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Investigating Production Technology and Raw Material Sourcing in 17th-Century Bricks of Ghar El Melh Fortress (Tunisia) Through Geoarchaeological Analytical Techniques

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ABSTRACT

This study aims to characterise the historical bricks used in the construction of the fortress of Ghar El Melh "El Borj El Loutani", dating back to the 17th century, with the focus on identifying the raw materials used in their manufacture. Understanding these materials is not only of historical significance but also critical for future restoration efforts, ensuring compatibility with the original bricks. Petrographic, mineralogical, chemical, and petrophysical analyses were conducted on three historical brick varieties. Additionally, granulometric, mineralogical, and chemical analyses were also performed on seven clays sourced from local outcrops. Results show that bricks share a similar mineralogical composition characterized by the presence of quartz and neoformed minerals (gehlenite, diopside, and anorthite), indicating the presence of a significant amount of carbonate in the initial mixture and leading to deduce that the firing temperature ranges from 800 up to 900°C. Analysis confirm that the raw material used for manufacturing the studied bricks originates from Raf-Raf formation. Notably, quartz grains are not included as a degreasing agent but are integral to the raw material itself. In summary, this research underscores the significance of geological techniques in identifying the raw materials and elucidating the brick manufacturing technology employed during the 17th century in Tunisia. These findings offer valuable insights for preservation and restoration projects.

Keywords: Mediterranean Architectural Heritage, 17th-Century Bricks, Firing Temperature, Clays Minerals, Petrophysical Characterisation, Polarizing Microscope, XRD.

INTRODUCTION

Historic buildings are valued for their artistic, architectural, and historical significance. They not only define our identity but also embody our nation's heritage and cultural wealth. These structures play a vital role in our spiritual and educational lives, contributing to cultural and economic development by attracting visitors and tourists. Sadly, ageing, inadequate preservation, human interventions, wars, and natural disasters pose significant threats to these monuments leading to their deterioration and sometimes their complete loss.

To safeguard our heritage for current and future generations, it is imperative to protect these historical monuments and halt their degradation through preservation and restoration efforts. One of the fundamental building materials used in such monuments is bricks. The use of bricks dates back to the dawn of civilization, with fired bricks appearing around 3500 BC (Campell, 2005). They spread across civilizations, influencing architectural styles from Mesopotamia to the Far East and, subsequently, to North Africa and the Iberian Peninsula (Perez-Monserrat et al., 2017).

In the Tunisian context, the utilization of bricks dates back to the Roman period, and their presence is ubiquitous in Islamic, Andalusian, and Ottoman heritage. However, these bricks have experienced varying degrees of decay, influenced by factors such as exposure, orientation, raw material quality, and firing processes. Understanding the causes of this decay is crucial before undertaking restoration works and replacing deteriorated original pieces with compatible and durable materials.

In Tunisia, the scientific conservation of historical monuments is still in its early stages, with several researchers dedicating their efforts to preservation (Ben Ouezdou et al., 2006; Labiadh et al., 2006; Zoghlami & Gómez-Gras, 2009; Zoghlami et al., 2004, 2005, 2016, and 2017; Zornoza-Indart, 2016-2018).

This paper focuses on Borj El Loutani Fortress (Figure 1), constructed during the Ottoman Empire's rule in Ghar El Melh (also known as Porto Farina), Tunisia, under the leadership of Mouradite Hamuda Pacha (1631-1666) as documented by Jalloul in 1999. This fortress is one element within a larger architectural triad of fortifications. Ghar el Meleh, located within the vicinity of the city of Bizerte occupies a strategic coastal location approximately 60 km northwest of Tunis (Figure 2), has attracted various civilizations over the centuries, making it a site of historical and archaeological significance (Chelbi, Paskoff, & Trousset, 1995). Notably, in 1922, recognition of the historical and architectural significance of the three Ottoman fortresses, along with the old arsenal harbor, culminated in their designation as Tunisian Historic Monuments. This formal recognition underscores their importance as cultural heritage sites in the region. Nevertheless, the architectural complex has witnessed substantial transformations and degradation throughout its history. Notably, Borj El Loutani Fortress, owing to its role as a civil prison. From the late 1960s until 1990, The Fortress experienced a period of abandonment. It was during the early 1990s that the Tunisian government launched an extensive enhancement and restoration program for national monuments, including this architectural complex (Ben Ouezdou et al., 2006). Subsequent restoration efforts followed, but regrettably, documentation of these interventions is lacking. Despite their considerable historical, social, and economic significance, the construction materials have not been thoroughly investigated until now. Inside the Ghar El Melh fortresses, three distinct types of bricks are discernible: red, yellow, and light-yellow bricks. Notably, the red bricks are the most susceptible to decay.

Two types of mortar were employed in the construction. The first is a lime-based mortar, skillfully used to bond the limestone blocks together, while the second comprises plaster mortar, specifically utilized to connect the bricks (Figure 2).

The building materials employed in these fortresses exhibit varying deterioration patterns, primarily attributed to the presence of soluble salts and the use of incompatible mortars in relation to the substrate, be it stone or bricks (Figure 2). It is worth noting that the most recent restoration endeavor, as reported by Zoghlami and Ben Lagha (2018), proved to be ineffective. The substitute bricks used in this restoration displayed severe decay over a relatively short period, in stark contrast to the original bricks, which are comparatively less damaged and, in some instances, remain intact.

Due to limited historical information, this study aims to identify the raw materials used in the manufacturing of the bricks, characterize their petrophysical properties, and deduce firing temperatures. This information, besides its historical significance, is crucial for producing custom-made replacement bricks resistant to decay agents prevalent around the monument and harmoniously compatible with the original building materials. Raw material sourcing is a vibrant research area, focusing on identifying the origins of fine-grained deposits used in pottery (Xanthopoulou, Iliopoulos, & Liritzis, 2020), bricks, and construction mortars. Such investigations involve petrological, mineralogical, and geochemical analyses. The manufacturing procedures and technologies employed also profoundly affect time-induced decay.

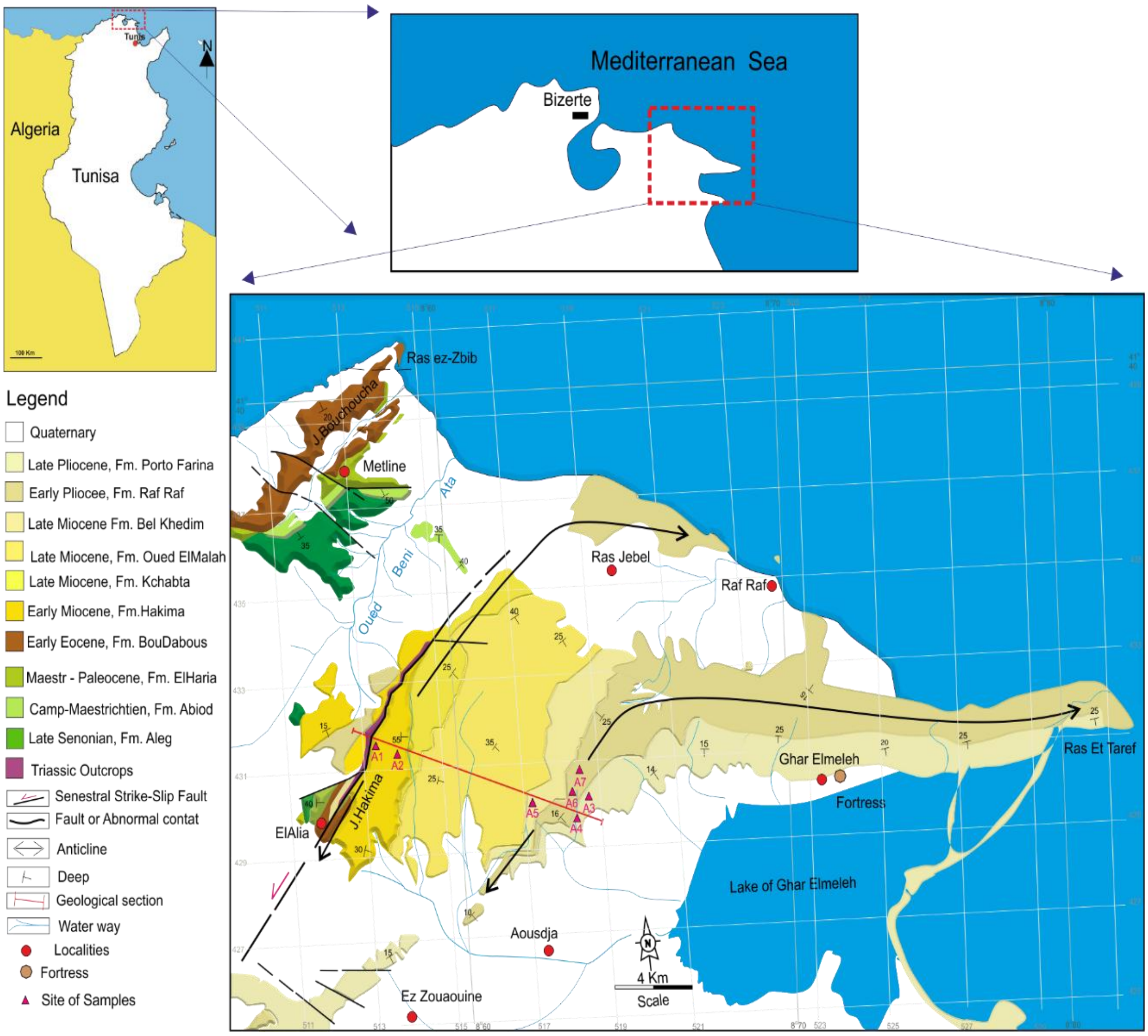

Figure 1. Geographic Localization and Geological Map of the Ghar El Melh Region, Illustrating Various Formations and Indicating the Location of Samples and the Geological Section

The composition of aggregates in pottery sherds and bricks can be readily identified through thin-section analysis, employing traditional petrological techniques (Gonzales, Arakawa, & Koenig, 2015). Mineralogical and geochemical analyses are crucial steps in discerning the raw materials utilized in brick production (Owens et al., 2016; Perez-Monserrat et al., 2017; Atlihan, Koralay, & Sahiner, 2018; Han, Pei, & Liu, 2022).

It is imperative to acknowledge that the final characteristics and resistance to decay of these materials are profoundly influenced by various factors. These include the initial mixture composition, heating rate, firing conditions, kiln temperatures, and whether oxidizing or reducing reactions predominate (Maggetti, 1982; Fort et al., 2004; Maritan et al., 2006, Lourenço, van Hees, Fernandes, & Lubelli, 2014; Xanthopoulou et al., 2020). Furthermore, manufacturing processes and technologies employed in ceramic materials have a substantial impact on their susceptibility to time-induced decay (El-Gohary & Al-Naddaf, 2009; López-Arce, Garcia-Guinea, Gracia, & Obis, 2003; El-Midany & Mahmoud, 2015; Illampas & Ioannou, 2023).

This study aims to analyze the petrophysical properties and the durability of the historical bricks of the Ghar El Melh fortress. Through this analysis, we seek to establish guidelines for minimizing decay and, when necessary, manufacturing new bricks for replacement units.

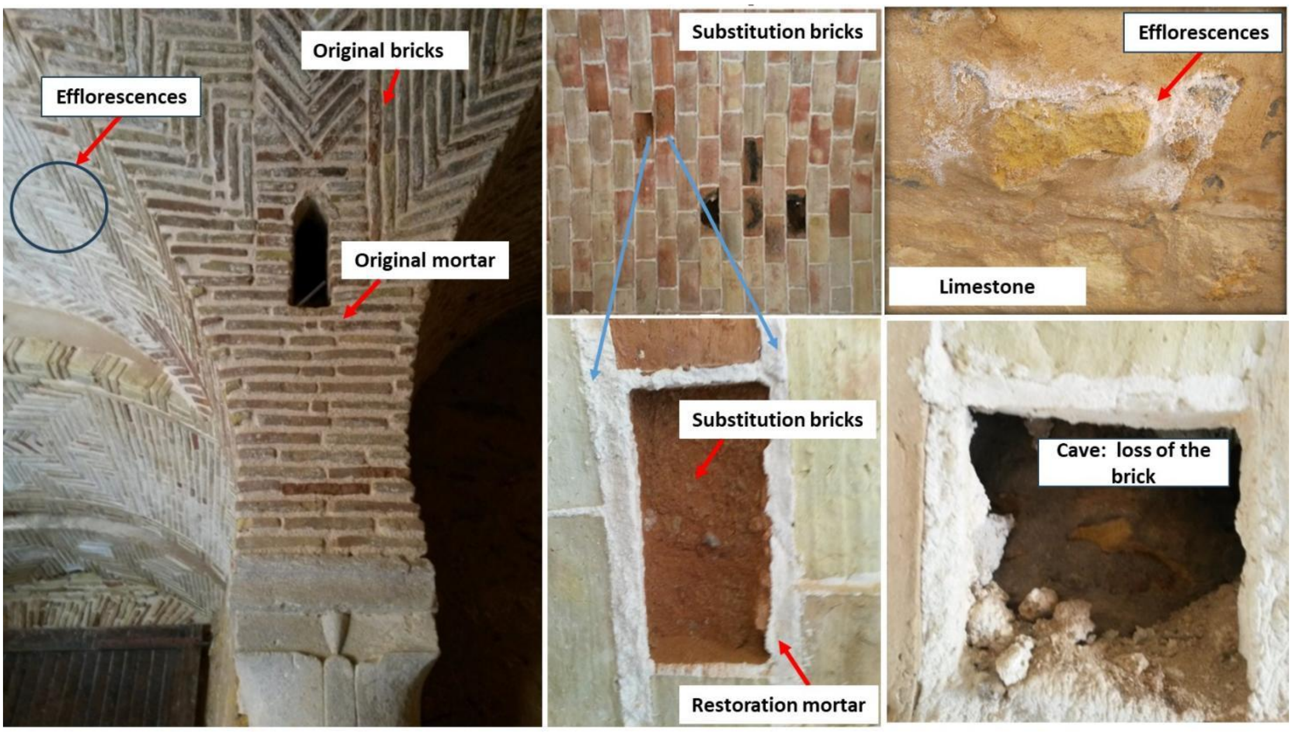

Figure 2. Building Materials and Decay Patterns

LITERATURE REVIEW

Geological Setting

The study area is part of the Easterner North of Tunisia. It is limited geographically by the following UTM coordinates: X = 592100; X = 615500; and Y = 4115500; Y = 4135500. This includes a portion of the Metline and Porto Farina geological maps. Delegations from Metline, El Alia, Ras El Jebal, Aousdja, Raf Raf, Ghar El Melh, and Utica are included (Figure 1). The study area is part of the large structural edifice of northern Tunisia, which is the Neogene basin of Kechabta, which itself constitutes the dip, towards the north-east, of the diapir zone (Perthuisot, 1974). The tectonic structures of the Neogene Kechabta basin have been the subject of several geological (sedimentological and structural) and geophysical works whose objectives are the geometric characterization of the tectonic structures (mainly folded) as well as the understanding of the roles of major faults in the Mesozoic structural evolution of the Kechabta basin (Ennabli, 1980; Haj Ltaief, 1995; Kacem, 2004; Melki et al., 2011; Mejri, 2012; Ramzi, & Lassaad, 2017).

From a sedimentological point of view, the series that outcrops in the study area extends from the Triassic to the Quaternary. These outcrops are dominated by sedimentary series of the Mio-Pliocene to Quaternary age. The study area of Ghar El Melh is formed essentially by a Neogene series about 4000 m thick, represented mostly by clay, sandstone, and gypsum (Figure 1). The nature of the outcrops as well as their lithostratigraphic facies have been extensively studied by Burollet (1951). The Miocene consists of a 525 m thick variegated detrital series of the Hakima formation made up of conglomerate layers at the base surmounted by consolidated ochre-colored sandstone very rich in iron oxides and red/brown silty marls with gypsum intercalations towards the top. The lower part of the Miocene series shows a gap at the outcrop, which can be explained by the shoaling position during the western Mediterranean extension. (Rehault, Boillot, & Mauffret, 1984; Yaich, Durlet, & Renard, 2000; Brahim et al., 2002; Bouaziz, Barrier, Soussi, Turki, & Zouari, 2002). In detail:

Hakima Formation: This formation consists of sands, sandstones, and marls with varied colors, reaching over 500 meters in thickness. It is prominent at Jebel Kechabta and Jebel Hakima. The base comprises alternating layers of sand, clay, and pudding, while the upper part includes coarse polygenic conglomerates, red marls, and coarse sandstones (Mejri, 2012).

Oued El Melh Formation: This formation is over 630 meters thick and primarily composed of grey clays and marls rich in gypsum. It corresponds to the Lower Tortonian and represents a transgressive lagoon or Sebkha system associated with subsidence (Kacem, 2004; El Euch-El Koundi, 2007).

Kechabta Formation: This formation features alternating clay-sandstone and clay-sandy layers, often exceeding 2000 meters in thickness. It is mainly located at Jebel Kechabta and is attributed to the Upper Tortonian. The base includes clayey complexes with sandstones, followed by sandstone layers with galena cement. The rest of the formation alternates between grey clayey levels and yellowish sandstones (Burollet, 1951).

Oued Bel Khédim Formation: This formation spans 483 meters and primarily consists of alternating grey and black clays rich in gypsum. It represents the Messinian age and exhibits lagoonal and lacustrine facies, with basal lagoon facies transitioning to lacustrine facies towards the summit (El Euch-El Koundi, 2007; Burollet, 1951).

The Pliocene is composed of two formations: the Raf Raf formation of the lower Pliocene age and the Porto Farina formation of the upper Pliocene age (Beseme & Lajmi, 1985; G. Bizon, J. J. Bizon, Burollet, & Jelisejeff, 1980).

The Raf Raf Formation includes grey and greenish clayey marls with sporadic yellowish indurated sandstone layers. Towards the upper layers, marls become increasingly saliferous. It exhibits varying thicknesses, such as 966 meters in the Utique exploration drilling well and 133 meters south of Jebel Kechabta. North of Raf Raf, the series features bioclastic marly deposits, a conglomeratic base, and lagoonal deposits at the top (Kacem, 2004; Burollet, 1951).

The Porto Farina formation outcrops at Ghar El Melh, Jebel El Nadhour, and Jebel Ed Demina to the east of Sidi Ali el Makki. This formation shows a yellowish sandstone dominance and is notable for its rich fossil content. It begins with a marine level formed by marl-clay sands overlain by a conglomeratic and sandy continental level (Burollet, 1951). The Porto Farina sandstones represent a significant detrital series that can reach a thickness of up to 504 meters (Melki et al., 2011; Harrab, Mannai-Tayech, Rabhi, & Zargouni, 2013).

METHODOLOGY

Fieldwork (Sampling)

Bricks dating back to the 17th century were sampled from the Ottoman fortress "El Borj El Loutani". Sampling was based on color difference and degree of damage. Three distinct varieties were identified: red (BR), yellow (BJ1), and light-yellow (BJ2) each exhibiting different degrees of decay. A total of three samples were collected for each brick variety.

Additionally, seven marl samples were obtained from various outcrops within the Ghar El Melh region (Figure 1). A stratigraphic log (Figure 5) and geological section were meticulously prepared to elucidate the arrangement of different geological formations in the area. This information was invaluable for pinpointing sampling locations and determining the source of raw materials used in the production of the examined bricks.

Experiments

Petrographic, mineralogical, and chemical analyses were conducted separately on both brick and clay samples, with additional assessment of the petrophysical behavior of the examined bricks.

Mineralogical analysis was carried out by X-ray powder diffraction (XRD) for both brick and clay samples previously grinded allowing the identification of the mineralogical composition. The analysis was carried out using a PANalytical X'Pert PRO X-ray diffractometer with an automatic divergence slit, a spinner, an X'celator, and a CuKα radiation at a scan speed of 0.011 2y/s. The acceleration power applied was 40 kV, with a current of 40 mA. XRD analysis of the clay fraction (i.e., the fraction with grain size < 2 μm) was studied by using oriented aggregates (air-dried, ethylene-glycol, and 2-hour heated slide at 550 ℃) to specify the clay minerals of the raw material. The diffraction data were processed by the X'pert High Score Plus program.

Chemical analyses were carried out using X-ray fluorescence spectrometry (XRF) with a wavelength dispersion Thermo Fisher Scientific ARL™ 9900 Simultaneous-Sequential XRF Series (Bizerte cement Plant), in order to identify the major elements, expressed as a percentage by weight abundant chemical oxides (SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, K_2O, SO_3, and P_2O_5). Samples were firstly dried in an oven at 105℃, then ground until a powder with a particle size less than 2 μm was obtained. The obtained powders were also dried at 500–550℃ for 24 h to remove the organic compounds. The powders were prepared by pressing the tablets on boric acid support for XRF analysis.

A petrographic study was performed using an optical polarizing microscope following the standard test UNE-EN 12407 (2007) to identify compositional and textural variations among brick varieties.

Petrophysical behavior was assessed by water absorption and transfer tests through porous media. Compressive and flexural strength tests were carried out to assess mechanical properties.

The hydric and mechanical behavior of the bricks was performed on prism-shaped samples (3×3×6 cm) obtained from the historical fortress.

(I) Water absorption under vacuum to determine the total open porosity ρ(%) which is calculated with the formula ρ=(M3-M1/M3-M2)×100, where M1: dry mass in grams, M2: is saturated mass in grams, and M3: water mass in grams, following the standard tests (NF EN 1963 B 10-615) and (NF B 10-503);

(II) Capillarity water absorption, which allows determining the capillary absorption coefficient C indicative of the water suction rate, as it occurs in areas of the buildings in contact with the ground, and which is defined with the formula C =(M/S×√t)×100, with M: the total mass of water in grams absorbed by the brick since the beginning of the immersion, S: section of the immersed face expressed in cm2, and t: total time in minutes elapsed since the beginning of the immersion according to standard test (NF B 10-502).

(III) Compressive strength was tested and calculated with the formula R=F/S, where R: compressive strength expressed in mega Pascals, F: maximum load supported by the specimen in Newton's, and S: the cross-section of the specimen (s=a*b) in square millimeters, according to the standard NT 21.317(2008);

(IV) Flexural strength was evaluated on prism-shaped samples (3×6×18 cm), which is calculated with the formula $Rtf=3Fl/2bh^2$ following the standard test NT 21.316 (2007). A granulometric study of the raw material was conducted by wet sieving and laser granulometry (ANALYSETTE 22 Nano Tec (FRITSCH)).

RESULTS

Brick Characterization

Petrographic, Mineralogical, and Chemical Characterization

Microscopic observation shows that red bricks (BR)are made up of 25% of well-sorted grains composed of fine-grained (0,031 a 0, 25 mm) monocrystalline quartz grains (5%), bioclasts (10%) planktonic foraminifera and micritic grains (10%). The porosity is high (25-30 %), partially filled by calcite and gypsum cements. The matrix is not vitrified and constitutes about (60%), the brownish color (Figure 3) together with the presence of bio-clasts and micritic grains indicates that the firing temperature cannot exceed 800°C. Type BJ2 also has a non-vitrified matrix. However, bioclasts are not observed. The porosity is higher than the BR type containing a moldic porosity corresponding to the oxidation of organic matter (straw).

Type BJ1 has the most vitrified matrix and the highest porosity and is partially filled with gypsum cement. The percentage of quartz grains is 10% less than in the first two types of bricks.

The mineralogical components of the three varieties of bricks (red brick BR, yellow brick BJ1, and light-yellow brick BJ2) were determined by XRD (Figure 4). Results reveal that the studied bricks have a similar mineralogical composition. Quartz (SiO_2), calcite ($CaCO_3$), gehlenite (Ca_2Al [$AlSiO_7$]), and gypsum ($CaSO_4$ $2H_2O$) are present in all samples but in different proportions, while anorthite ($CaAl_2Si_2O_8$) is only present in BJ1 and diopside ($CaMgSi_2O_6$) lack in BR.

Chemical analysis (Table 1) shows that the bricks have a similar composition with a high fraction of silicon oxide SiO_2 (37 to 39%), calcium oxide CaO (18 to 21. 4%), and aluminum oxide Al_2O_3 (11 to 12 %). The SiO_2/Al_2O_3 ratio is similar for the three varieties of bricks ranging from 3.13 to 3.32. It seems evident that the main chemical components of the clay used in the manufacture of these bricks are relatively uniform. Bricks present a high loss of ignition ranging from 8 to 12%, which is due to the presence of gypsum and precipitation (recrystallization) calcite.

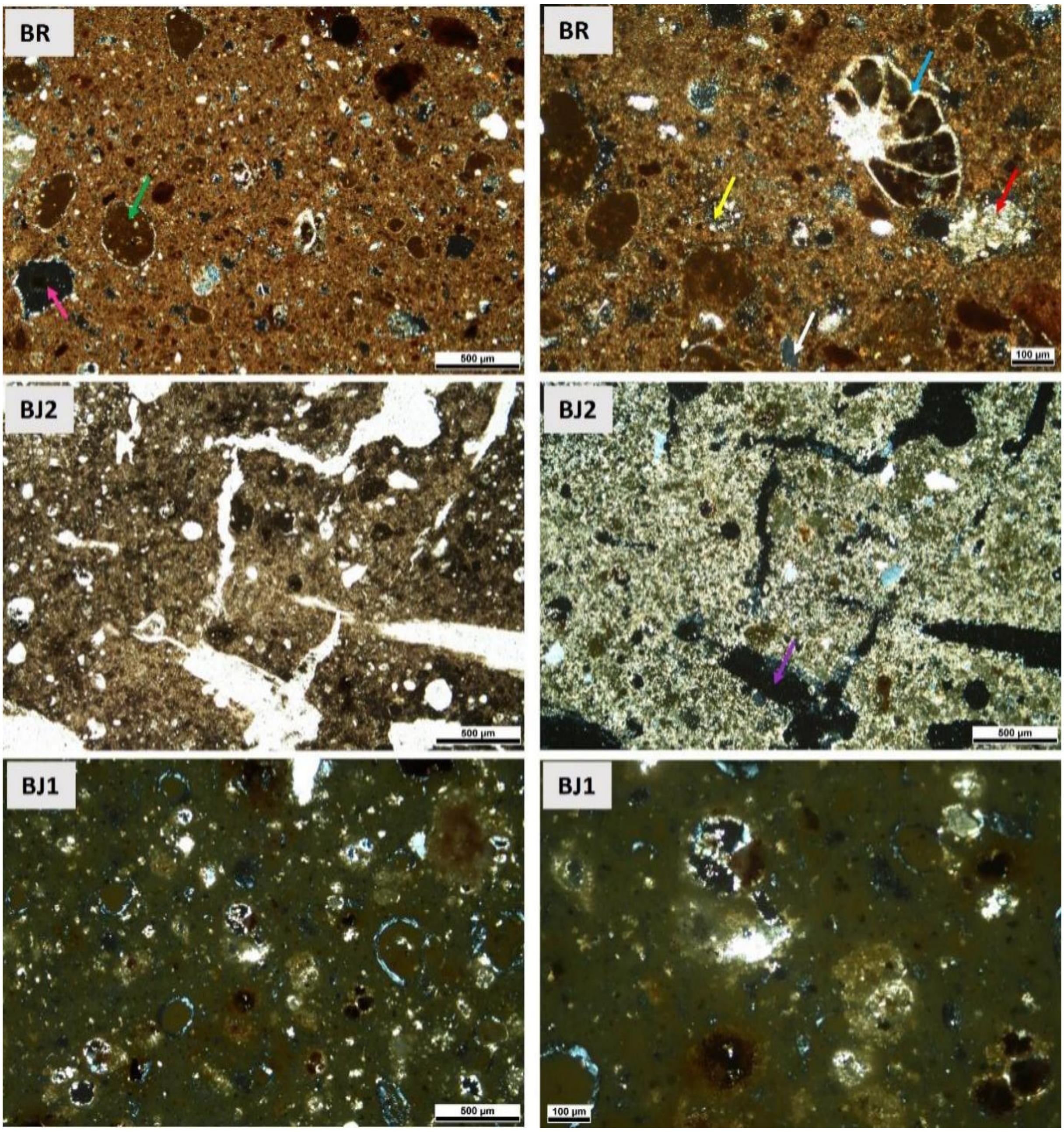

Figure 3. Micro-photomicrographs of the studied Bricks Illustrating Key Petrographic Features. White Arrow: Quartz, Yellow Arrow: Gypsum, Red Arrow: Recrystallization Calcite, Pink Arrow: Primary Porosity, Green Arrow: Micritic Grain, Blue Arrow: Planktonic Foraminifera, Purple Arrow: Secondary Porosity

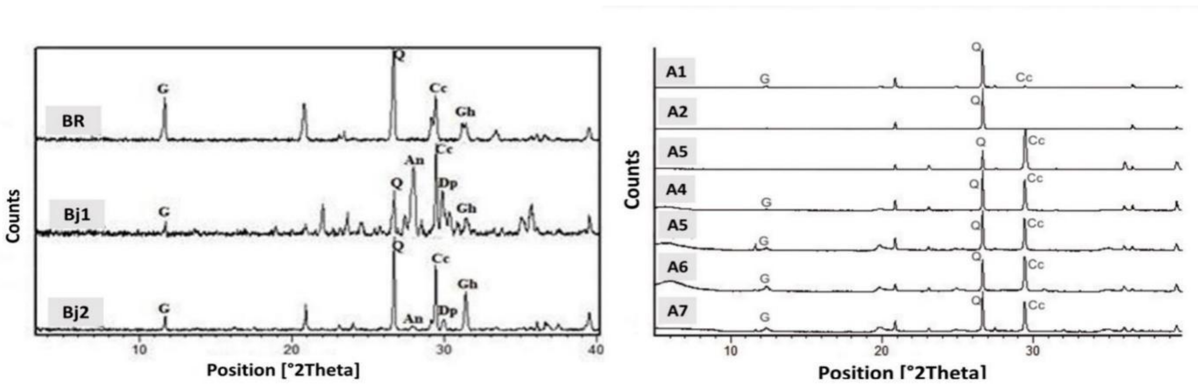

Figure 4. X-Ray Diffraction Patterns of the Studied Bricks and Clays. Q: Quartz, G: Gypsum, An: Anorthite, CC: Calcite; Gh: Geh-Lenite; Dp: Diopside

Petrophysical Characterization

The studied bricks show high porosity percentages, ranging between 20 and 36 % (Table 2). Indeed, porosity is an important parameter due to its influence on other properties such as chemical reactivity, mechanical strength, durability, and quality of the brick (Fort et al., 2007; P. M. Velasco, Ortíz, Giró, & L. M. Velasco, 2014; Antonelli, Cancelliere, & Lazzarini, 2002; Zoghlami and Benlagha, 2018). The capillary water absorption test reveals that the suction degree of the red (BR) and light yellow (BJ2) bricks is high. Nevertheless, the capillary coefficient of the yellow bricks (BJ1) is low, which is consistent with the porosity values. Mechanical tests (Table 2) show that the studied bricks have a low compressive strength. Indeed, the quality of the bricks, both in terms of strength and durability, increases with the decrease of the porosity. In addition, the results obtained by the flexural strength prove that the red bricks are the most resistant to bending (12.19 MPa), more than the yellow bricks (10.44 MPa) and the light-yellow bricks are the least resistant (5.62 MPa), as shown in Table 2.

Origin of the Raw Material

Field Results

The studied section was taken from the region of Ghar El Melh, it is NW-SE direction, and subdivided into several geological formations ranging from Triassic to Quaternary times as shown in Figure 1 and Figure 5. The geological field section and stratigraphic log served to localize samples and to estimate outcrop extension to identify the raw material used in the manufacture of the studied bricks (Figures 5 and Figure 6).

Table 1. Bricks and Clays Chemical Results

Samples	Loss en ignition	SiO_2	Al_2O3	Fe_2O_3	CaO	MgO	K_2O	SO_3	P_2O_5	SiO_2/Al_2O_3	Al_2O_3/Fe_2O_3
A1	8.20	56.88	15.10	6.52	9.85	1.70	0.42	0.15	0	3.77	2.32
A2	2.94	87.30	7.88	5.03	8.79	0.57	0.16	0.18	0	11.08	1.57
A3	33.19	23.13	4.34	2.66	39.57	1.06	0.06	0.25	0.05	5.33	1.63
A4	21.90	38.11	9.97	4.78	24.26	2.37	0.22	0	0.04	3.2	2.09
A5	14.34	44.87	12.84	6.92	15.14	3.43	0.40	2.33	0.08	3.49	1.86
A6	17.62	40.40	12.58	6.01	18.21	3.66	0.25	5.03	0.18	3.21	2.09
A7	16.10	42.54	13.38	6.52	15.53	3.35	0.48	3.57	0.06	3.18	2.05
BR	12.86	38.43	12.03	6.08	18.71	2.55	1.00	1.76	0.19	3.19	1.98
BJ1	8.44	39.02	12.45	6.11	21.22	2.56	0.84	1.57	0.21	3.13	2.04
BJ2	10.96	37.40	11.28	4.70	21.50	2.44	0.44	0.18	0.24	3.32	2.40

Table 2. Hydric and Mechanical Parameters of the Studied Bricks

Samples	BR	BJ1	BJ2
Porosity (%)	28.51 (±1,05)	20.57 (±3,64)	36.75 (±4,06)
C (g/m2.min1/2)	2.78 (± 0,25)	1.69 (± 0,69)	3.04 (± 0,31)
Compressive strength (MPa)	0.8 (±0,07)	1.17 (±0,13)	0.60 (±0,31)
Flexural strength (MPa)	12.19 (± 1,17)	10.44 (± 1,55)	5.62 (± 1,21)

Experiment Results

According to Table 3, samples A2 and A3 are coarse sediments (56-65% > 63µm), while samples A1, A4, A5, A6, and A7 have a percentage of fine fraction ranging between 83 and 99%. Indeed, the percentages of silts are the highest compared to the percentages of the clay fraction in most samples. So A2 and A3 representing Hakima and Porto Farina formations cannot be the materials used for the fabrication of the studied bricks.

Table 3. Granulometric Analysis Results of Sampled Clays

	Coarse fraction (%)	Fine fraction (%)	
Samples	Ø > 63µm (sands)	63 > Ø >2µm(silts)	Ø < 2µm (clays)
A1	13	57	30
A2	56	22	22
A3	65	27	8
A4	17	76	7
A5	4	77	19
A6	4	92	4
A7	1	85	14

The mineralogical characterization by XRD of the sampled clay shows that they are formed essentially of quartz and calcite. Gypsum is present in a minor quantity (Figure 4).

The study of the clay minerals by oriented aggregation shows that they mostly consist of kaolinites, illites, and smectites as shown in Figure 7.

Chemical analysis

All clays present a high amount of SiO_2 and a low amount of Al_2O_3 (Table 4). The SiO_2/Al_2O_3 ratio is very high for all samples. Nevertheless, in the case of A6 and A7, it is very close to the values of the SiO_2/Al2O_3 ratio obtained in the bricks (Table 1).

Except for A2, clays present a high ignition loss ranging from 8 to 33%, which is probably related to coordinated water, the presence of clay minerals, hydroxides, and organic matter.

DISCUSSION

The petrophysical study reveals that the studied bricks exhibit a high porosity ranging from 20% to 30%. This porosity is closely associated with their capillary absorption rate and mechanical properties. Despite having a relatively low resistance to compressive strength, these bricks display significant resistance to bending, which may explain their durability in an environment contaminated by soluble salts.

The observed high porosity can be attributed to various factors such as dissolution or oxidation phenomena (Kostova & Mihaylova, 2023). It is well-documented that decay often leads to increased porosity due to or the formation of cracks (Dufourni, 2014; P. M. Velasco et al., 2014; Zoghlami et al., 2016).

Additionally, the presence of quartz, as confirmed by mineralogical and chemical analyses in this study, plays a role. Peters and Iberg (1978) have demonstrated that a high percentage of quartz can result in fissures during the cooling process due to the volume reduction of quartz as it transitions from form B (high temperature) to alpha form (low temperature). Furthermore, the carbonate content in the bricks can contribute to the formation of cracks and pores with diameters less than 1μm when the bricks are fired within the temperature range of 800℃ to 1000℃ (Cultrone et al., 2004). This phenomenon underscores the impact of carbonate-rich initial mixtures on the firing process.

Mineralogical and chemical results confirm that bricks are manufactured from the same raw material. Indeed, X-ray diffraction (XRD) of the bricks reveals a uniform mineralogical composition characterized by the presence of quartz and high-temperature minerals such as anorthite, gehlenite, and diopside. These minerals are known to form during the firing process, indicating the initial mixture's richness in carbonates (Lopez-Arce et al., 2003; Cardiano et al., 2004; Fort et al., 2007). Importantly, the mineralogical and chemical analysis of the raw materials (clay samples from local outcrops) confirms that all samples consist of calcite minerals, suggesting that carbonates are a natural component of the raw material rather than an additive. Moreover, the significant abundance and intense peak of quartz in these samples indicate its presence as a primary component (A. K. Mishra & A. Mishra, 2021). The presence of diopside in BJ2 and diopside and anorthite in BJ1 further confirms that the three brick varieties were fired at different temperatures. The coexistence of calcite and gehlenite in BR indicates a firing temperature range between 800–850°C, as not all the calcite has converted to gehlenite. This phenomenon may be attributed to factors such as the size of bioclasts and micritic grains and/or a brief firing duration (A. K. Mishra & A. Mishra, 2021). In contrast, both the yellow (BJ1) and light-yellow (BJ2) bricks were fired at temperatures exceeding 900°C, with BJ1 experiencing the highest firing temperature as evidenced by the presence of anorthite and its notably low porosity rate (Viani, Cultrone, Sotiriadis, Ševčík, & Šašek, 2018). Nevertheless, it is important to note that quartz is resistant up to 1000℃–1100℃ (Papadopoulou, Lalia-Kantouri, Kantiranis, & Stratis, 2006; Aras & Kiliç, 2017). This mineral still exists in the studied samples which define firing below this temperature.

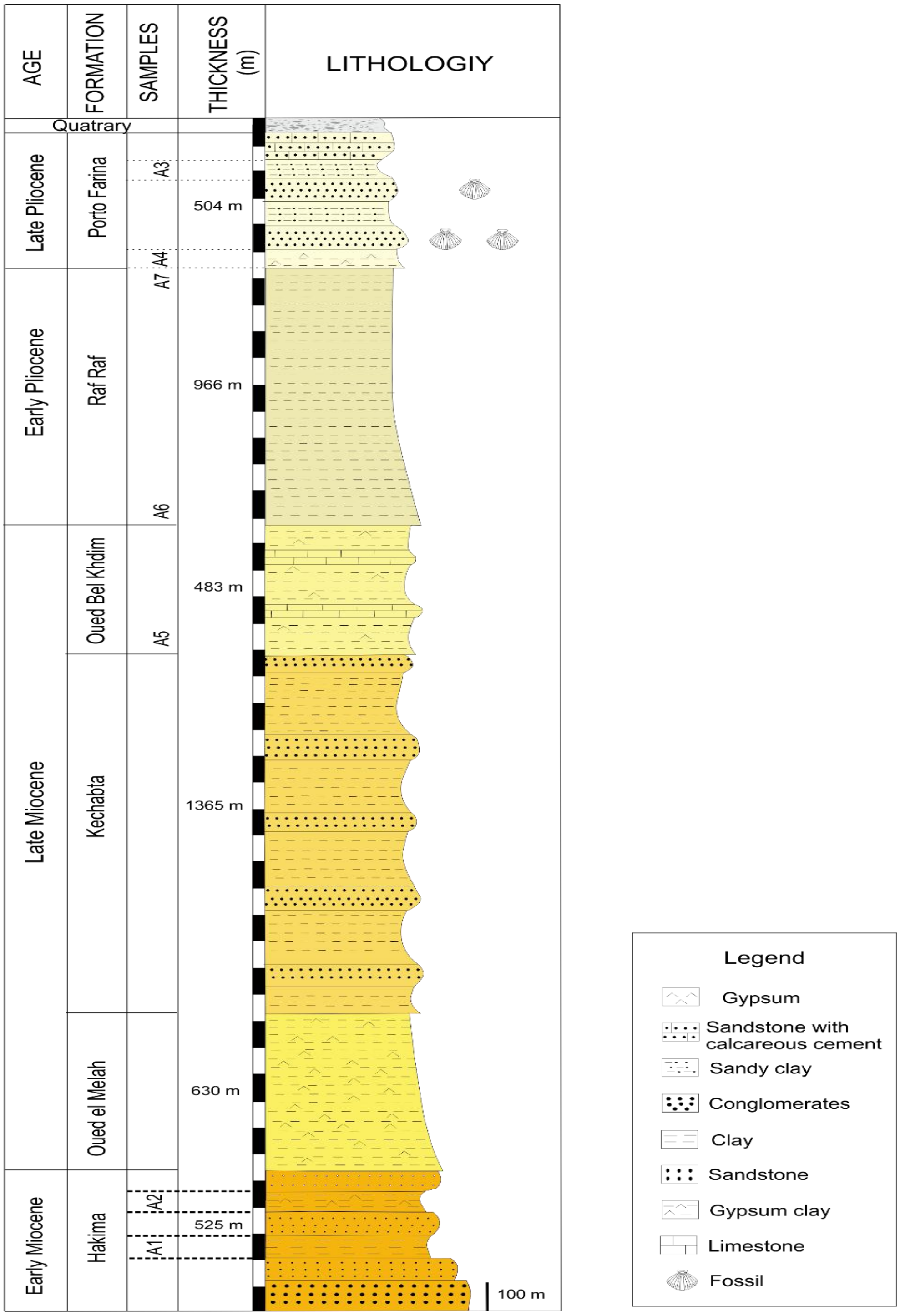

Figure 5. A Stratigraphic Log Carried Out in the Northeastern Part of Ghar El Melh

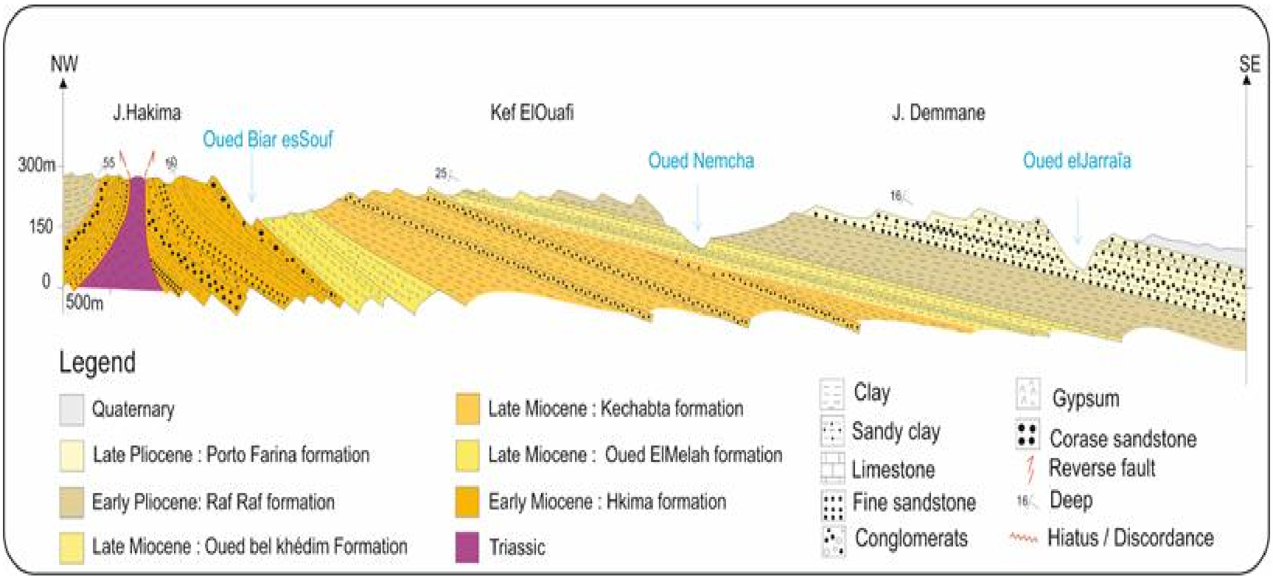

Figure 6. Geological Succession Cropping Out in the Ghar El Melh Region

In addition, the variation in the color of the bricks may be attributed to separate firings in different environments. Specifically, the red bricks (BR) were fired at temperatures ranging between 800°to 850°C in an oxidizing atmosphere, whereas the yellow (BJ1) and light-yellow bricks (BJ2) underwent firing at temperatures ranging between 900°and 1000°C in a reducing atmosphere. Furthermore, it has been established that a heterogeneous firing process can lead to temperature differences of up to 300°C within the same oven, even when using the same original dough. Such variations can result in the appearance of different minerals, as observed in this case (Peters & Iberg, 1978; Gargiulio Riccio, Fontan, Tollon, & Fortuné, 2001). The presence of gypsum and carbonates identified in the composition of the bricks through X-ray diffraction (XRD) and chemical analysis is indicative of recrystallization minerals. The presence of gypsum likely results from the dissolution of gypsum mortar typically used to bond bricks together. Similarly, carbonates are probably products of the dissolution of lime mortar and adjacent carbonate rock ashlars. It's worth noting that these carbonates cannot be provided from the raw material itself, as the firing temperature may have exceeded 900°C, as indicated by the presence of high-temperature minerals.

The granulometric study, conducted on thin sections of bricks, reveals that the quartz ranges from fine-grained sand to silt, constituting a significant percentage of the composition. Furthermore, the granulometric analysis of the raw materials indicates that A2 and A3 samples consist of coarse sediments (with more than 56-65% > 63µm), while A1, A4, A5, A6, and A7 samples contain a fine fraction, accounting for percentages ranging from 83% to 99%. These findings exclude the upper level of the Hakima formation and the upper level of the Porto Farina formation as potential sources of the studied bricks' raw materials. This analysis also reaffirms that quartz grains were not intentionally added but rather constitute original components of the raw material. Additionally, miner-alogical analysis using oriented aggregates reveals that the clay minerals mainly consist of kaolinites, with minor proportions of illite and smectite (Figure 7).

From a chemical standpoint, the SiO2/Al2O3 and Al2O3/Fe2O3 ratios in the brick samples closely align with those obtained for A6 and A7 (Table 1). To sum up, and according to granulometric, petrographic, and mineralogical analysis, the possible origin(s) of raw material used for the manufacture of the studied bricks can be A1, A4, A5, A6 and A7. Nevertheless, the kind and extension of outcrops (proximity) (Figure 1) and the presence of bioclasts attested by dissolution type porosity (moldic porosity) make the A6 and A7 clays from Raf formation the most probable raw material used to manufacture the historical bricks.

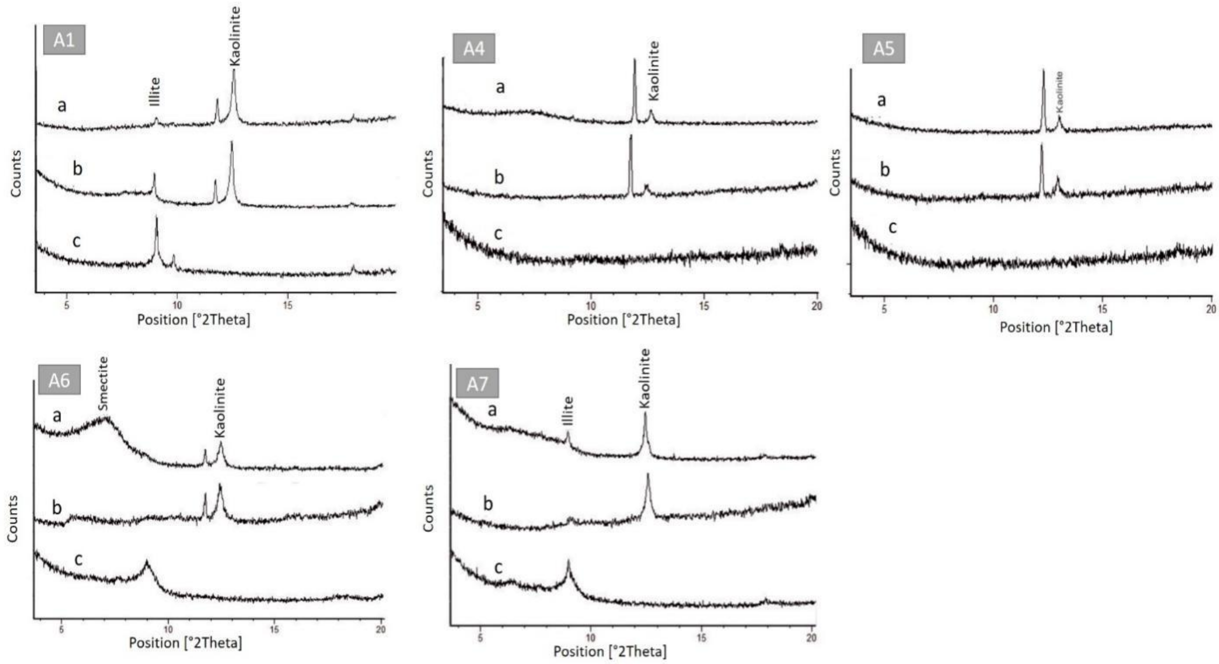

Figure 7. XRD Results of the Clay Mineral Fraction (a) No-Treated Samples; (b) Samples Treated with Ethylene Glycol; (c) Heated Samples.

CONCLUSION

Based on the results obtained from various analyses, it can be inferred that the most probable source(s) of the raw materials used in the construction of the 17th-century for-tress "El Borj El Loutani" is associated with the Raf-Raf formation.

The presence of very fine-grained quartz and its percentage in both the bricks and the clays strongly suggests that quartz was not intentionally introduced as an additional component during the brick-making process but it was naturally present in the clay rock.

Based on the mineralogical and petrophysical analysis of the studied bricks, it can be concluded that the firing temperature ranges between 800℃and 850℃ for brick type BR and between 900℃ and 1000℃ for both brick Types BJ1 and BJ2. This conclusion is supported by the successive appearance of high-temperature minerals.

Thin section observations and mechanical study results consistently demonstrate that BJ1 is the best-fired brick with superior mechanical performance. Thus, for any brick re-placement needs, clays from the Raf-Raf formation should undergo firing at temperatures exceeding 900℃. This will guarantee optimal matrix vitrification and improve mechanical resistance.

The adopted method, which involves the comparison of mineralogical and petrographic characteristics between clay outcropping materials and the final product (bricks), has proven to be highly effective in identifying the raw materials used in the manufacture of historical bricks and determining production technology.

Finally, the characterization of clays and original bricks in this study provides crucial insights to produce replacement bricks that are compatible with the originals and resistances in the current monument environment. Future work will involve a series of laboratory manufacturing tests aimed at defining the parameters of the firing process and determining the appropriate component ratios.

AUTHOR CONTRIBUTIONS

Karima Zoghlami: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing - original draft, Writing – review & editing. Aida Zaddem: Data curation, Formal analysis, Investigation, Resources, Validation, Writing – original draft. Taoufik Haji: Field study, Validation. David Gomez: Formal analysis, Naima Azaeiz: Funding acquisition, Supervision & editing.

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