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The influence of clay composition and lithology on the industrial potential of earthenware

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HIGHLIGHTS

- Earthenware properties of different clay lithologies have been evaluated.
- Argillite exhibits the highest mechanical strength among the studied lithologies.
- The difference between clay fraction and total clay content may depend on lithology.
- Particle size has more influence on mechanical strengths than fluxing agents.
- Various clay lithologies are suitable for construction materials industry.

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ABSTRACT

This paper investigates the influence of clay composition and lithology on the industrial potential of earthenware. The 26 ceramic pastes collected were subjected to particle-size analysis, X-ray diffraction and X-ray fluorescence to investigate their compositions. The shards properties were determined through porosity tests, shrinkage, flexural and compressive strengths. Based on obtained results, all studied ceramic pastes turn out to offer a great potentiality for the earthenware industry despite their varied lithology. Argillite-based pastes exhibit the highest mechanical strengths while colluvium and schist-based pastes exhibit the lowest mechanical strengths. Further, the results have established a reference database for the perpetual restoration work of Marrakech monuments and can help to tracing back the source areas of the archaeological materials of this city.

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1. Introduction

Clays and clay minerals have been widely used as the main component of raw materials in the fabrication of diversified ceramic products [1]. The properties of clays that are of interest to the ceramics industry are their plasticity which facilitates the shape of the body, chemistry, mineral composition, thermal properties, colour, refractoriness and mechanical strength after firing [2].

Because of the geological variability of its basement, Morocco has extensive clay deposits [3]. These clays are being used for earthenware in both for industrial ceramics scale as well as for the traditional production of small-scale ceramic factories. In the Marrakech region (central region of Morocco), the ceramic activity has been known since several centuries. There are still hundreds of small traditional ceramic factories around the city. Each site is located in a specific geological region and this ceramic activity generally uses raw material from nearby geological formations [4]. Except for some recent local work [5–9], clays used in ceramic within the Marrakech area have not been fully characterized. Furthermore recent works were mostly focused on the study of archaeological and historical buildings of the city [10,11]. The characterization of the materials around the city is one of the most important steps, as this helps identifying the original materials of historical buildings and enables to make decisions about restoration [12].

In this study, the characterization of clayey materials in the region of Marrakech aims to determine their composition and the properties of the fired bodies taking into account their lithological

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nature. These knowledges are crucial: 1) for evaluating the impact of clay composition and lithology on the industrial potential; 2) for evaluating their potential suitability as raw materials in other various ceramic applications; 3) to enrich the database of raw materials used for earthenware activity; 4) to contribute to the knowledge of clayey material nature of archaeological materials and historical buildings of the Marrakech imperial city used in their manufacture centuries ago; 5) to help identifying the composition of clays around the city and to use the best of them for restoration.

2. Study areas

The studied area is located in the central part of Morocco between 31° and 32°30′ North and between 7° 30′ and 8° 30′ West. From a geomorphological point of view, this region comprises the north flank of High Atlas, the massifs of Jbilet and Rehamna, and the Haouz and Bahira plains.

On the geological side, the studied area displays a wide variety of facies. The upstream part of the Atlas Mountain range situated at the south of the area, as well as the Northern Jbilet and Rehamna massifs, consists of a Precambrian and Paleozoic basement composed of igneous and metamorphic rocks. In the piedmont of the High Atlas, the basement is covered by epicontinental and marine sediments of Mesozoic and Tertiary age dominated by limestone, clayey sandstones and marls [13]. In the central part of the area, the Haouz plain is composed of Neogene and Quaternary formations dominated by conglomerates, sandstones, siltites and clays. Those detrital formations resulted from the dismantling of the Atlas mountains and are composed by dejection cones and fluvial terraces intimately linked in time and space [14].

Twelve sites of earthenware occur within a range of 10–80 km around the city of Marrakech and are distributed on the three geological areas: 1) Bouchane and Oulad Dlim sites are respectively located near the Paleozoic Rehamna and Jbilet massifs; 2) Saada, Agafay, Tamsloht and Mzouda sites are located in the Haouz plain; 3) the six remaining sites (Tamazouzt, Talatast, Ait Boussaid, Ourika, Amezmiz and Anougal) are situated in the north flank of the High Atlas Mountain. More than 850 potters are spread over about 550 workshops in these sites. According to their firing temperature, porosity and red colour, the ceramic products of the region are classified as coloured earthenware [15].

3. Material and methods

3.1. Material

Depending on their geological situation, each pottery site uses clayey materials from the nearby deposits. Consequently, the raw material used manifests various geological and lithological nature. Globally, we distinguish six types of raw materials: Paleozoic schist, Quaternary soil, Mio-Pliocene and Triassic argillite and colluviums, washing sludges resulting from gravel pits and fine rivers sediments. The most used materials are soils and schists. Argillites and colluviums are used with the same frequency followed by rivers sediments. Whereas the washing sludges is the less used.

The argillites of the region have been affected by different degrees of diagenesis. The argillites of the Ait Bousaid site have undergo a low degree of diagenesis and are in the form of scree fragments of 5 mm diameter globally and are friable by hand pressure. On the contrary the argillites of the sites of Ourika or Mzouda are not friable and require grinding with porcelain mortar to reduce them to powder. The schists form the substratum of the geological formation of almost all sites. They are moved and deposed in fragments form in Mzouda, Anougal and Ourika sites. While for the site of Oulad Dlim and Bouchane, they are extracted directly from the bedrock and require a preliminary mechanical crushing. The soils used at the Agafay and Tamsloht sites are in the form of a cover of plain. In the Bouchane, Amezmiz and Oulad Dlim sites, the soil results from the alteration of schists with some additional components from proximal granitic intrusions [14]. Colluviums are detected on the mountain slopes of the sites of Ourika, Talatast and Mzouda. The colluviums used in the region are characterized by the presence of coarse grains of feldspars and quartz with some clay minerals that provide plasticity. The river (wadi) sediments used in earthenware of the Saada site, are located in small channels formed after flooding events. They are detected by the presence of drying slits, indicating the presence of plastic materials necessary for the shaping process. The sediments of Zaraba are turbiditic muds. They correspond to the filling of the Rocade Canal and are regularly extracted along the canal. The washing sludges used in the Tamazouzt site come from the aggregate production plants. These aggregates, originating from the rivers (wadis), comprise various rocks types and generate washing sludges of wide-ranging compositions.

While some potters use only one type of clay, others are forced to mix two or more types of clays-according to a local know-how, in order to adjust the properties of both the unfired ceramic body and the final product. The soil layers above the schist are generally mixed to them to improve the plasticity and to react as a binding agent. The sand and silt are mixed to argillite. Colluvium naturally contains significant amount of plastic clays. Wide variety of samples are present in Mzouda site, the potters try to mix the plastic raw material with the non-plastic one. The argillites show various degree of diagenesis from one site to another and this affect their properties.

3.2. Sampling

In each earthenware site of Marrakech region, the clay raw materials extracted from one single formation are relatively homogeneous and behave similarly from the point of view of ceramic properties, except sediment from the river (wadis). Therefore, for each studied site, one single raw sample for each type of raw material was collected directly from the potters' stocks or from quarries with local assistance. The pastes composed of a mixture of several raw materials were prepared in the laboratory according to the formula used by the potters.

In total, 34 samples of different clayey raw materials used by the artisans of the twelve studied sites are collected. Those raw clay materials allow preparing 26 pastes. 13 pastes are made up from one single type of raw material (Table 1). While 21 raw materials (Table 1) are mixed to form the remaining 13 pastes: OD = $1/3OD_1 + 1/3OD_2 + 1/3OD_3$; Bo1 = $2/3Bo_1 + 1/3Bo_2$; Bo2 = $1/2Bo_1 + 1/2Bo_2$; Am = $1/2Am_1 + 1/4Am_2 + 1/4Am_3$; An = $1/2An_1 + 1/2An_2$; Mz1 = $3/4Mz_5 + 1/4Mz_6$; Mz3 = $1/3Mz_2 + 2/3Mz_1$; Mz5 = $1/2Mz_4 + 1/2Mz_5$; Our4 = $3/4Our_1 + 1/4Our_2$; Mz2 = $1/2Mz_3 + 1/2Mz_6$; Mz4 = $1/2Mz_4 + 1/2Mz_3$; AB = $1/2AB_1 + 1/2AB_2$; Our5 = $1/2Our_1 + 1/2Our_3$.

The abbreviation used for samples nomenclature is based on the source area of each raw material. For the Saada site particularly, potters use the clay sediments provided from different sources: Zaraba Canal (Zar), N'Fis (Nfs) and Tensift (Ten) rivers.

3.3. Methods

The particle size distribution of the raw samples was determined by wet sieving for the fraction with grain diameter greater than 40 μm. The fraction with grain diameter less than 40 μm was introduced for 24 h into a 100 ml deionized water tank, and was stirred from time to time before being analysed by a Horiba LA-300 laser diffraction analyser.

The identification of the mineralogical phases of raw materials and neoformed phases of fired products were carried out by X-ray diffraction using a Bruker D8-Advance diffractometer with CuKα radiations (scan step size: 0.02°; time/step: 0.6 s; anode: copper with Kα = 1.5418 Å). Qualitative identification was based on the method used by Moore and Reynolds [16]. Semi-quantitative estimation have been described in Cook et al. [17] and in Boski et al. [18] for bulk sample, and in Fagel et al. [19] for clay fraction. The latter was prepared from the fraction <2 μm of decarbonated sediments. After removing carbonates from the crushed rock with 0.2 N HCl, deflocculation of clays was done by successive washing with deionized water and centrifugation at 3000 rpm. The size-fraction <2 mm was separated by sedimentation following Stokes's law, and later placed on glass slides as oriented aggregates. The intensity of each peak was multiplied by a corrective factor. The intensity of the clay fraction was measured on ethylene glycol spectra.

Major elements (Si, Al, Ca, Fe, K; Mg, Mn, Na, Ti, P) were analyzed by X-ray Fluorescence (XRF) on lithium–borate fused glass [20]. The Loss-On-Ignition (LOI: combined water, organic and inorganic matter) were measured on dried samples heated at 950 °C for 2 h [21].

The porosity was determined by the absorption of water [22] in specimens fired at 1050 °C.

The flexural and compressive strength were also carried out on cylindrical and rectangular specimens fabricated respectively by rolling and pressing and then fired at 1050 °C. The machines used are a "Testometric Micro350" for the flexural strength and a "compression machine made in University of liege" (ArGEnCo Laboratory) for the compressive strength, equipped with a worm gear motor NEFF and controlled by a Microstep Drive (Parker) and a compumotor PC23 adaptator.

Part of the experiments was carried out at the University of Cadi Ayyad (particle-size) and other at the University of Liege (mineralogical and chemical compositions).

4. Results

4.1. Particle size distribution

The results show large particle size variations from one sample to another. For the pastes, the clay fraction has variable contents

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