

Late Cretaceous and Palaeocene clays of the northern Tunisia: potential use for manufacturing clay products

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Abstract Late Cretaceous and Palaeocene clay deposits of the northern Tunisia were studied for potential valorization in clay product manufacturing. To achieve this goal, physico-chemical characterization based on multidisciplinary approaches including mineralogy, geochemistry and thermal analyses was carried out. Clay samples collected from the Bir M'Cherga and Tajerouine study sites have undergone chemical analysis by using atomic absorption spectroscopy. The obtained results showed that the studied samples were mainly composed of silica, alumina, calcium and iron oxide. Mineralogical analysis confirmed the results of chemical analysis; it showed the main peaks of quartz, calcite, kaolinite, smectite and illite. Those clay samples were used as natural resources for manufacturing earthenware tiles. Mixtures of clays and dolomites were prepared from the Late Cretaceous and Palaeocene clays and then heated to the desired temperature to obtain different earthenware products. Finally, the prepared clay products were tested via different geotechnical analyses. It was found that the clay percentage needed to be decreased in the starting mixture to enhance the properties of the final products. Those results also indicated that industrial application of both Late Cretaceous and Palaeocene clays of

the northern Tunisia could be done, especially for manufacturing clay products.

Keywords Clay · Mineralogical analysis · Geochemistry · Industrial tests · Clay products · Late cretaceous · Palaeocene

Introduction

Valorization of natural clays for various applications, especially in ceramics and earthenware industry, is fundamentally important for the development of raw material sector in Tunisia. The desired industrial application depended on the physico-chemical characteristics and the thermal treatments as important properties for the industrial application extension of the studied materials. For these reasons, numerous researchers paid special attentions to the outcropping and easily accessible clay deposits (Baccour et al. 2008a, b; Mahmoudi et al. 2008a, b, 2010; Srivastava and Mankar 2013). Those studies showed that clay deposits could be effectively used for the manufacture of various ceramics. In this context, natural clay samples from the northern Tunisia (i.e. Bir M'Cherga and Tajerouine areas) were studied for potential use in manufacturing clay products. Mineralogical, geochemical and technological aspects were carried out to evaluate the potential application as raw materials for the manufacture of earthenware products.

Two main clay deposits were selected for the present study; the first lied 30 km to the south of the Tunis city (i.e. Bir M'Cherga) whereas the second belonged to Tajerouine area, northwestern Tunisia (Fig. 1).

From structural geology viewpoint, the area of Bir M'Cherga belongs to the field of the northern Atlas of Tunisia; it consists of a series of anticlines affected by NE-SW submeridian faults (Jauzein 1967; Zargouni 1975) which in turn cut by major E-W faults (Burolet 1956; Ben Ayed 1993;

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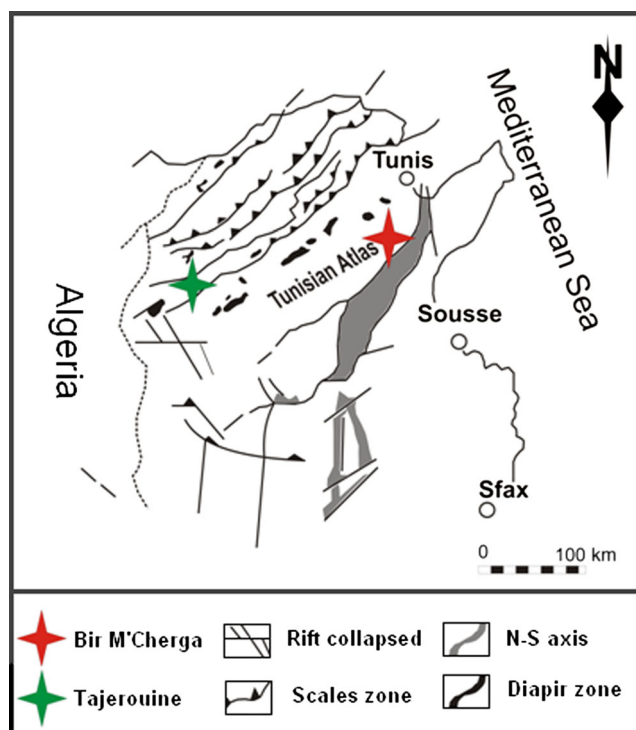


Fig. 1 Location of study areas

Boukadi 1994; Marzougui et al. 2014; Aridhi et al. 2014) and associated with grabens orthogonal to collapsed folds (Solignac 1927; Castany 1954; Ben Ayed 1993; Turki 1985; Chihi 1995; Chikhaoui 2002). This region is characterized by tectonic phenomena in compliance with the relevant tectonic framework of Tunisia. According to Jauzein 1967, it consists of parallel axes to the major Tunisian central accident, where faults and flexures are generally orthogonal to the NE-SW folds of the major phase.

As for Tajerouine area, it belongs to the northwestern edge of Tunisia as an intermediate zone between the central Atlas and northern Atlas of Tunisia with diapir and grabens (Burolet 1956; Burolet and Sainfeld 1956; Ben Ayed 1993). This region is dominated by folded features dispersed within rift basins of the Kalaat Khasba, Rohia and Bou Ghanem grabens (Chihi 1995).

Materials and methods

Study sites

Fifty clay samples were collected in two reference sections of the northern Tunisia (i.e. Bir M'Cherga and Tajerouine). The Bir M'Cherga study site is located 30 km to the south of Tunis (Lambert coordinates $X_A=363.5$, $Y_A=520.5$, $X_B=364$, $Y_B=$

522; Bir M'Cherga map N° 28, 1/50,000) where the marly series of the Upper Maastrichtian-Palaeocene age were overlain by the Eocene limestone bars (Fig. 2).

The Tajerouine section, belonging to the Kef district (38 km towards Tunisian-Algerian borders), is limited by the high reliefs of Thala to the south and those of Kef to the north (Lambert coordinates $X_A=382$; $Y_A=383$; $X_B=287$; $Y_B=286$; Tajerouine map N° 51, 1/50,000). It consisted of a thick marly series of the Upper Maastrichtian-Palaeocene system to the southwestern flank of Jebel Houdh covered by the Eocene limestone bed (Fig. 3). The collected clay samples were dried at 105 °C for 24 h, crushed and sieved; the <40- μm -sized subsamples were kept in sealed bottles for further analyses from where the clay fraction (<2- μm -sized particles) was separated by centrifugation according to the procedure of Holtzapffel (1985).

Chemical analysis by AAS

Chemical analysis was carried out by atomic absorption spectroscopy (AAS) using a PerkinElmer apparatus with peroxide acetylene flame. This analysis allowed the determination of the major elements' relative percentages (i.e. SiO_2 , Al_2O_3 , Fe_2O_3 , CaO , K_2O , MgO , Na_2O , SO_4) and loss on ignition.

Mineralogical analysis by XRD

Mineralogical analysis of the collected clay samples was determined by X-ray diffraction analysis using a Philips PW 1730 diffractometer equipped with Cu-K α filter. XRD patterns of the original clays were obtained from randomly oriented powders (<40- μm -sized fraction); those of the clay fraction subsamples (<2 μm) were determined on oriented pastes as the following: (1) without pretreatment (air-dried), (2) saturated with ethylene glycol and (3) heated at 550 °C for 2 h. Percentages of clay species were obtained from the relative height of the 001 reflections (Moussa et al. 1992; Ben M'barek et al. 2002; Baccour et al. 2008a, b; Jeridi et al. 2008) recorded on the diffractograms of the oriented pastes (Moore and Reynolds 1989).

Micro-granulometric and granulometric analysis

Particle size analysis of the clay samples (grey clays with foliated or platelet layers and iron oxide) was carried out by simple sedimentation using laser granulometry analyzer (Micromeritics SediGraph D5000 analyzer). The overall particle diameter was determined by measuring the sedimentation speed (V) of suspended particles. It is given by Stokes law as follows:

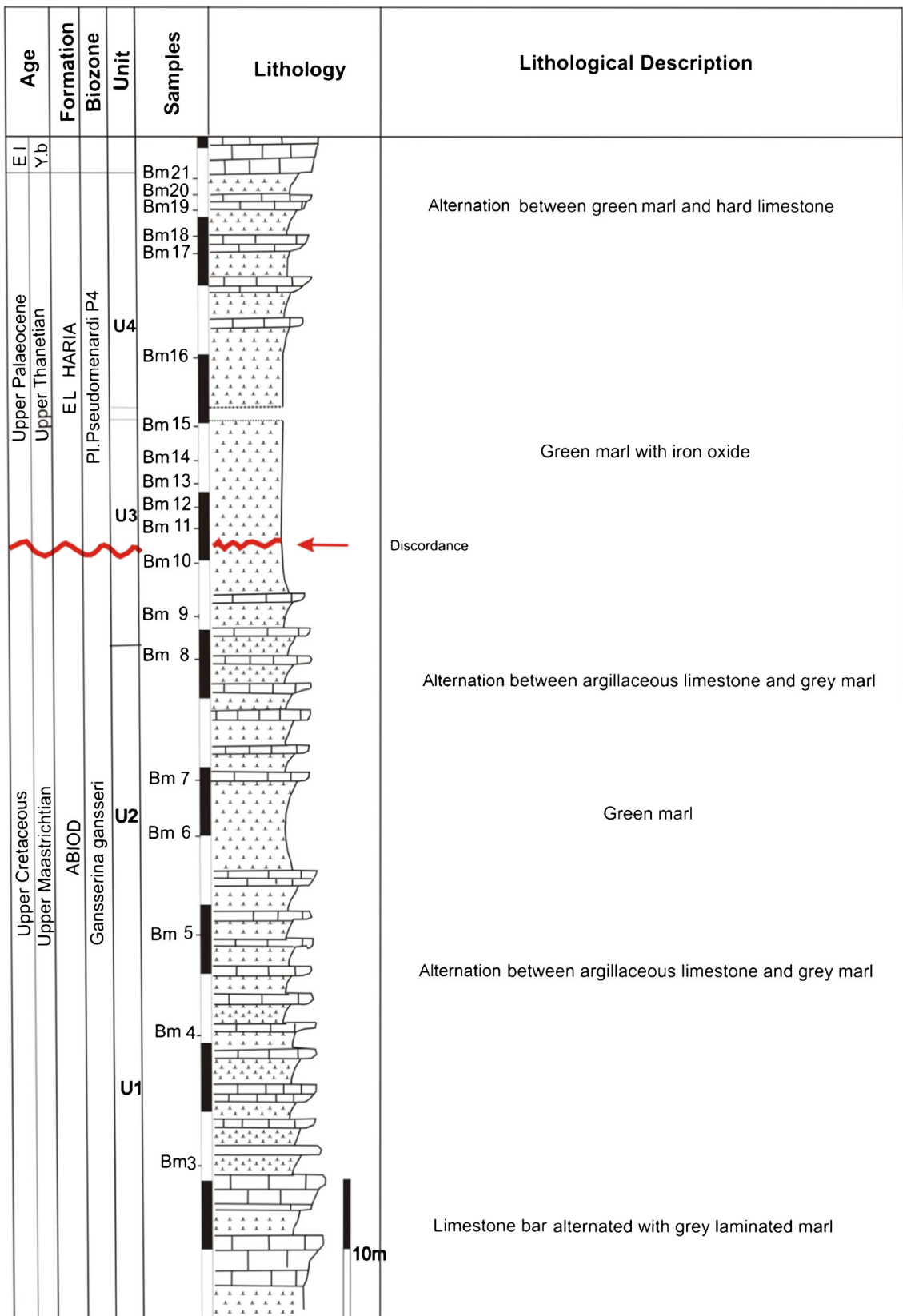


Fig. 2 Cross section of Bir M'Cherga study site

$$D = KV^{1/2} \tag{1}$$

where *K* is dependent on the viscosity of the dispersing liquid, the density of the liquid fluid, the volumetric mass of the particles and the acceleration of gravity. This device determines, via an X-ray beam, suspended particles at a different sedimentation height. The distribution of cumulative weight percentages is calculated for diameters between 0.1 and 100 μm. Micro-size analysis was carried out not only for the determination of <2 μm fraction percentage but also for reliable information about the mode of transport and sedimentation (Rivière 1977).

Thermal analysis

Thermogravimetric analysis (TG) was performed using a Rigaku Thermoflex apparatus on a representative sample of

the collected clays. Differential thermal analysis (DTA) was also carried out to determine the reactions that may occur during the progress of the heating programme. Dilatometric and Bigot’s tests were plotted using an Adamel-kind device DI24 reaching 1200 °C at an increment rate of 5 °C/min.

Semi-industrial tests

Physical properties of the prepared tiles were ascertained for a mixture of raw white clay (85 %) and dolomite (15 %) (*M_r*) to prepare clay products (i.e. tiles). Three main steps were needed to prepare various clay products: (1) tile preparation, (2) drying and (3) firing. A brief description of the experimental procedure is given below

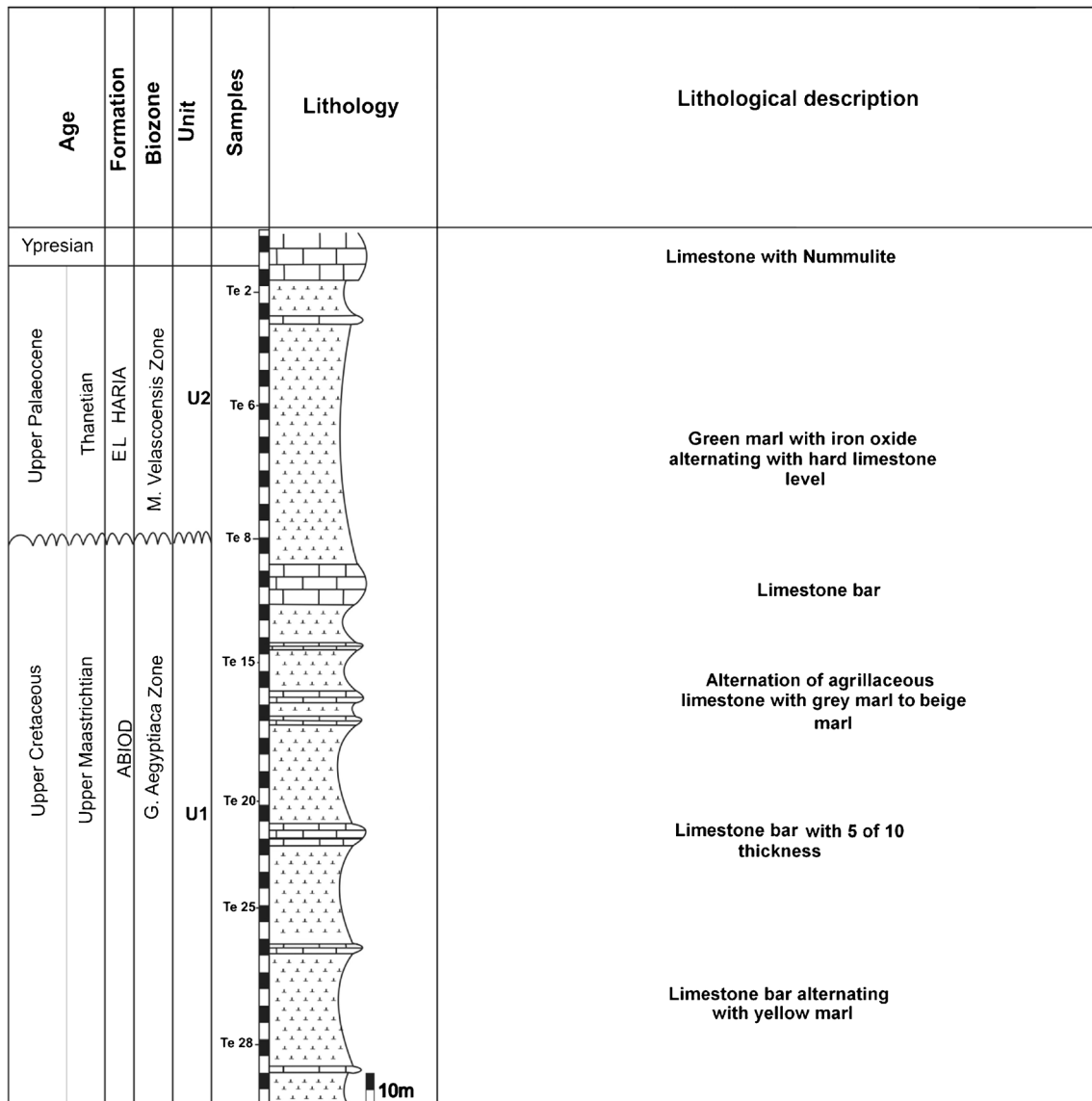


Fig. 3 Cross section of Tajerouine study site

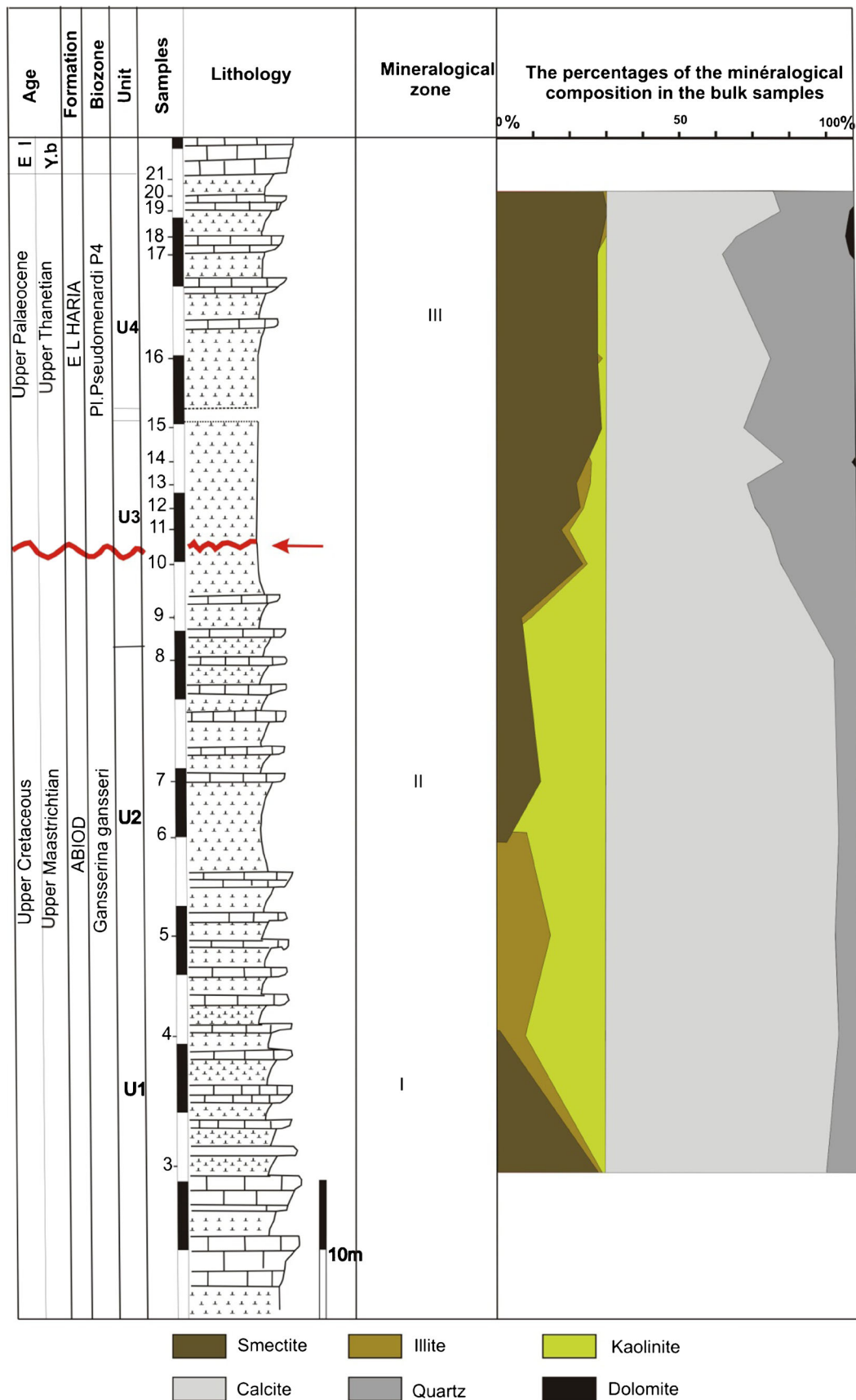


Fig. 4 Mineralogical study of clays from Bir M'Cherga section

Table 1 Mineralogical composition of the studied clay deposits (in weight %)

Location	Mineralogical Zonation	Sample	Clay mineral			Non-clay mineral				
			Smectite	Illite	Kaolinite	Calcite	Quartz	Dolomite		
Bir M'Cherga	ZB1	BM ₃	94.39	2.76	3.32	87.16	12.83	nd		
		BM ₄	2.39	22.80	70.17	91.48	8.51	nd		
	ZB2	BM ₅	–	50	50	89.58	10.41	nd		
		BM ₆	7.89	17.54	75.43	91.48	8.51	nd		
		BM ₇	40	nd	60	92.14	7.85	nd		
		BM ₈	30.92	nd	69.07	90.27	9.72	nd		
		BM ₉	24.62	9.09	66.28	82.16	17.83	nd		
		ZB3	BM ₁₀	79.90	4.11	15.98	69.39	30.60	nd	
			BM ₁₁	60.93	7.81	31.25	64.76	35.23	nd	
			BM ₁₂	77.7	4.10	18.18	58.09	41.90	nd	
			BM ₁₃	73.1	14.11	12.77	56.42	43.57	nd	
			BM ₁₄	83.76	2.80	13.42	70.5	28	1.66	
			BM ₁₅	97.93	0.32	1.74	54.26	45.73	nd	
		ZB2	BM ₁₆	96.29	1.44	2.26	65.07	34.92	nd	
			BM ₁₇	94.29	0.86	4.84	45.37	52.86	1.76	
			BM ₁₈	97.46	1.07	1.46	51.93	45.06	3.00	
			BM ₁₉	99.35	0.36	0.68	67.93	29.34	2.17	
			BM ₂₀	98.29	1.43	0.38	66.31	33.68	nd	
		Tajerouine	ZT1	Te28	92	3	5	56	44	nd
				Te25	87.5	2.5	10	55	45	nd
ZT2	Te20		49.5	5.5	45	56	24	18		
	Te15		40	6	44	25	75	nd		
	Te8		19	2	79	45	55	nd		
ZT3	Te6		4	9	87	37	63	nd		
	Te2		nd	30	70	58	26	nd		

nd not detected

1. Preparation of earthenware tiles: a powdered raw clay was mixed with sand and dolomite to get a mixture containing clay (90 %), sands (5 %) and dolomite (5 %). This mixture was a <500- μm -sized fraction obtained after crushing and grinding of small portions of 60 g (Fig. 8). The mixture was introduced in a hand-pressed mould that was put under 240 bars (Jouenne 1990).
2. Drying: tiles were air-dried for 24 h and then oven dried at 110 °C until constant weight.
3. Finally, the dried sample was fired in an electric furnace at different temperatures of 850, 900 and 1050 °C. The firing programme was set at a temperature increment of 5 °C/min.

illite and kaolinite, as main clay minerals. Associated minerals were calcite, quartz and dolomite (Table 1). According to the distribution of the percentages of clay and non-clay minerals, one could define three distinct mineralogical zones including the lower (zone I), medium (zone II) and top interval (zone III). Clay abundance varied according to the section and zone (Figs. 4 and 5). For instance, smectite was abundant in the zone I of the Bir M'Cherga section, while in that of Tajerouine, kaolinite was dominant. Similar observation was made for both sections in zones II and III: zone II showed abundant kaolinite associated with smectite and illite while zone III revealed the abundance of smectite.

Bir M'Cherga section

Mineralogical analysis of clay samples from Bir M'Cherga cross section revealed the dominance of smectite and kaolinite reaching 30 and 21 % of raw clay samples, respectively (Table 1). In contrast, illite content was lower than 15 %. Non-clay minerals were mainly composed of quartz, calcite

Results and discussion

Mineralogical analysis by X-ray diffraction

Mineralogical analysis of the Late Cretaceous and Palaeocene clay sections showed the presence of smectite,

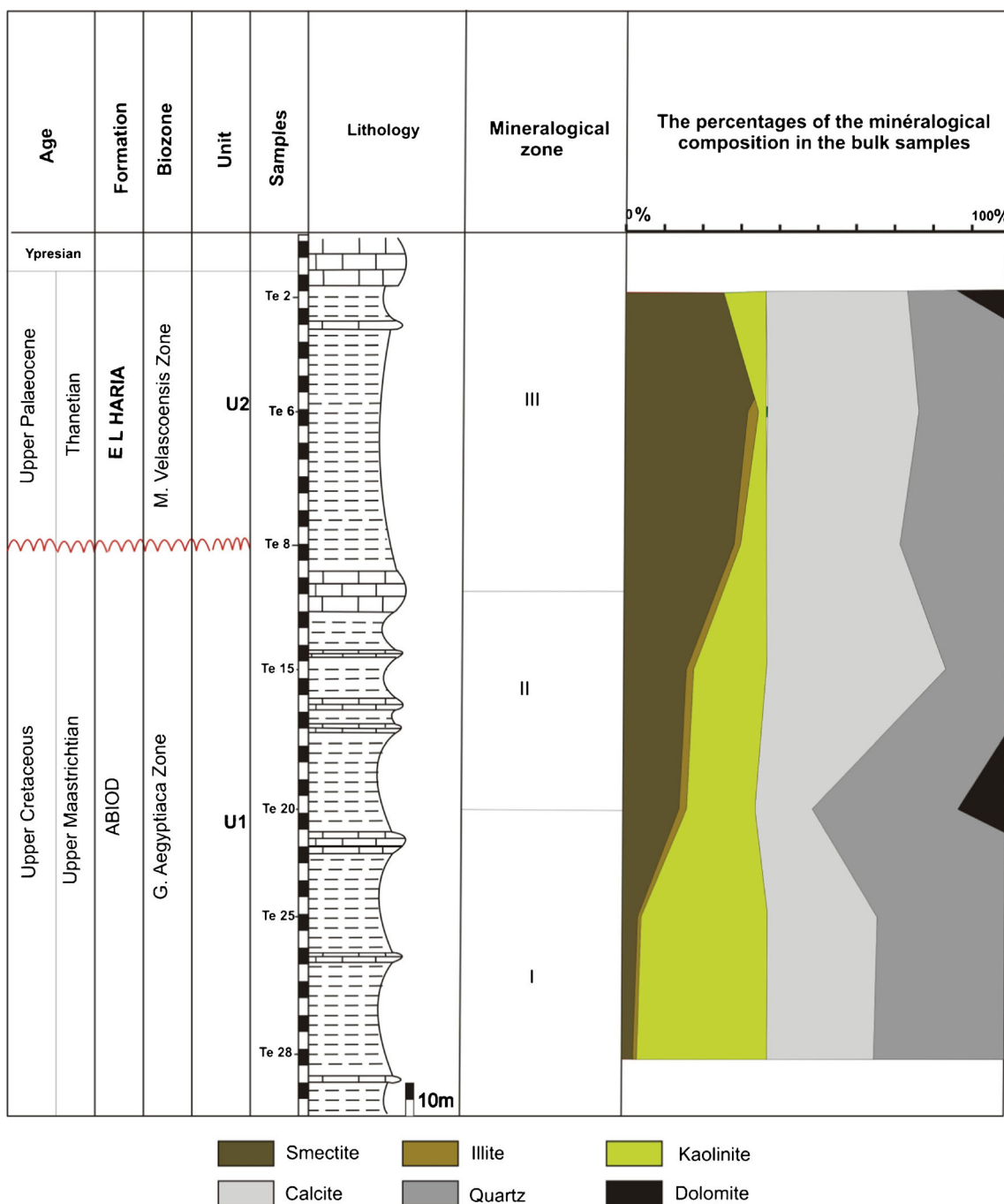


Fig. 5 Mineralogical composition of clays from Tajerouine section

and with small amounts of dolomite. A brief description of the distinguished mineralogical zones was given below (Fig. 4):

Zone I is the lower part of the Bir M'Cherga cross section which was dominated by smectite (28 % BM₃), with small amounts of kaolinite and illite. As for non-clay minerals, calcite represented 61 % while quartz 9 %, indicating the absence of dolomite.

Zone II lied to the medium of the section with dominant kaolinite, ranging from 15 to 22 %, subordinated by illite (3 to 15 %). The proportion of smectite increased upwards from 1 to 12 %. Associated minerals were mainly composed of calcite (57 to 64 %) and quartz (6 to 13 %), but no dolomite (Table 1).

Zone III was dominated by smectite with a percentage of 18 to 30 %; that of kaolinite ranged between 1 and 10 % (Fig. 4, Table 1). Those values were much higher than

Table 2 Chemical composition of the collected clay samples (in weight %)

Location	Sample	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	K ₂ O	Na ₂ O	SO ₃	LOI
Bir M'Cherga	BM ₁	17.20	34.00	12.68	4.04	4.23	1.96	4.35	0.14	22.98
	BM ₂	17.51	34.99	14.71	4.37	3.53	1.01	3.14	0.28	21.11
	BM ₃	34.47	17.44	9.94	2.60	0.74	0.93	3.08	0.13	32.41
	BM ₆	38.16	12.64	7.27	2.23	0.63	0.93	3.14	0.13	35.69
	BM ₉	32.33	20.01	9.68	4.05	1.34	0.57	1.58	0.14	30.31
	BM ₁₀	26.22	27.48	10.82	4.54	2.18	0.65	2.01	0.15	25.95
	BM ₁₁	18.41	34.20	15.58	5.02	2.57	1.11	3.80	0.18	21.86
	BM ₁₄	23.14	28.16	12.99	2.71	2.71	1.00	3.34	0.19	25.29
	BM ₁₆	15.09	34.29	9.13	2.22	2.22	1.15	3.95	0.16	21.99
	BM ₁₇	9.05	44.81	15.75	4.94	4.25	0.79	1.84	0.17	18.40
Tajerouine	Te ₂	23.31	20.01	10.82	3.14	3.14	1.04	3.94	0.16	25.16
	Te ₂	24.72	24.35	13.46	5.29	5.04	0.15	0.12	3	23.88
	Te ₆	3.24	33.87	15.76	8.74	30.53	1.56	0.50	0.55	11.97
	Te ₈	9.83	31.87	15.29	7.98	12.40	1.23	0.41	4.15	15.73
	Te ₁₅	27.34	24.85	10.95	4.64	1.01	0.75	0.23	0.69	25.85
	Te ₂₀	29.58	21.26	9.92	4.59	2.53	0.19	0.05	1.19	28.47
	Te ₂₅	9.11	34.24	9.63	6.33	14.02	0.12	0.04	9.43	13.13
	Te ₂₈	14.05	31.57	14.52	7.19	1.49	0.62	0.21	2.23	28.08

LOI. Loss on ignition

those in illite contents, which showed less than 2 % for all samples. High amounts of calcite (32 to 49 %) and quartz (20 to 35 %) as non-clay minerals were recorded.

those in zone II, but slight increase in dolomite content was observed near the top of the section (Table 1).

Tajerouine section

Tajerouine clay samples were mainly composed of abundant kaolinite and smectite, reaching 32 % for the Te₂₈ sample; illite content did not exceed 1 % in all samples, except for the Te₂ sample (11 %, Table 1). Associated minerals are represented by calcite, quartz and occasionally dolomite. Similar to the Bir M'Cherga cross section, three distinct mineralogical zones were identified (Fig. 5):

Zone I (lower part) is where kaolinite reached 32 % with low amounts of illite (<2 %, Table 1). Associated minerals were quartz (35 %) and calcite (29 %).

Zone II (middle part) was marked by a slight increase in smectite content, reaching 16 %; those of illite varied between 1 and 2 % and kaolinite ranged between 18 and 19 %. Abundant calcite (47 %), quartz (16 to 37 %) and low dolomite content (<12 %) constituted the non-clay minerals.

Zone III represented the top of the Tajerouine section. This is a smectite-enriched zone that varied between 26 and 32 %. Illite content ranged between 1 and 11 % with slight decrease of kaolinite contents (2 to 7 %). Associated minerals (calcite and quartz) showed similar trends as

Chemical composition by AAS

Major element contents of the collected clay samples were shown in Table 2. The obtained results showed high amounts CaO and Fe₂O₃. Those of SiO₂ indicated the importance of detrital contribution. Al₂O₃ has a relatively low percentage relative to the total rock. Its origin cannot be attributed to the tetrahedral layer of clay minerals. K₂O content was high; it increased with illite percentage as this was directly related to illite contents. Amounts of alkaline oxides (Na₂O and K₂O) were high in all clays due to the large amounts of illite (Monterio and Vieira 2004). Effects of sulfur can be overcome by addition of barium carbonate to form a more stable barium sulfate (Tajerouine section). Besides its fluxing role, Fe₂O₃ also provided the fired products with the distinctive reddish colour. However, Fe₂O₃ was not the only factor responsible for the coloration of earthenwares as other constituents (e.g. CaO, MgO, MnO and TiO₂) may significantly affect the colour of fired clays. In addition to these components (i.e. chemical contents), firing temperature, Al₂O₃ contents and the furnace atmosphere played an important role in the final colour of fired clay (Fisher 1984).

As for the chemical composition of argillaceous sediments, the depositional conditions are crucial factors in the

determination of the final lithology, therefore the dominant chemical element (Figs. 6 and 7). For instance, during the Late Cretaceous when conditions are met to develop carbonate levels, CaO contents significantly increased simultaneously with a clear decrease in silica (SiO₂) and alumina (Al₂O₃). During the Palaeocene, detrital character asserts from the base to the top of this stage showing increased Al₂O₃/K₂O ratio. This was further confirmed by the increase in iron/magnesium ratio (Fe₂O₃/MgO) during the Late Palaeocene. Both sections (i.e. Tajerouine and Bir

M’Cherga) showed somewhat similar siliceous clayey deposits (Ben M’Barek 1996; Ben M’Barek et al. 2000; Ben M’Barek 2001; Ben M’Barek et al. 2002).

Granulometric analysis

Particle size distribution of studied clay samples was a key factor in determining its potential applications as clay products, particularly clay fraction (<2 μm; Mahmoudi et al.

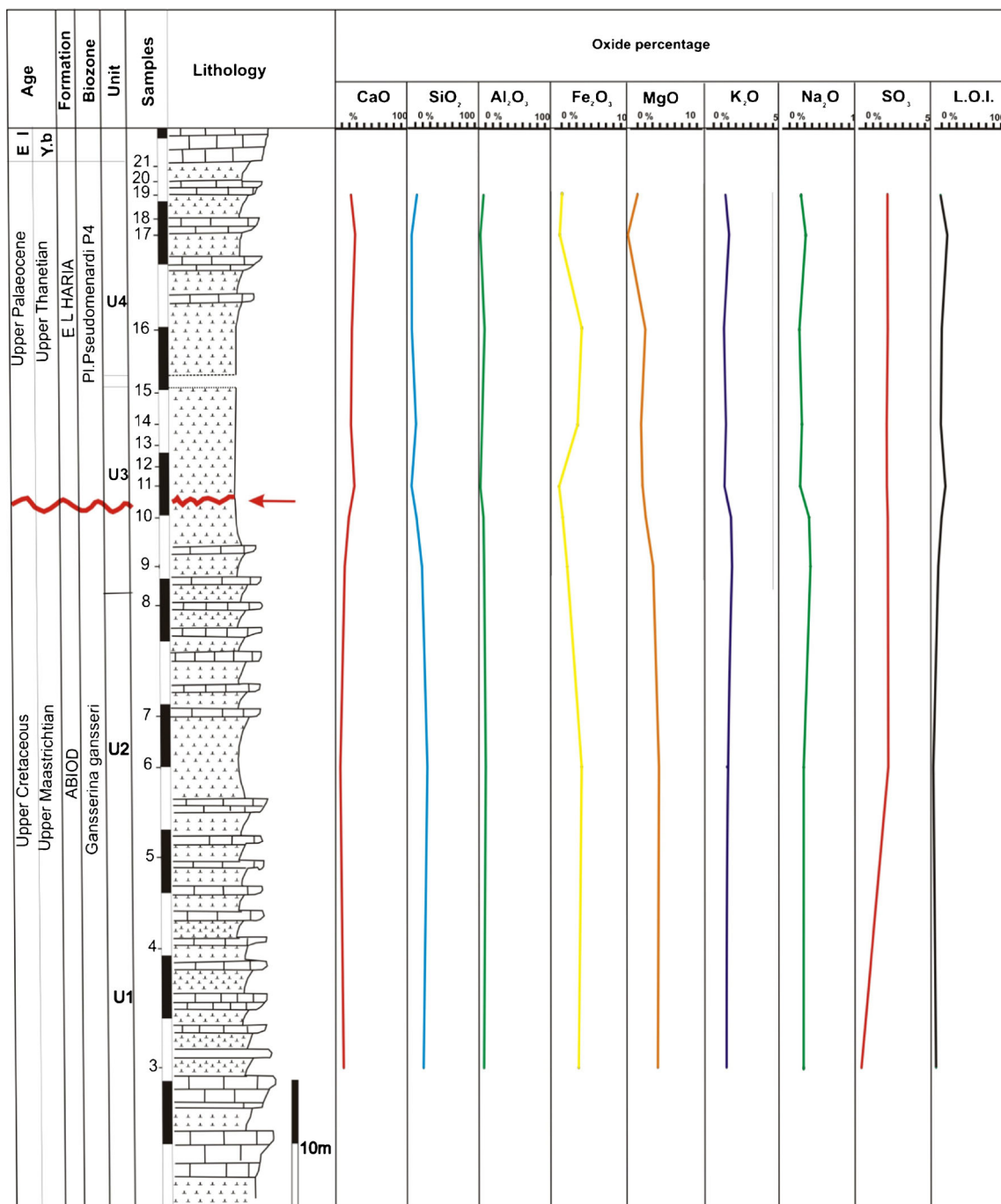


Fig. 6 Vertical distribution of the geochemical study of clays from Bir M’Cherga section

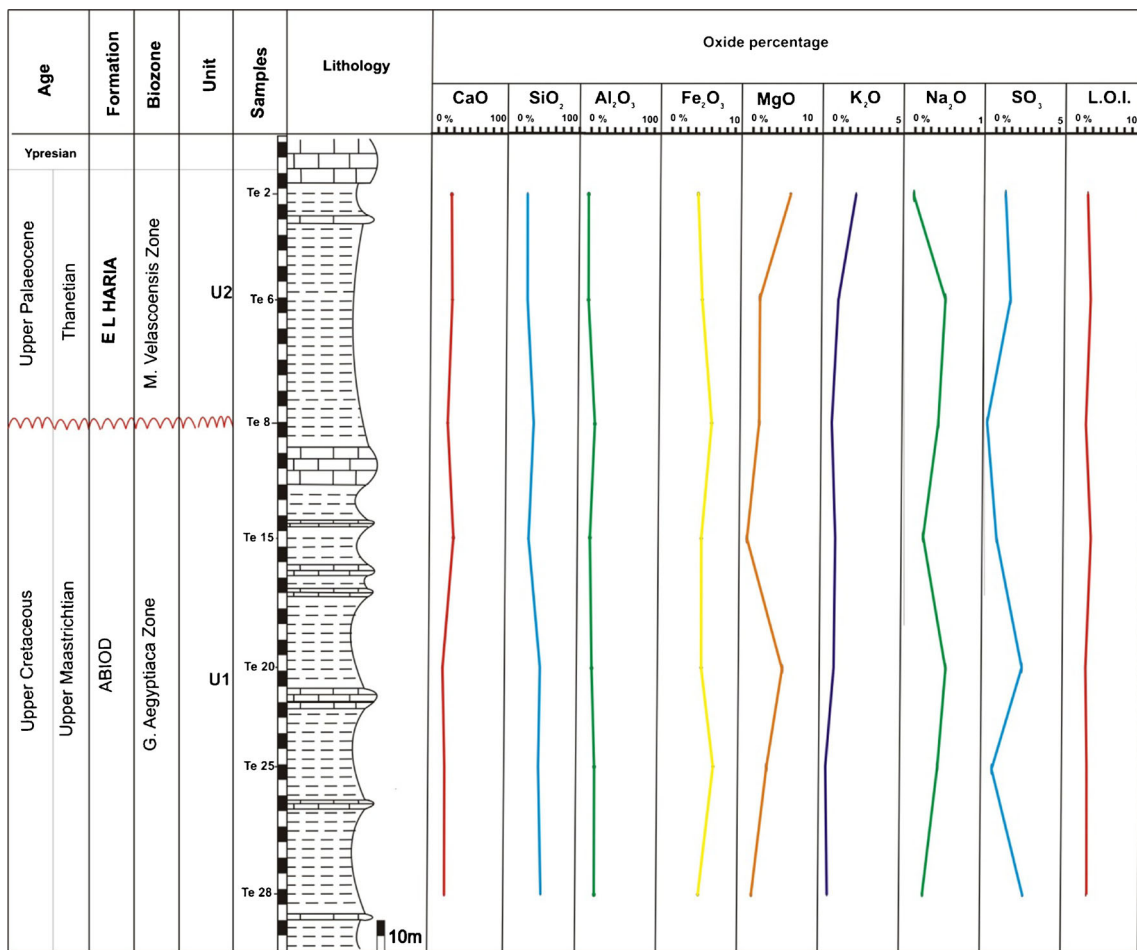


Fig. 7 Vertical distribution of the geochemical study of clays from Tajerouine section

2008a, b; Venkatraman et al. 2011). Granulometric analysis of the Tajerouine and Bir M’Cherga clays clearly indicated the homogeneous distribution. The percentage of >100- μm -sized grains reached around 50 % for both sections (Fig. 8).

Micro-granulometric analysis of clay mixtures showed that the <2- μm -sized fraction (clay fraction) reached 30 % for the Bir M’Cherga clay and 37 % for the Tajerouine samples. This analysis indicated that all of the studied clay samples were excellent candidates for clay product manufacturing.

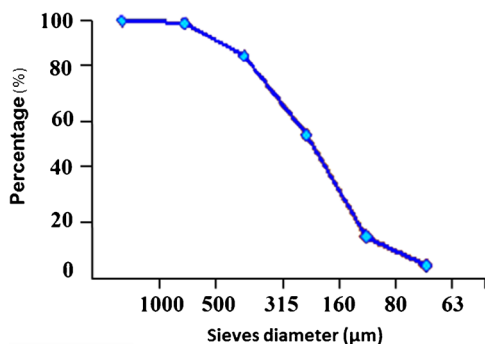


Fig. 8 Grain size distribution of representative clay sample

Imperfections that may arise because of the coarse sand fraction (200–2000 μm) can be remediated by simple grinding.

Drying and firing behaviour

According to the physicochemical properties of clays from the studied sections (Bir M’Cherga and Tajerouine), two mixtures were prepared from natural clays and dolomite (weight ratio of 90/10). Then, the mixture was analysed by the firing process described earlier to assess the behaviour of the Bir M’Cherga and Tajerouine clays.

Drying behaviour (Bigot’s curve)

The swelling/shrinkage behaviour of a common clay material may be assessed by the well-known Bigot’s test (Bigot’s curve). When clays dry out, water-dispersed particles tend to approach each other and agglomerate, resulting in cracks and consolidation or cohesion. When the particles are consolidated, no further shrinkage can be

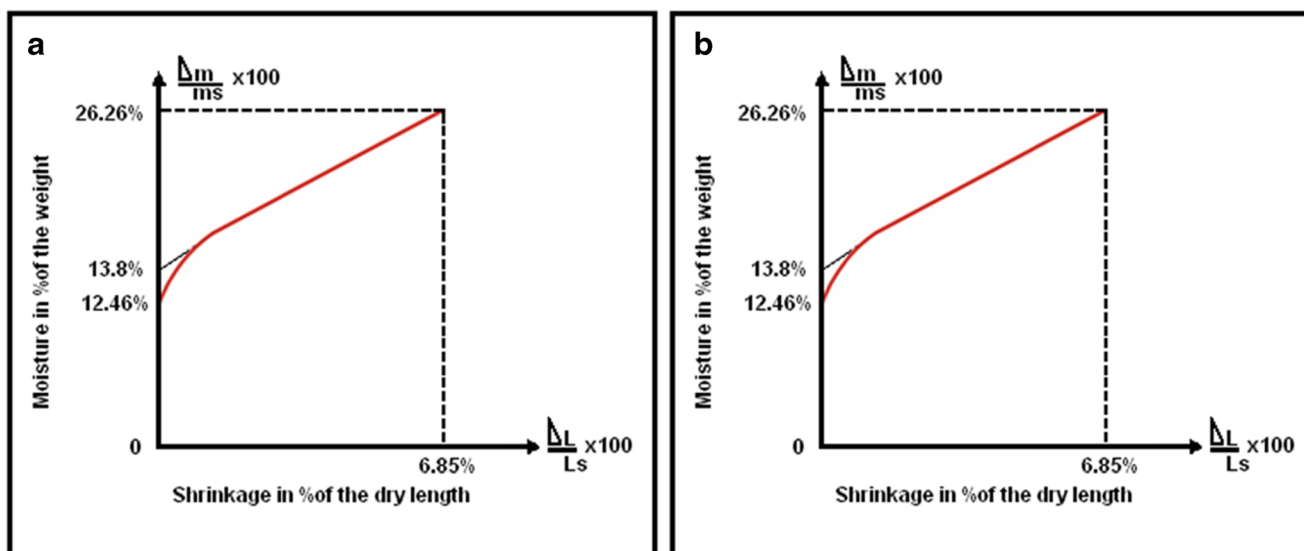


Fig. 9 Bigot's drying curve of clays from **a** Bir M'Cherga and **b** Tajerouine study sites

observed. It is therefore plausible to admit two stages of drying process: (1) water flow is accompanied by shrinkage and (2) drying without dimensional variation (Fig. 9). The evaporating water during the first phase is called colloidal water; that of the second phase is called interposed water. Drying percentage is also called the shaping water content or total water which represents the sum of all evaporated water (colloidal water and interposed water). The drying process can be ascertained through a drying curve; the results of which are shown in Fig. 9. The drying behaviour of the Bir M'Cherga clay showed total water percentage of 26.6 %; interposed water (12.8 %) and colloidal water (13.8 %) were quite similarly distributed. The final drying shrinkage was 6.85 %. The Tajerouine clay mixture showed a bit lower shrinkage with total water percentage of 19.43 %. The percentage of interposed water was 5.04 % while that of colloidal water reached 14.39 % with a final drying shrinkage of 2.47 %.

Dilatometric curves

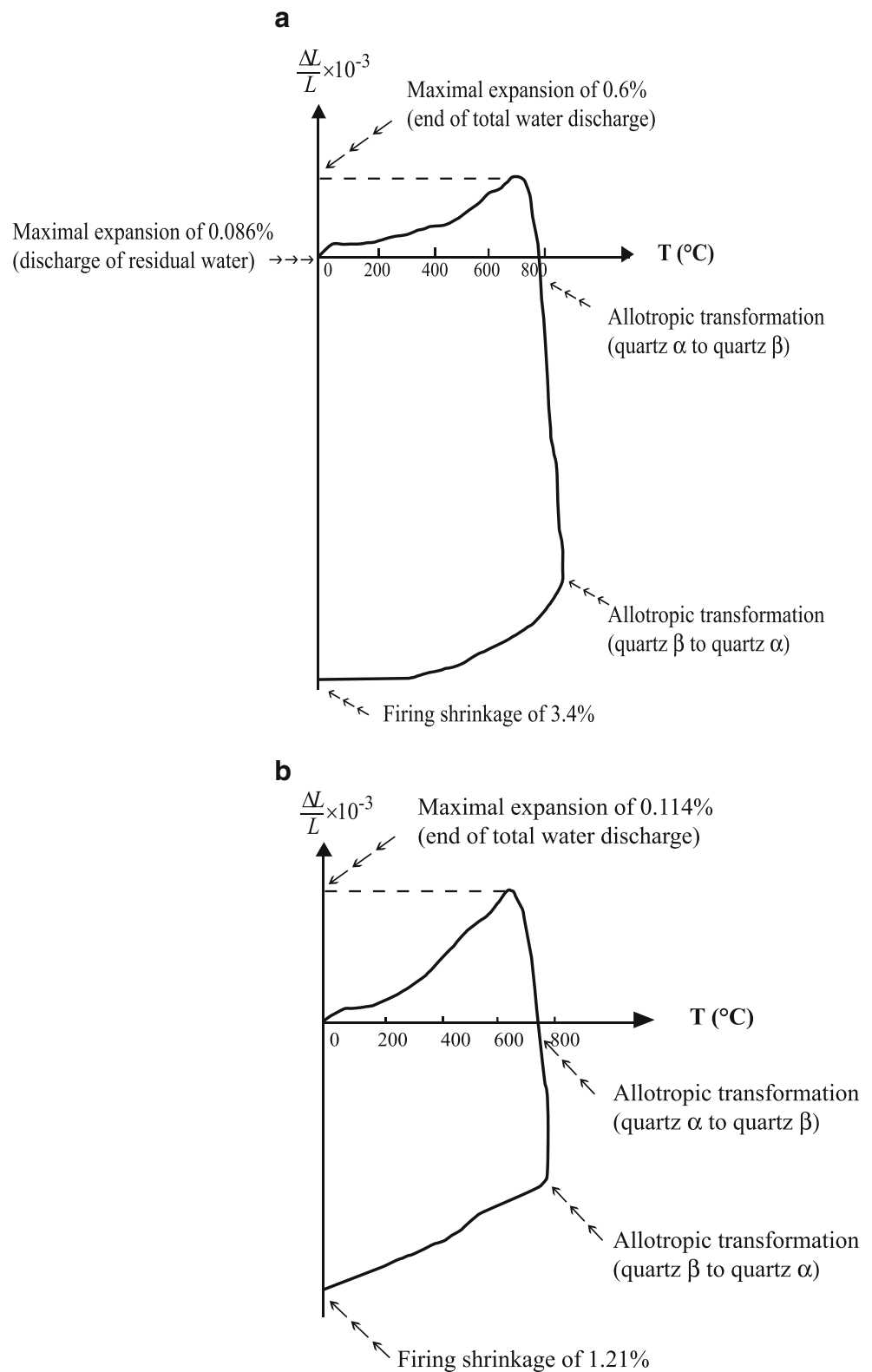
Dimensional changes observed after firing of the raw clays are given in Fig. 10. The Bir M'Cherga section developed a slight dilation of less 0.6 % due to the desorption of adsorbed water from ambient temperature to 110 °C. The expansion was followed by larger changes between 500 and 600 °C due to the $\alpha \rightarrow \beta$ quartz transformation and kaolinite breakdown (transforms into meta-kaolinite); it reached 6.06 % around 789 °C. Singer and Singer (1963) pointed out that α -quartz transformed into β -quartz at 573 °C with a volume increase of 2 %. Further slow heating of β -quartz changed to $\beta 2$ -tridymite at 870 °C with 12 % as volume change. Maximum expansion rate between 500 and

600 °C was observed near 583 °C. After a slight shrinkage around 850 °C, a sharp shrinkage near 952 °C was attributed to sintering and vitreous phases. This meant that the temperature interval gives optimum reaction time for the grains to react correctly; the obtained material would show high resistance to the deformation at high temperature, confirming the potentialities for earthenware production. Bigot's plot significantly deviates from its usual trend between 800 and 1000 °C, indicating the end of the fusible phase of the product. This curve was almost linear, indicating a cooking withdrawal of 3.4 %. The dilatometric curve of the Tajerouine representative tile showed slight dilation of 0.2 % from ambient temperature to 180 °C, followed by maximum dilation of 1.14 % at 740 °C. The beginning of sintering phase can be recognized at 750 °C with its great deviation between 800 and 900 °C, indicating the end of this phase. The cooling phase indicated a final firing shrinkage of 12.12 %. High content of K_2O in all raw materials may contribute to the rapid vitrification, especially in $< 2 \mu m$ fraction.

Technological tests on the prepared clay tiles

M_1 and M_2 were prepared as described earlier from the mixture of 5 % sand, 5 % dolomite and 90 % raw clay of Bir M'Cherga or Tajerouine study sites, respectively. M_1 and M_2 mixtures were used for the preparation of a sample set for comparative purposes. After air-drying for 4 h, test pieces were placed in an oven at different temperatures (30 °C, 50 °C and 110 °C). The drying cycle of the prepared tiles was then continued in an electric furnace up to 850 °C, 900 °C and 950 °C with an increment

Fig. 10 Dilatometric curve of clay mixtures from **a** Bir M'Cherga and **b** Tajerouine sections



temperature of 5 °C /min. The obtained results for manufacturing tiles are shown in Table 3. Drying shrinkage of the Bir M'Cherga clay mixture (M_1) was estimated to be 0.6 %, while that of Tajerouine mixture (M_2) was

slightly higher (0.8 %). Overall firing shrinkage of the prepared tiles was satisfactorily appreciated; it reached 3.8 % at 950 °C. In addition, flexural strength ranged from 4.95 to 16.15 N/mm² for M_1 and 4.3 to 5.83 N/

Table 3 Properties of ceramic tiles prepared from the clay samples of Bir M'Cherga (M₁) and Tajerouine (M₂)

Mixture	Drying shrinkage (%)	Firing temperature (°C)	Firing shrinkage (%)	LOI (%)	Porosity (%)	Flexural strength (N/mm ²)
M ₁	0.60	850	2.45	17.17	19.18	4.95
		900	3.74	17.57	17.25	9.68
		950	3.80	18.61	14.73	16.15
M ₂	0.80	850	1.10	4.85	12.57	4.30
		900	1.21	5.10	12.5	5.82
		950	1.40	5.61	11.22	5.83

mm² for M₂ at 950 °C. The porosity was 19.18 % for M₁ and 11.8 % for M₂. Loss of ignition increased slightly with temperature; it varied accordingly with carbonate contents in the studied clays. Its value was closely related to the molecular water, the oxidation of FeO, the decomposition of carbonates and the presence of organic matter. All raw material mixtures had red and yellow colours. The increased redness and yellowness can be attributed to the high amounts of some oxide impurities. Red-coloured appearances of the final product are very tolerable, making the studied clay materials good candidates for the desired industrial exploitation (Cherni 2008).

Conclusion

Late Cretaceous and Palaeocene clays from the northern Tunisia were studied to evaluate their potential application in the manufacture of earthenware tiles. Mineralogical analysis by X-ray diffraction on the <2 μm clay fraction revealed substantial mineralogical variation. Semi-quantitative calculations indicated that clay fraction was dominated by smectite and/or kaolinite, associated with illite. Considerable variation from the bottom upwards was observed in both sections (i.e. Late Cretaceous and Palaeocene).

It was also clearly observed that clay fraction (<2 μm) of the Late Cretaceous sediments showed homogeneous evolution with dominant kaolinite (40 at 90 % of the clay fraction), associated with illite and low content of smectite.

For Palaeocene clay of the Bir M'Cherga study site, clay mineralogical evolution clearly showed substantial gap near Cretaceous-Palaeocene passage. This was materialized by a dominance of smectite and illite but higher contribution of smectite estimated to 79 % in the Late Cretaceous and 87 % in the Palaeocene. The increase in smectite was more pronounced in the Bir M'Cherga site (northern Tunisia). Chemical and mineralogical analyses indicated kaolino-illitic marls, relatively enriched in alumino-ferruginous elements and smectite. The SiO₂/Al₂O ratio was estimated to 2–3, confirming the smectitic nature of the studied clays. K₂O

content was <2 %; that of iron (Fe₂O₃) was slightly higher (2–7 %). High percentage of CaO was confirmed by the high value of loss on ignition.

From the technological tests carried out on the prepared tiles, it was concluded that a firing temperature of 900 °C was high enough to cause a drying shrinkage of 2.47 to 6.85 % and water absorption of 8 %. Furthermore, the studied clays showed rapid drying with satisfactory firing shrinkage in line with the relative standards.

In conclusion, the clayey deposits of the Bir M'Cherga and Tajerouine study sites, northern Tunisia, showed the required specifications and satisfactory technological tests that would allow their use as raw materials for the preparation of good-quality earthenware. Further in-depth studies on the physico-chemical properties of the Bir M'Cherga and Tajerouine clay may be envisaged.

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