Main parameters influencing the plastic behavior of clays used for traditional ceramics production

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Article Info

Abstract

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This paper investigates the main parameters influencing the plastic behavior of clays used for traditional ceramics production. For this, twenty-six clayey pastes were selected from twelve traditional ceramic plants around the city of Marrakech (Morocco). According to the lithology, six different types of materials are used as raw material in the ceramic industry of this region. Emphasis is placed on the impact of the characteristics of these clayey materials upon the plastic behavior of these clays. The pastes were characterized through their consistency using the Atterberg limits. It has been concluded that the grain size, the mineralogical and the clay mineral composition and content, the effect of diagenesis and the presence of talc-pyrophyllite association play the most important role in the control of the plasticity behavior.

1. Introduction

Plasticity is one of the most important parameters for manufacturing clayey ceramic products. It allows to ceramic paste to be easily workable during shaping process (Idrissi et al., 2021a). Several factors have been suggested in literature to be responsible for the plasticity behavior of soils and sediments. The knowledge of these factors leads to the optimization of the clay deposits used in local or regional ceramic industries (Idrissi et al., 2014, 2021b; Idrissi, 2017; El Ouahabi et al., 2019).

The plasticity level of an hydrated soil is mainly influenced by the clay mineralogy of the material, its particles distribution, the diagenesis level, the organic matter and the presence of amorphous clay-size materials (Hajjaji et al., 2010; Daoudi et al., 2014, 2015; Idrissi et al., 2014). This latter influences differently depending on its content and on the curing period in presence of water (Husein Malkawi et al., 1999). Generally, identifying the respective influences of all these parameters on the plasticity behaviour of the sedimentary material is difficult.

Research in this area has been traditionally focused on the influence of clay minerals on the plasticity properties of soil materials (Bain, 1971; Schmitz et al., 2004; Hajjaji et al., 2010; Idrissi et al., 2015). Dumbleton and West

(1966) studied the links between clay contents and the Atterberg consistency limits of natural soils around the world, trying to determine the contribution of the clay mineral to the engineering properties of soil. Kaolinite, chlorite and illite are considered as clays of low to moderate plasticity (Aiban, 2006)

Smectite clay rich soils are described in engineering as the most active and responsible for swelling problems, and are among the highest plastic clays (Low, 1980; Sridharan and Choudhury, 2002).

Fibrous clays (palygorskite and sepiolite) also manifest very high capacity for sorption (adsorption and absorption) (Galan, 1996) and then influence highly the plasticity behavior of soils. Their plasticity limit is even higher than that of smectite (Daoudi et al., 2015). The other clay species manifest low to moderate plasticity behavior.

The region of Marrakech, subject of this study is known for its geological variability (Idrissi et al., 2016) which consequently generates clay deposits of variable nature and then of variable plasticity. To adjust the properties of unfired ceramic body, some regional ceramic workshops mix different clayey materials. They generally mix coarser materials like schistose grains with finer soil to obtain the desired workability.

Previous studies have discussed the link between the technological choices and other properties (Rye, 1981; Müller et al., 2013, 2014). In this study we will demonstrate that the technological choices for actual and archaeological ceramic materials have also a close link with the plasticity of the ceramic pastes. In this study, the contribution of each parameter on the plasticity behaviour of the ceramic pastes will be pointed. This study aims to present the plasticity behavior of several clay used in Marrakech ceramic industry and to evaluate the main parameters influencing it such as lithology, grain-size, and mineralogy.

2. Studied area

From a geomorphological point of view, the region of Marrakech comprises the north flank of High Atlas, the massifs of Jbilet and Rehamna, and the Haouz and Bahira plains. Briefly, on the geological side, the studied area displays a large sort of facies with ages allocated from Precambrian to Quaternary. In summary, twelve sites of traditional ceramics occur within a spread of 10 to 80 km round the city of Marrakech. Bouchane and Oulad Dlim sites are respectively located near the Paleozoic Rehamna and Jbilet massifs. Whereas Saada, Agafay, Tamsloht and Mzouda sites are located in the Haouz plain. The six other sites (Tamazouzt, Talatast, Ait Boussaid, Ourika, Amez Miz and Anougal) are located in the north flank of the High Atlas Mountain. A detailed description of the

studied area and the studied materials can be found in Idrissi et al. (2018).

3. Material

The studied clay manifests various nature. Globally, we distinguish six forms of raw materials: Paleozoic schist, Quaternary soil, Mio-Pliocene and Triassic argillite and colluviums, washing sludges resulting from gravel pits and fine rivers sediments. In order to adjust the properties of both, the unfired ceramic body and the corresponding final product, the regional ceramic plants mix different clayey materials. Thus, the soil layers above the schist are generally mixed to them to improve the plasticity and to react as a binding agent. However, colluvium contains naturally, a significant quantity of plastic clay. Large choice of clays is 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 plasticity degree (Idrissi et al., 2018).

In total, 26 pastes have been studied, they are used by the potters to manufacture different kind of products. The Table 1 resumes the details of each sample. The abbreviation used for samples nomenclature is based on the source area of each raw material. For the Saada site particularly, potters use the silty clays provided from different sources: Zaraba Canal (Zar), Wadi N'Fis (Nfs) and Wadi Tensift (Ten).

Table 1 : Nature and sources of pastes used to manufacture various products.

Samples	Area	Lithology	Added materials	Color
OD	Oulad Dlim			Brown
Bo1	Bouchane	Schist	Soil	Green
Bo2				
Nfs	Saada	Silty clays	-	Gray
Ten				
Zar				
Ag1	Agafay	Soil		
Ag2				
Tms	Tamsloht			
Mz1	Mzouda	Schist	Sand	Yellow
Mz2		Argillite		Red
Mz3		Colluvium		Orange
Mz4		Argillite	Silt	Red
Mz5		Schist		Gray
Mz6		-		Orange
Tmz1	Tamazouzt	Washing mud	-	Gray
Tmz2				
Ta	Talatast	Colluvium	-	Red
AB	Ait Boussaid			Brown
Our1	Ourika	Argillite	-	Red
Our2				
Our3		Schist	-	Green
Our4		Colluvium	Silty clays	Brown
Our5		Argillite	Colluvium	Red
Am	Amez Miz	Schist	Soil	Brown
An	Anougal		-	Orange

4. Methods

The used methods for X-ray diffraction, X-ray fluorescence spectroscopy and particles size distribution are described in Idrissi et al., (2018). The Atterberg limits was carried out on the fraction of the material passing through the 425 μ sieve. The required weight is about 150 g of the undersize of this sieve.

Liquid limit

The measurement of the liquid limit was executed by a Casagrande apparatus based on the method of Atterberg. The operation consists in spreading a liquid paste in a container so as to obtain a layer of about 1 cm thick in the middle.

The paste is grooved in the cup so as to divide it into two equal parts separated by a V-shaped groove having at bottom of the cup exactly 2 mm wide. The cup is then dropped onto the base by actuating the crank at the rate of 2 strokes per second until the groove is closed over a length of 10 mm. We count the number of blows required to cause the groove in the clay bed (Méthode de Mesure, 1981).

The number of shocks required for closing is noted and about 10 g are sampled in the area where the groove has closed to measure the water content of the paste. The number of shocks must be between 15 and 35 for the measurement to be valid. The operation is repeated on the same paste after retracing the groove to check if the number of strokes remains the same, otherwise it is necessary to knead the paste again on the cup and repeat the operation.

It is necessary to cumulate at least three tests (number of shocks with the corresponding water contents) in order to draw up a curve which makes it possible to deduce the liquidity limit which is the water content corresponding to 25 shocks.

Plastic limit

The plastic limit is determined by rolling a small ball of semi-soft paste (which does not stick to the fingers) to form a clay coil 10-15 cm in length and 3 mm in diameter. After obtaining the clay coil, we put it back into a ball and repeat the coiling clay procedure. The operation is repeated until the paste breaks during winding before reaching 3mm in diameter. At this time, the coil clay is weighed on a small beaker of known mass to calculate the wet mass (Mw). The sample is placed in an oven for 24 hours at 105 °C in order to deduce the dry mass (Md). The water content (%W) is calculated relative to the dry mass with the equation 1:

$$\%W = \frac{(Mw - Md) \times 100}{Md} \quad \text{Eq. 1}$$

The same operation is repeated on another coil clay. The difference between the two water content values

should not be more than 1%. And we take the arithmetic mean of these two values as the plastic limit.

Plasticity index

The plasticity index was measured through the Atterberg limits using the equation 2:

$$PI = LL - PL \quad \text{Eq. 2}$$

Where: PI is the plasticity index, LL is the liquid limit and PL is the plastic limit.

5. Results and Discussion

Plastic behaviour

Plasticity is mainly influenced by the mineralogy of the material, but also by the sand, silt and clay fraction. These latter exhibit a wide range of engineering characteristics. To make terms more useful in engineering practice, the Casagrande's plasticity chart classifies clayey and silty soils according to their behaviour (Holtz and Kovacs, 1981).

The results for consistency limits (i.e., plastic limit [PL], liquid limit [LL]) of the studied samples, and the plasticity indexes (PI) are shown in Table 2. The studied samples are plotted in various areas on the 'Liquid Limit-Plasticity Index' Chart (Figure 1), meaning that these pastes tend to have different engineering behavior. In fact, Holtz and Kovacs (1981) consider that soils in which fine grains do control the engineering behavior are classified according to their plasticity characteristics. Plasticity, swelling capacity and organic matter content are taking a part in this classification (Figure 1). The A-line generally separates the more claylike materials from those that are silty and also the organics from the inorganics. Almost all studied samples are above the A-line, but they have low to high values of plasticity and swelling (32% < LL < 55%) as indicated in the top of the diagram.

The Plasticity index (PI) indicates the presence of two main groups of pastes. The first group is characterized by PI values ranging between 15 and 20. This group comprises principally the pastes composed of schist, washing muds, silty clays and soil. In this group, we can see that the plasticity index and the liquid limit of schist are the lowest (LL $\leq$ 37%). A low liquid limit means that the content of retained water is low, so the paste is less plastic and more stable. For the second group, the PI ranges between 21 and 32. This group comprises principally the pastes composed of colluvium and argillite. All pastes made by colluviums are characterized by high PI (25-27).

The result in Figure 2 reveals that almost all investigated pastes are plotted inside the optimal moulding region, except Ten, Nfs and Our5 that are located inside the acceptable area, while the other are located outside both the acceptable and optimum moulding regions.

Furthermore, most sample could exhibit low shrinkage due to the fact that their plastic limits are relatively low and consequently imply less colloidal and intergranular water responsible of shrinkage.

Table 2: Particle size distribution and consistency limits (%) of the studied samples.

Lithology	Samples	Consistency limits (%)		Plasticity indexes (PI)	Particle size distribution (%)		
		Liquid limit (LL)	Plastic limit (PL)		Clay fraction (<2µm)	Silt fraction (2–20µm)	Sand fraction (>20µm)
Schist	Bo1	34	19	15	18	26	56
	Bo2	35	19	16	20	27	53
	OD	36	21	15	25	23	52
	Am	37	19	18	19	25	56
	An	36	19	17	18	22	60
	Our3	35	18	17	23	12	65
	Mz1	34	17	16	20	26	54
	Mz5	33	15	18	19	39	42
	Mz6	32	16	16	30	27	43
Wadi sediment	Nfs	48	32	16	34	34	32
	Ten	47	30	17	40	53	7
	Zar	48	24	24	62	33	5
Soil	Ag1	38	21	16	50	35	15
	Ag2	39	21	16	51	28	21
	Tms	45	25	20	34	43	23
Argillite	Mz2	34	17	17	38	17	45
	Mz4	37	19	18	40	35	25
	AB	40	22	18	27	12	61
	Our1	52	22	30	66	25	9
	Our2	40	19	21	48	30	22
	Our5	55	23	32	41	31	28
Colluvium	Mz3	43	18	26	25	34	41
	Ta	49	24	25	26	26	48
	Our4	46	19	27	36	27	37
Washing sludges	Tmz1	39	23	16	60	22	18
	Tmz2	39	24	15	64	24	12

Figure 1: Plotted samples on Casagrande Chart (Casagrande, 1948) [after Howard (1977) and Dakshanamurthy and Raman (1973)]. CL: Clay of Low liquid limit. ML: Silt of Low liquid limit.

Figure 2: Studied pastes workability through the Atterberg limits (Bain and Highly, 1978).

Impact of particle size distribution on the plasticity

The particle size distribution results of the studied samples are listed in Table 2. They show wide variation, having clay fraction ($<2 \mu\text{m}$) from 18 to 66%, silt fraction from 12 to 53% and sand fraction from 5 to 65%. In order to understand the influence of particle size distribution on plasticity, we first projected the studied pastes on the Shepard classification diagram (1954) following the relation between sand, silt, and clay (Figure 3). The samples generally plot in the sand-silt-clay, silty sand, silty clay and clayey sand fields. Schist samples have relatively more sandy composition, whereas washing muds, silty clays and soils are relatively more clayey. Samples composed of argillite are characterized by wide variation in particle size and are plotted in various fields on Shepard's diagram. The Shepard's classification does not consider the type of sediment and its origin. The Unified Soil Classification System developed by Casagrande (1948) considers that materials in which silts and clays do not affect the engineering performance are classified according to their grain size characteristics.

The studied samples were previously plotted on the McManus Diagram (2018) to show their degree of permeability and porosity. It has been noticed in that previous study that most of the studied samples have a low porosity and permeability which allow them to have good workability due to the good cohesion between particles. And even the plastic properties of coarse pastes could be improved by better crushing process.

The results obtained in Figure 4 show that there is no obvious relationship between the clay fraction content of the studied sample and their plasticity behavior; samples with approximately the same clay fraction content can display very different plasticity indexes and reversely which is normal because the correlation coefficient is very low ($\text{COR}[X,Y] = 0.25$). Furthermore, the Shepard

diagram shows that the pastes based on schist and colluviums have relatively the same grain size composition; they are characterized by coarser materials, however, while the former displays the lowest plasticity, the second have medium to high plasticity. the mineralogical data are crucial to explain these behaviors.

Impact of mineralogical composition on the plasticity

Whole rock composition

The mineral composition of the bulk-samples consists mainly of clay minerals, quartz, feldspar, hematite and carbonate (calcite and dolomite). Quartz and feldspar are ubiquitous with a proportion varying between 20 to 55 wt. % and 5 to 35 wt. % respectively. Hematite is present in minor amounts in most of the samples (0-10 wt. %). The total carbonate (calcite + dolomite) ranges between 0 and 15 wt. %. The clay minerals content ranges between 25 and 60 wt. %. During the shaping phase, clay minerals act as plastic and binder components, while other silicates and carbonates as skeletal components.

Clays composition and plasticity

The mineralogical data of whole-rock are not sufficient alone to explain the plasticity behavior and the clay minerals composition is necessary. In fact, this latter is fundamental to the understanding of engineering behavior of fine-grained soils.

The clay minerals of the studied paste in the fraction $< 2 \mu\text{m}$ are diversified. They are made by a mixture of illite (10-40%), kaolinite (5-15%), smectite, mixed layers ($<10\%$), vermiculite and chlorite ($<5\%$), pyrophyllite-talc association (5-10%) is present in the samples Our1, Our2, Our4, Our5 and Tms. Fibrous clay is only present in one paste (Ta), with a content of 18%.

Clay fraction-total clay duality

The fraction $< 2 \mu\text{m}$ could include clay minerals and very fine non-clay minerals. Total clay designates all clay minerals even if their size exceeds $2 \mu\text{m}$. The Figure 5 depicts the clays mineralogy and the clay fraction rate of the whole of studied pastes, with an indication of their lithological nature and of their CF/TC ratio. This figure includes the mineralogical parameter of clays in order to

explain the decorrelation highlighted previously between the grain-size and the plasticity. In fact, the pastes with plasticity indexes ≥ 24 are composed of significant content of smectite, fibrous clay and “smectite + talc-pyrophyllite” association. The content of smectite could not have great impact if the clay fraction is very low ($< 20\%$). More discussion about the difference between the total clay and the clay fraction is to find in Idrissi et al. (2018).

Figure 3: Classification of studied clay raw materials based on the sand-silt-clay ratios (Shepard, 1954).

Figure 4: The plasticity indices of the samples studied as a function of the percentage of the fraction < 2 microns (COR[X,Y]=0.25).

Figure 5: Total clay mineralogy (Obtained by XRD) and clay fraction rate (Obtained by Laser Diffraction Analyzer) of studied pastes with an indication of their lithological nature and their clay fraction / total clay ratio (CF/TC).

Daoudi et al. (2010) show that the burial diagenesis is the main factor controlling the genesis of the clay assemblages of the sedimentary series of the Lower Mesozoic of the Western High Atlas. The effects of burial on these clay assemblages are expressed as the downward increase in the proportions and size of the illite crystallites. In addition, depending on their granulometry, mineralogical and chemical composition, argillites represent the most heterogeneous materials studied. They belong to different sedimentary series which have undergone varying degrees of diagenesis and this has affected their degree of plasticity. The plasticity indices of the argillites are distributed between two extreme poles: clay of low plasticity and clay of high plasticity (Figure 5) and their “clay fraction/total clay (CF/TC)” ratios are between 0.7 and 1, 7. This suggests that the effect of diagenesis seems to have played a major role in the behavior of the plasticity of these materials.

In addition, the observations in scanning electron microscopy show that the Bouchane paste based on shale has illite particles with large sizes ($> 20 \mu\text{m}$; Figure 6). This explains the low clay fraction / total clay ratio in the case of shale-based pulps (Figure 5).

Figure 6: SEM micrographs of schistose fabrics from Bouchane showing the large particles of illite (I) with a grain-size $> 20 \mu\text{m}$.

The difference in the plasticity behavior between pastes based on schist ($\text{PI} \leq 18$) and on colluvium ($25 \leq \text{PI} \leq 27$), despite the similarity expressed by their coarser grain-size distribution, can be explained by their clay composition. Indeed, in most schist samples, the clay composition is dominated by the illite and the kaolinite which are considered as clays of low plasticity. Whereas the colluviums are characterized by a high content of some specific clays: fibrous clays [Ta], pyrophyllite-talc association [Our4], smectite and smectitic mixed layers [Ta, Our4 and Mz3].

The most influential minerals on plasticity

It is well known that the smectites and the fibrous clays display the highest plasticity behavior of clays; this

explains the high plasticity indexes of samples Mz3 ($\text{PI} = 26$) and Ta ($\text{PI} = 25$). For the third paste made by colluvium (Our4) as well as in other pastes made by argillite (Our1 and Our5), and soil (Tms), the clay composition is characterized by low amount of smectite, but coupled with a variable amount of the talc-pyrophyllite association which result from the dismantling of metamorphic rocks of Paleozoic and Triassic (Daoudi and Pot De Vin, 2002). The talc has good ceramic properties. It is an ideal complement to ball clay (clay of high plasticity) to create a dryable body that still has excellent working properties with a good feeling in the hands of the artisan. Talc and Pyrophyllite are close to the smectite in structure and chemical composition, but behave very differently from smectite (Giese et al., 1991). The former are hydrophobic (Steenberg and Harris, 1985) while the latter is hydrophilic. Thus, talc and pyrophyllite are not able to intercalate water and swell due to the relatively weak interaction between the surfaces 001 of these two minerals and water molecules (Giese et al., 1991). Consequently, talc powder has a low plasticity (Norsker, 1990). But these hydrophobic minerals are known to possess a weak van der Waals bond giving them the property of dispersing easily in water and keep them from clumping (Mukherjee, 2013). This author explains that with such dispersing minerals (anti caking), other clays are being dispersed and then easily dissolved in water without clumping. Thus, a greater surface area of the clays will be made available to react with water and to increase the amount of water absorbed by smectite. This explains why the plasticity indexes are the highest in samples with the couple of talc-pyrophyllite and smectite. The highest plasticity indexes are identified in the pastes based on argillites from Ourika site (pastes Our1 and Our5). This high plasticity allows to artisans of Ourika site to produce the most complicated decoration forms at the region of Marrakech (Fig. 2f). These samples are characterized by: a high rate of clay fraction (66 and 41 % respectively), a high total clay content (41 %) and the presence of the pyrophyllite-talc and smectite association.

The plasticity indexes of soils are not very different from those of schist except for the paste of Tamsloht which contains the pyrophyllite-talc association and reaches a PI of 20. The PI of washing muds is still low despite the high rate of clay fraction. This is due to the low content in total clay ($< 28\%$).

6. Conclusion

This study shows as expected, in most of the studied samples, there is a clear relationship between the plasticity behaviour in one hand and the grain size distribution, clay content and clay mineral composition in the other hand. Fine grain-sized materials as well as those containing smectite and fibrous clays display the most plastic

behaviour, whereas the pastes with coarser grain-size associated with the presence of illite display the lowest plasticity. Adding that despite its low plasticity, the talc-pyrophyllite association can display the highest plasticity indexes when coupled with smectite in the same materials. However, in some categories of samples (argillites), the plasticity can be influenced by the effect of diagenesis.

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