretaceous by

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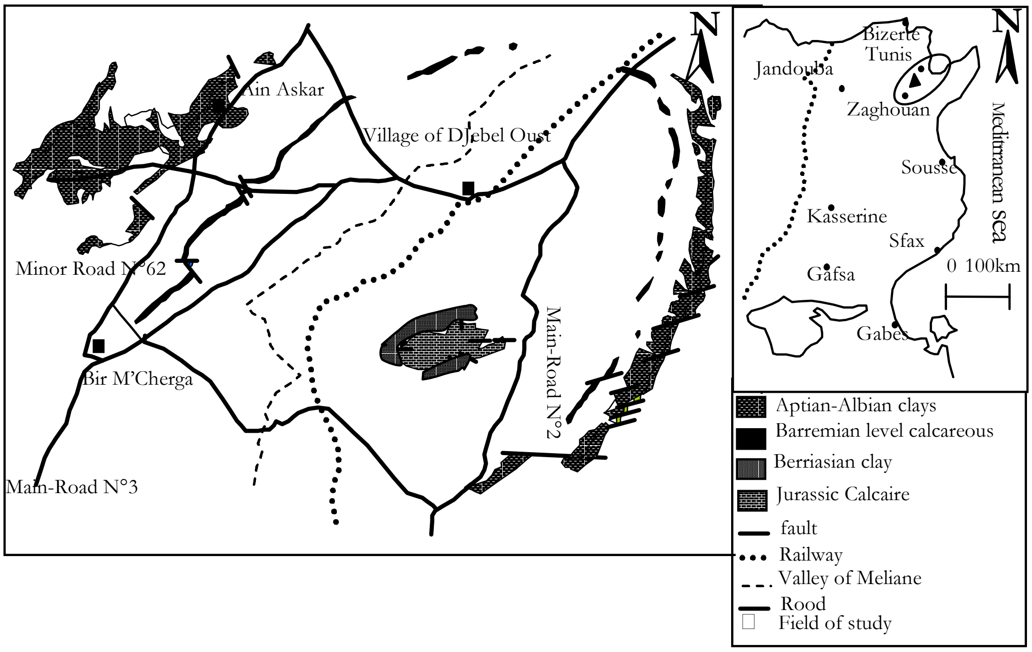

Figure 1: The geographic situation of studied sector.Extacted from the simplified geological map of Bir M'Cherga N°28

improving the former subdivisions[10].

MATERIALS AND METHODS

The X-ray diffractometer used in this study is a Philips X' Pert Pro system. The wave length (λ) is 1.5418Å (Cu Kα). The generator of the X-Ray diffractometer operates at 40Kv and 40 mA. The cation exchange capacity is measured with standard acetate ammonium procedure[11]. The major element compositions are determined by atomic absorption. The differential thermal analysis coupled with thermogravimetry (DTA/TG) was obtained with a SETRAM type 124. The expansion and firing shrinkage were measured using dilatometer Adamel Lhomargy type BI. The samples were heated from ambient temperature to 900°C. The rate of temperature rise was 10°/min. The parameters of plasticity (liquid limit (L_L), plastic limit (P_L) and index plasticity (I_P), $I_P = L_L - P_L$) were determined in accordance with the French Standard NF P 94-051. Drying behaviour was studied using barelatograph D124. The drying and firing shrinkage were evaluated using the formulas: $|(l_g - l_d) / l_g| \times 100$ and $|(l_f - l_d) / l_d| \times 100$, respectively. Where l_g, l_d and l_f are the measured length of green, dried and fired samples, respectively.

The clays intended for the industrial tests were dried at 110°C, and later finely crushed. The preparation of ceramic pieces necessitates the mixture of two kilograms of clay with 7 % of water. For each mixture, we make 20 earthenware squares, each one had the following dimensions: 100 mm ∗ 50 mm ∗ 7 mm, and was pressed at 250 bars. Initially, we dried them at ambient temperature during four hours. Then, we dried them at 30°C, after that at 50°C and finally at 110 °C until we have the constant weight. The firing was carried out in an electric furnace at different temperatures (900, 950, 1000 and 1050°C) with a rise of 300°C per hour. For each mixture, we carried out the firing of five ceramic pieces[12-14].

RESULTS AND DISCUSSION

Clay characterization

1. X-ray diffraction study

X-ray diffraction patterns of the studied Berriasian clays (Figure 1) show that the percentages of the crude sample and the fraction below 2μm do not present a

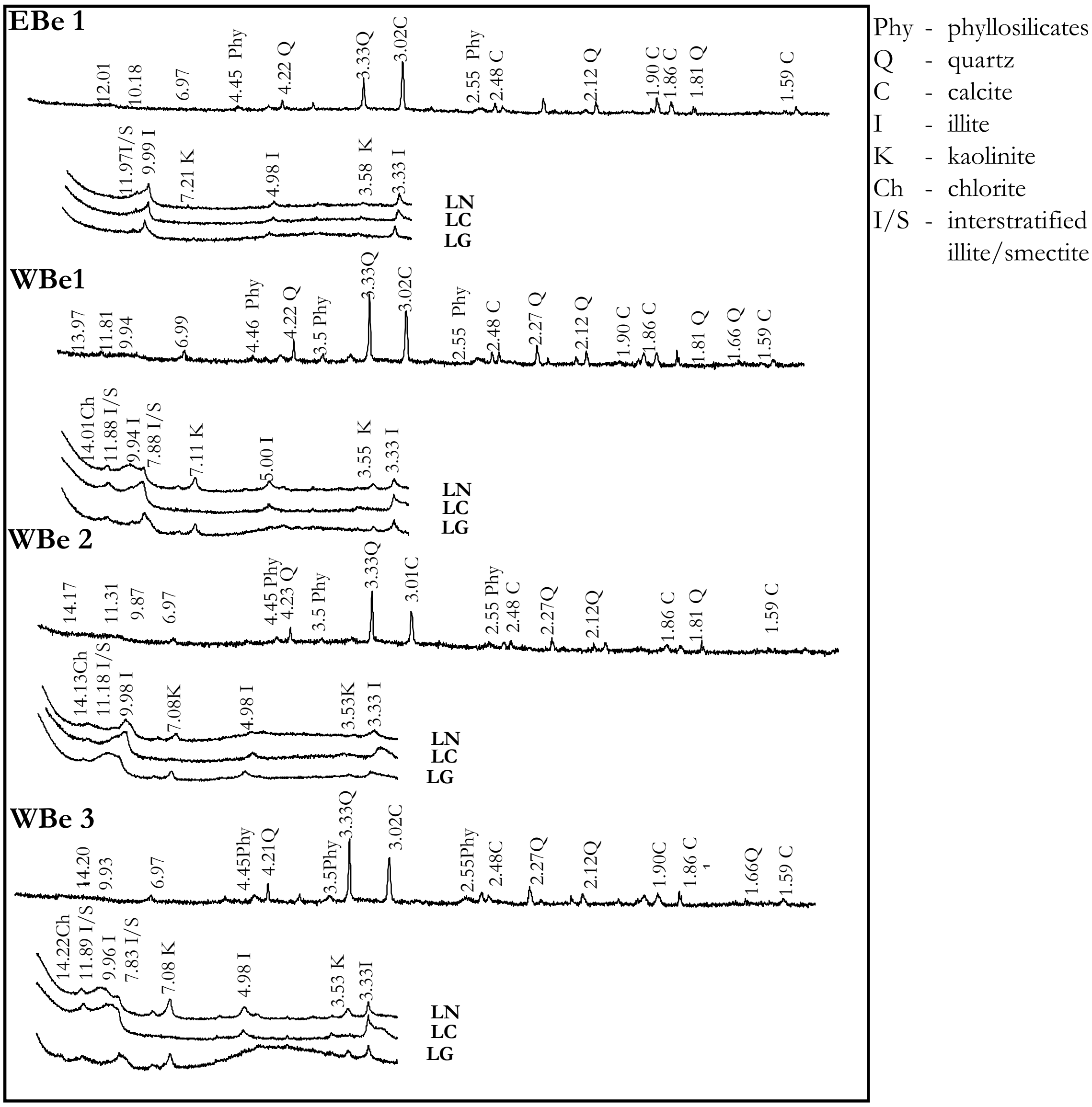

Figure 2: RX Diffractogms of J. Oust Berriasian clays

noticeable change along the geological series of the Berriasian clays. Indeed, quantitative semi calculation of the fraction below 2µm allowed us to show the presence of the proportions in quartz that varied between 12% and 25% and of the calcite which, oscillates between 25% and 42%. The <2µm fraction varies between 43% and 63%. The crude sample is dominated by illite with a percentage oscillating between 57% and 71%. Kaolinite is the second mineral with a rate varying between 11% and 24%. There are similar quantities of chlorite and interstratified illite/smectite, with a percentage of 9 % for each mineral (TABLE 1).

TABLE 1: Mineralogical composition (wt. %)

	Whole sample			<2µm Fraction			
	Qz	**Ca**	**Phy**	**I**	**K**	**I/Sm**	**Ch**
W Be 1	20	33	47	71	15	4	9
W Be 2	25	27	48	61	14	6	18
W Be 3	12	25	63	58	25	7	10
E Be1	15	42	43	69	11	20	0

2. Chemical study

The chemical analysis of J.Oust Berriasian clays shows that they contain alumina varying between 11.27% and 14.03% and proportions of silica alternating between 34.47% and 46.12%. The percent-

TABLE 2: Chemical composition (wt. %)

	E Be 1	*WBe 1*	*WBe 2*	*WBe 3*
SiO_2	34.47	45.15	46.12	43.75
Al_2O_3	12.70	11.27	14.03	12.58
Fe_2O_3	4.10	4.27	5.38	5.19
CaO	20.66	16.49	13.12	15.21
MgO	1.71	0.68	1.02	1.20
Na_2O	2.59	1.60	1.76	1.97
K_2O	3.02	2.56	2.43	2.33
Loss of ignition	20.63	17.84	16.01	17.54

ages of iron vary between 4.10% and 5.19%. The amount of CaO varies between 13.12% and 20.66%. The existence of the zones containing K_2O near to 3.02% (TABLE 2) testifies the presence of illite. This has been already ascertained in the mineralogical analysis.

3. IR-spectroscopy

Figure 3 depicts the results obtained from the infrared spectra of Berriasian clays.

The Al-Al-OH stretching frequency is observed at 920 cm^{-1} and 3650 cm^{-1}. This can be considered as a characteristic of a dioctahedral clay (illite). The intense band at 1110 cm^{-1} and 1035 cm^{-1} are those of the Si-O stretching frequencies. The tetrahedral bending modes are at 570 cm^{-1} and 485 cm^{-1}. The shoulder at 700 cm^{-1} suggest the presence kaolinite. The non-clay impurity which is easily removed by sedimentation is essentially calcite and quartz detected by doublet at 1445 cm^{-1} and 876 cm^{-1} for calcite 803 cm^{-1} for quartz.

4. Cation exchange capacities (CEC)

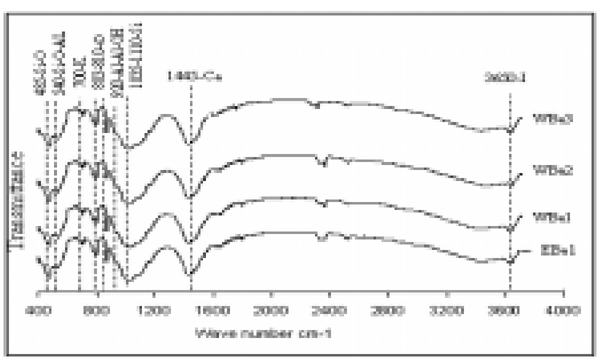

Figure 3: Infrared spectra fo J.Ouse Berriasian samples

In aqueous solution the phyllosilicates can exchange certain ions with others, Thus according to the nature of clay, the interlayer cations can substitute with other compensation cations of load to balance the electric charge of 100 g clay. The results described in TABLE 3 show that the values of the cation exchange capacities are low and does not exceed 12. This is explained by the presence of impurities (carbonates and silica), the abundance of clay minerals formed primarily by illite and kaolinite, and the absence of clay minerals with strong cation exchange capacities such as smectites and vermiculites. This result is in agreement with the X-ray diffraction. The cation exchange capacities the crude samples rose due to the elimination of carbonates.

Ceramic properties

1. Plasticity

The test of plasticity of J.Oust Berriasian clays shows that a the liquid limit varies between 32.33% and 38.50%, the plastic limit between 18.75% and 20.96% whereas the plasticity index oscillates between 13.58% and 17.54% (TABLE 5). These values lead us to conclude that these clay samples are illitic clays little to fairly plastic as referred to results of degree and plasticity index given in TABLE 4 and figure 4.

2. Drying curves

Few Berriasian clay samples selected from J. Oust were submitted to a drying test. The test provides information on the clay aptitude for drying and their capacity to expel or retain water[17] and also to determine the drying parameters (the shaping (total) water, the interposition water, colloidal water and drying shrinkage).

Berriasian Clays need an amount of shaping water varying between 13.40% and 19.07%. .The interposition water lies between 9.35% and 14.45%, whereas the drying shrinkage varies between 2.52% and 3.22%. The obtained results are shown in table

TABLE 3: Cation exchange capacities of J.Oust Berriasian samples

	EBe 1	WBe 1	WBe 2	WBe 3
Rough clay C.E.C. (meq/100g)	4.00	8.00	10.66	10.00
Raw clay C.E.C. (meq/100g)	5.03	9.73	12.69	12.55

6 and diagram 5.

3. Thermal analysis

3.1. Differential thermal and thermogravimetric analysis (DTA/TG)

DTA and TG were merely used as complementary methods with respect to other technique. The DTA curve of J.Oust Berriasian of the crude sample (figure 6), show four endotherms peaks. The first peak at 90 and 99°C corresponds to water hydration. The second peak at 297-301°C illustrates the presence of organic matter. The third peak at 569 and 570°C due to the dehydroxylation of clays. The fourth endothermic peak located at 760 and 817°C corresponds to the decarbonation. The exothermic peak at 931 and 965°C is due to structural change.

TABLE 4: Degree of plasticity according to the plasticity index (wt.%)[15]

Index of plasticity	Degree of plasticity
0 to 5	Not plastic
5 to 15	not very plastic
15 to 40	plastic
> 40	Very plastic

TABLE 5: Plasticity of Berriasian clays (wt. %)

	Liquid limit	Plastic limit	Index of plasticity
EBe1	38.50	20.96	17.54
WBe1	32.53	18.81	13.72
WBe2	32.33	18.75	13.58
WBe3	35.70	18.18	17.52

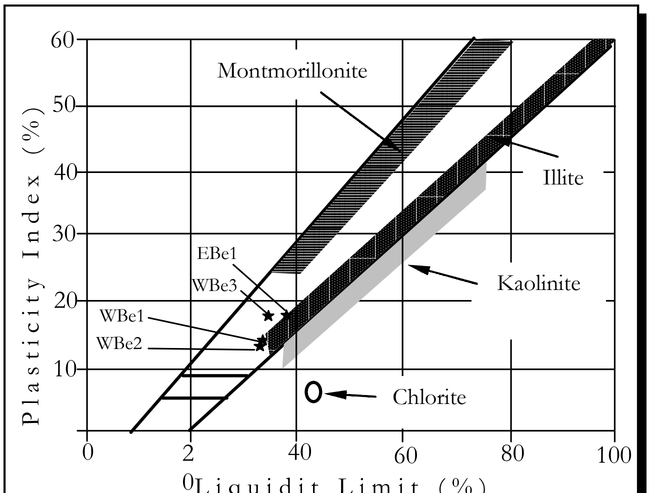

Figure 4: Position of the studied clays on the Holtz and Kovacs diagram[16]

The weight loss of the crude samples given by TABLE 7.

3.2. Dimensional change on firing

The examination of the dilatometry curves of J. Oust Berriasian crude samples clay initially shows a light expansion towards 105°C, due to the loss of the water interposition. The slope of the curves is accentuated towards 573°C, shows the characteristic α-to-β quartz inversion, and the typical clay behaviour[18] characterised by maximum expansion varying between 1.1% and 1.27%. The firing temperature oscillates between 863°C and 835°C. Finally, the contraction of Berriasian clay material is marked by a hook on the curves, resulting from the reversible $\beta \rightarrow \alpha$ of quartz, followed by final shrinkage ends between 1.92% and 2.08%. The parameters of firing are deferred on the figure 7.

4. Ceramic tests

TABLE 6: Parameters of drying of J. Oust Berriasian clays (wt. %)

	Shaping water	Interposition water	Colloidal water	Drying shrinkage
EBe1	19.07	13.60	5.47	3.22
WBe1	13.40	9.35	4.05	2.76
WBe2	13.82	9.62	4.20	2.52
WBe3	18.98	16.45	4.53	2.82

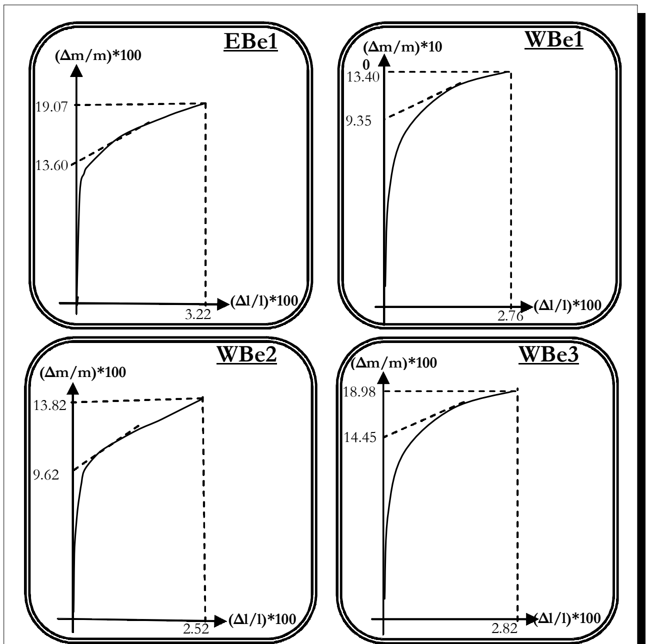

Figure 5: Drying curves of J.Oust Berriasian clays

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Taking into account the physical-chemical[19-20] and geotechnical results, the clay referred to as EBe1 has greater values of plasticity, drying shrinkage, firing shrinkage and maximum expansion than WBe2 clay. For this reason, the addition of a grease-remover (sand) is essential for a better use of these clays manufacturing ceramics. The ceramic tests on the ceramic pieces were carried out on two types of mixtures: M1 and M2, both of them are representatives of the Berriasian clays of J.Oust.

M1= 85% EBe1 + 15% sand
M2= 93% WBe2 + 7% sand

The ceramic tests on pieces (table 8) show that the flexural strength in raw is 3.88 N/mm ^{2} for M1 and 3.16 N/mm ^{2} for M2. However, the drying shrinkage is 2.11% for M1 and 1.88% for M2. The firing shrinkage increases when temperature increases. It stabilizes at 1000 with 19% for M1 and 1.32% for M2. The loss of ignition follows also the same analogy and it increases when temperature increases.

TABLE 7: Thermogravimetric analysis

Samples	1st loss (%) Water hydration	2nd loss (%) Organic matter	3rd loss (%) Dehydroxylation	4rt loss (%) Decarbonation	Total (%)
EBe1	1.56	0.22	2.11	16.14	20.03
WBe2	2.10	0.43	3.12	6.43	12.08
Range of temperature (°C)	45-240	280-340	465-640	700-830	

TABLE 8: Ceramic tests of the ceramic pieces

Mixtures	Raw flexural strength (N/mm²)	Drying shrinkage (%)	Temperature of firing (°C)	Firing shrinkage (%)	Loss of ignition (%)	Water absorption (%)	Cooked flexural strength (N/mm²)
M1	3.88	2.11	900	0.95	12.44	23.38	7.58
			950	1.52	15.95	17.32	10.38
			1000	1.90	19.92	15.4	18.521
			1050	1.91	19.91	13.51	23.60
M2	3.16	1.88	900	0.82	13.51	21.51	6.42
			950	1.27	16.32	19.38	10.55
			1000	1.32	16.81	16.51	16.33
			1050	1.32	16.80	16.30	20.52

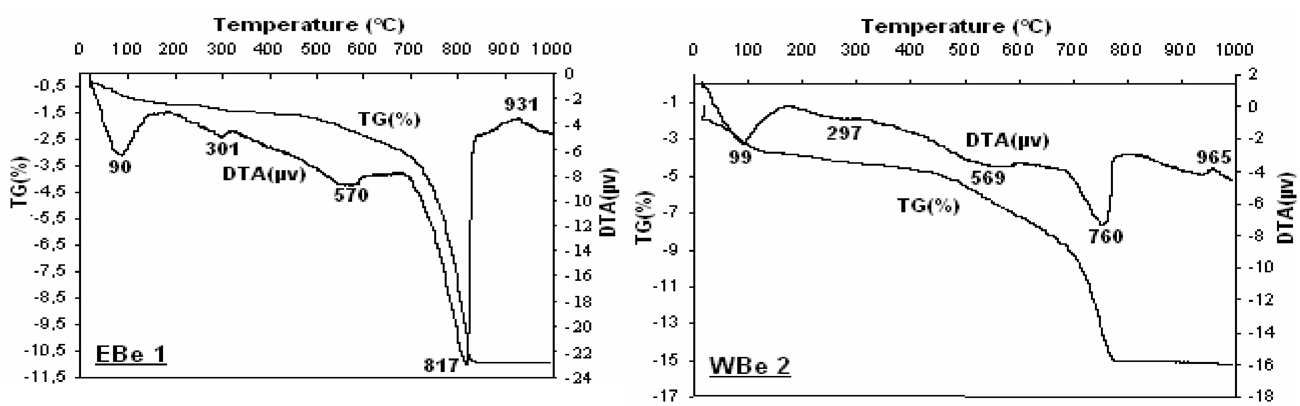

Figure 6: Curves DTA/TG of J.Oust Berriasian clays

The values oscillate between 12.44% at 900°C and 19.91% at 1050°C for M1, and between 13.51% at 900°C and 16.8% at 1050°C. When temperature increases the absorption water decreases.

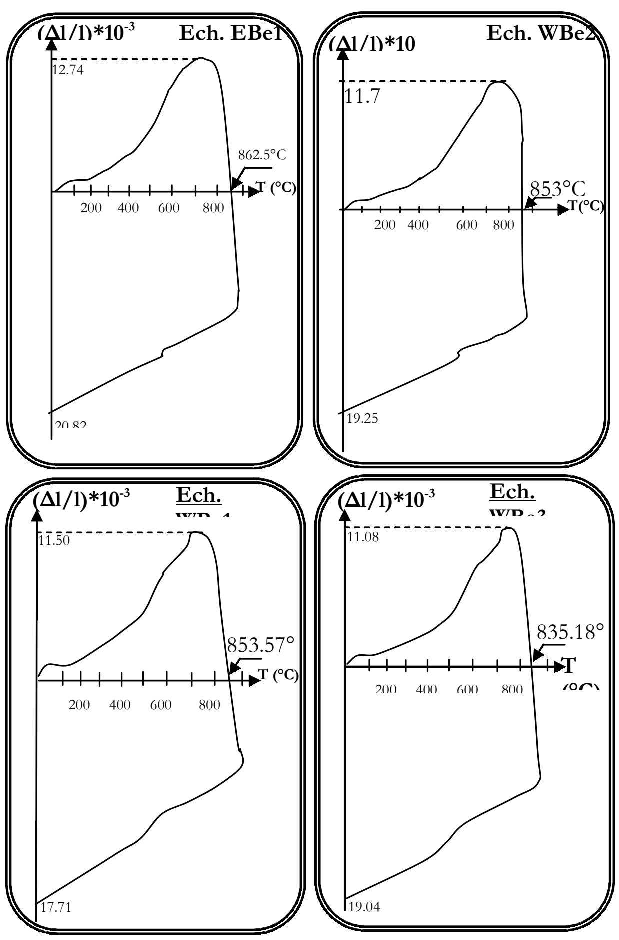

Figure 7: Dilatometry of crude samples of J. Oust Berriasian clays

CONCLUSION

This research focused on the physical–chemical study, supplemented by geotechnical characterization and industrial tests of J. Oust Berriasian clay, targeting a possible application in the field of ceramics.

These clays are characterized by the predominance of illite (57 % to 71%) and kaolinite (11 % to 24%). The proportions of interstratified illite/smectite (4 % to 20%) are close to chlorite (0% to 13%). They are notably siliceous (34.47% to 46.13%), carbonated (13.12% to 20.66%) and aluminous (11.27% to 14.03%). The quantity of iron oscillates between 4.1 % and 5.38 %. The existence of zones rich in K_2O (2.33% to 3.02 %) testifies the presence of illite, which is in agreement with the mineralogical analysis. The geotechnical identification shows that the drying shrinkage of clays is weak; it varies between 2.52% and 3.22 %. The firing shrinkage oscillates between 1 77% and 2.08 %. Moreover, we have established that the clays of the studied levels are little to fairly plastic, and that they adapt to fast drying.

Finally, the Berriasian clay of J.Oust could be used in the manufacture of ceramic products, namely ceramic pieces.

REFERENCES

[1] M.Solinag; Thesis és-Sciences, Lyon, 700 **(1927)**.

[2] G.Castany; Annales Mines Géologie, Tunisie, **8**, 595 **(1951)**.

[3] G.Castany; Annales Mines Géologie, Tunisie, **14(8)**, 632 **(1955)**.

[4] A.Jauzein; 'Carte géologique de la Tunisie au 1/50000', Ed., Serv. Géol. Tunisie, **(1957)**.

[5] A.Jauzein; Thesis d'Etat, Faculté des Sciences, Paris, Tunisie, 475 **(1962)**.

[6] L.Memmi; Notes Services Géologie, Tunisie, **31**, 41 -50 (1969).

[7] L.Memmi; Notes Services Géologie, Tunisie, **45**, 45 -53(1979).

[8] L.Memmi; Thesis d'Etat, Université Claud Bernard, Lyon I, 344 **(1990)**.

[9] Stranik et al; Notes Services Géologie, Tunisie, **41**, 20-25 **(1974)**.

[10] A.L. Maamouri, et al; Zemny Plyn a Nafta, Hodnin, Cerven, **39**, 73-105 **(1994)**.

[11] L.P.Meir, R.Nuesch; Journal of Colloids and Interfaces Sciences, **217**, 77-85 **(1999)**.

[12] S.Mahmoudi; Mastère, Faculté des Sciences, Tunisie, 76, **(2004)**.

[13] F.Jamoussi; Thesis d'Etat, Université Tunis El Manar, Faculté des Sciences, Tunisie, 437 **(2001)**.

[14] M.Ben M'Barek; Thesis de Doctorat, Université Tunisie, Faculté des Sciences, Tunisie, 223, **(2001)**.

[15] L.C.P.C., **10**, 26 **(1987)**.

[16] Holtz and Kovacs; Current Research in Earth Science Bulletin, **244**, 3 **(1981)**.

[17] G.Aliprandi; 'Matériaux Réfractaires et Céramiques Techniques', Septima, Edition, Paris, 225-292 **(1979)**.

[18] P.S Santos; Ciencia e Tenologia de Argilas, **I**, 3rd Ed., Edgard Blucher ltda, S.Paulo, **(1997)**.

[19] S.Ferraari, A.F.Gualteri; Appl.Clay Sci., **XX**, In press **(2005)**.

[20] A.Aras; Appl.Clay Sci., **24**, 257-26 **(2004)**.