Handmade clay bricks:

Chemical, physical and mechanical properties

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Abstract

The clay brick masonry that is much used in historical structures often is in a rather poor state of conservation. In order to intervene correctly in these buildings, it is convenient to characterize the old material. For this purpose, a large sample of clay brick specimens from the 12th to 19th century were collected from six Portuguese monasteries, and were characterized chemically, physically and mechanically. A large variability of the properties was found. Additionally, a sample of handmade new bricks, which are commonly used as replacing material, was also analysed. The results were compared to the old bricks and could be possibly adequate as substitution bricks. Still, significant differences were found in chemical composition, and in water absorption and porosity, which are much lower in modern handmade bricks. With respect to mechanical properties, the range of values found in old bricks was rather high and the degree of deterioration exhibited a large scatter, meaning that a conclusion is hardly possible.

Keywords: clay bricks, chemical properties, physical properties, compressive strength, raw material.

1 Introduction

In the last decades, the importance of historical buildings due to cultural and economical reasons caused a large increase in studies involving ancient building technologies and materials. Clay brick, in its forms of sun dried and burnt, has been around since the beginning of civilization, ten thousand years ago. It was easily produced, lighter than stone, easy to mould and formed a wall that was fire resistant and durable. The usage of brick masonry, often in combination with stone masonry and timber roof, or floors, is well distributed all over the world.

The characterization of old clay bricks is a hard task due to the difficulties in collecting samples, the scatter in the properties and the lack of standard procedures for testing, see Elert *et al.* (2003). Still, characterization is relevant to understand damage, to assess safety, to define conservation measures and even to make a decision on reusing or replacing existing materials, as modern materials can be unsuited, from a chemical, physical or mechanical perspective.

Information about old and handmade clay brick units is rather scarce. Ancient materials are generally different from modern ones, and frequently exhibiting low characteristics, such as high porosity and absorption, low compressive strength and elastic modulus. Most studies target at the ageing process, durability, physical and chemical deterioration process of clay bricks, Baronio and Binda (1984, 1985, 1987). Other authors studied the manufacture process in order to define physical (porosity and water absorption) and mechanical (compressive strength) properties, e.g. Dondi *et al.* (1999) and Elert *et al.* (2003), as well as the effects of additives on the properties of new clay bricks, Bogahawatta and Poole (1996). The effect of pollution and environmental effects were discussed in Cultrone *et al.* (2000).

The mechanical properties of brick are very relevant for the structural behavior of historical constructions, as this is the main influence factor on the compressive strength of masonry. For this reason, a sample of old clay bricks collected from six 12th to 19th century monuments in Portugal was characterized in detail, including chemical, physical and mechanical properties, namely: chemical composition, mass, porosity, water absorption and compressive strength. The same sample has been used to find a correlation between the compressive strength and a micro-drilling technique, see Fernandes and Lourenço (2007).

Besides providing a database of relevant information, the paper addresses two other aspects. The first aspect is a discussion about the possible identification of the source of raw clay. From the most important chemical constituents identified, the provenance of the raw clay source can be estimated and compared with the location of known clay pits. This information allows a qualitative definition of performance and the comparison of behavior with known bricks from the same provenance, see also Nyakairu *et al.* (2002).

A second aspect is that the knowledge of the physical and mechanical properties of old clay bricks and the identification of the most suitable raw material, based on the chemical composition of the clay, would allow to obtain new bricks suitable for replacing old ones. It would be possible to make similar replacements of missing parts, as traditional production techniques can still be found, and a mismatch between materials can lead to early occurrence of cracking (Robinson and Borchelt, 1994). For this reason, a sample of bricks from two manufacturers producing handmade clay bricks in a traditional way was characterized and compared to the old clay brick sample.

2 Description of the test samples

The sample of old clay bricks adopted in this study consists of about 150 bricks, with rather different dimensions and shapes, collected from six different Portuguese national monuments, most of them located in the northern part of the country, as illustrated in Figure 1. The monuments are from four main regions: the Monasteries of Pombeiro and São Martinho de Tibães are from Minho, the Church of Santo Cristo do Outeiro is from Trás-os-Montes e Alto Douro, the Monasteries of Salzedas and São João de Tarouca are from Beira Alta, all from the North of Portugal, and the Christ's Cloister in Tomar is from Ribatejo, located in the Centre of Portugal. The buildings were selected because University of Minho has been working in the monuments or the buildings were undergoing conservation works, which allowed an easy collection of bricks. Nevertheless, the number of bricks was rather limited, as only 10 to 34 units were available from each location. This was mainly due to the scarce number of specimens and of units with adequate size for samples to be extracted from. The bricks were usually collected from buried remainings and vault filling, which explains the difficulty in finding full units. Note that removing bricks from walls or vaults was strictly prohibited. Only the bricks from the Monastery of Salzedas and the Christ's Cloister in Tomar were obtained from building elements or closed doors/windows, explaining their much lower size variability. Table 1 reports a brief description of the sampled old material in terms of acronym, number of specimens gathered, dimensions, period and origin. In the production of most of these bricks, clay was normally extracted from local pits, as the selection of the raw material depended essentially on its availability in the construction location or nearby (Álvarez de Buergo and Limón, 1994). In addition, at that time, the transport network was inexistent or rudimentary for the transport of construction materials. In fact, the transport network in Portugal was

mainly developed during the 19th century and, therefore, most of the materials sampled were made from local raw materials. Afterwards, the bricks were dried and fired in rudimentary ovens until that same period, when the steam engine was introduced in several sectors in Portugal, inclusive, in some industrial processes and in the production of construction materials.

The second type of bricks is constituted by new handmade traditionally fired clay bricks. These bricks were obtained from two manufacturers from the region of Alentejo, located in the South of Portugal, and are often adopted as replacing material of old bricks or used to give a traditional image in new houses. The two manufacturers are from the towns of Galveias (GA) and S. Pedro do Corval (PC), see Figure 1.

Information about this sample, also of about 150 bricks, is reported in Table 2, indicating the name of the manufacturer, acronym, manufacture location, dimensions and number of specimens. These bricks have all a regular parallelepiped shape, although the dimensions differ greatly. In the production of these new bricks, the clay was not submitted to any type of special treatment, and modern quality control of the raw material was not adopted. In the case of the bricks from S. Pedro do Corval, the mixture of the clay and water was mechanically performed by rudimentary equipment, while in the case of Galveias, the clay mixture is processed manually. The bricks are moulded manually with bottomless moulds over a thin layer of sand and fired in traditional wood ovens.

Figure 2 illustrates typical examples of the old and the new handmade clay bricks. The examples show the colors and shape of the different bricks. The bricks from Salzedas (Figure 2a) and Tomar (Figure 2f) and the new handmade bricks exhibit rather regular shapes and dimensions, while the other old bricks present frequently deformations, damages and smaller bricks than normal. It must be noted that bricks

from São Pedro do Corval (Figure 2h) exhibit uniform coloring, while the bricks from Galveias (Figure 2g) have patches of burnt material, indicating a higher firing temperature.

It is noted that the ancient bricks originate from the North of Portugal, whereas the modern brick sample come from the South of Portugal. This is essentially due to the constraints in gathering ancient building materials and in finding traditional brick manufacturers. The latter concentrates in Alentejo, where some demand for traditional handmade bricks still exist.

3 Chemical analysis

Generally, the first step to characterize the raw clay is by means of chemical and mineralogical studies, see Baronio *et al.* (1985), Moropoulou *et al.* (1993), Pauri *et al.* (1994), and Cultrone *et al.* (2004). These are frequent in archaeology, for characterizing old ceramics and pottery, and in the characterization of old mortar properties, e.g. Barrios *et al.* (2000) and Binda *et al.* (2000). The chemical composition is mostly used to identify the raw material characteristics and the source of raw clay, information much relevant for the study of the history of construction and materials. The determination of the mineralogical composition of old bricks allows the identification of possible deficiencies that occurred during their production, like the presence of organic matter, lime nodules, harmful soluble salts and other impurities that might influence the durability of the brick. The presence of soluble salts and other impurities are one of the most important factors of old brick decay and are frequently found, Baronio *et al.* (1985) and Brocken and Nijland (2004). Mineralogical analysis can also provide information about firing temperature and degree of vitrification, Cultrone *et al.* (2000),

which is relevant for manufacturing replacing bricks, Elert *et al.* (2003), López-Arce *et al.* (2003) and Cardiano *et al.* (2004). As this work only deals with the chemical analysis of clay material from old bricks, the information retrieved will be used to identify the area of extraction of the clay used to manufacture the old brick specimens.

3.1 Testing methodology

The chemical composition of the samples was determined by X-Ray fluorescence spectrometry, much used for old ceramics by other authors, Elert *et al*. (2003). The equipment used is a Philips X'Unique II X-Ray fluorescence spectrometer (Figure 3a) connected to a computer used to control the set-up and for data storage. This equipment uses the following detector crystals: LiF220, Pe, Ge and TLAP.

In the present testing programme, small fragments were collected from the clay bricks with the use of a hammer and a rod (Erro! A origem da referência não foi encontrada. Figure 3b) and carefully prepared to obtain uniform conditions of analysis and avoid contamination by elements present in glass coatings and binders. After drying at 110 °C during 24 hours, samples are finely milled to obtain a fine powder.

Afterwards, 10 g of the ceramic material are accommodated in small 30 mm diameter plastic tablets (Figure 3cErro! A origem da referência não foi encontrada.) and put in the spectrometer for analysis.

The sample is then irradiated with X-Rays. The radiation that is generated by the different brick's elements is characterized by a specific wavelength and intensity, which is related with its concentration, allowing, therefore, their identification in the X-Ray spectrum. The chemical oxides and elements that were considered in this study are the most typical and abundant, including: silicon oxide (SiO₂), aluminum oxide (Al₂O₃),

iron oxide (Fe₂O₃), potassium oxide (K₂O), titanium dioxide (TiO₂), sodium oxide (Na₂O), calcium oxide (CaO), magnesium oxide (MgO), barium (Ba), zirconium (Zr), strontium (Sr), rubidium (Rb) and manganese (Mn).

3.2 Results and interpretation of the chemical analysis of old clay bricks

The average chemical composition of old and new handmade bricks is presented in Table 3. All old bricks from the sample were submitted to X-Ray fluorescence spectrometry, which resulted in a total of about 150 tests: OU (10), PO (34), SA (27), TA (28), TI (32) and TO (16). The proportion of the main oxides is expressed in percentage of the weight of material (%), while the contents of the remaining elements: Ba, Zr, Sr, Rb and Mn, is indicated in parts per million (*ppm*) because they are usually found in very small quantities.

The chemical composition of the old bricks' clay consists of 54 to 61 % of silica (SiO₂) and 22 to 32 % of aluminium (Al₂O₃). The average variability of SiO₂ and Al₂O₃ exhibit low coefficients of variation, ranging between 4 and 5 % and 11 and 15 %, respectively. Thus, it seems that the base chemical components of the raw clay used on the bricks is relatively uniform. In fact, the five bricks collected in monuments from the northern part of the country have a fairly similar base, with a variability of 3 and 11 % for SiO₂ and Al₂O₃, respectively. The bricks from Tomar (TO) showed some differences, although not very significant. They exhibit different proportions of SiO₂ and Al₂O₃, which are 6 % higher and 20 % lower than the average of the bricks from the northern part, respectively.

CaO, Na₂O and TiO₂ exhibit a significant dispersion. The presence of the first two elements is often due to contamination by lime mortars or salt, respectively. Na₂O

is rather low in all specimens except in the bricks from Salzedas (SA) and Tarouca (TA), which are located very close and suggests that those clays could have been extracted from close clay pits. The bricks from Tomar (TO) show a significant amount of CaO, most likely due to contamination by lime mortars, found attached on their surface, which could have migrated into the chemical samples.

It is noted that the bricks from Outeiro (OU) contain about two or three times more iron than the other bricks, while those from Salzedas (SA) and Tarouca (TA) are the ones where this element is less found, thus explaining in part their lighter color. See the bricks from Salzedas in Figure 2a.

3.3 Provenance of the raw clay

As discussed above, the chemical composition can provide information needed for an estimation of the provenance of the clay, Castro *et al.* (1997), Capedri and Venturelli (2005), making possible to manufacture suitable replacing bricks. The chemical composition of ceramics is often considered closely related to the chemical composition of the raw material employed in their fabrication. In elder times, commerce of clay type raw materials at long distances was rare, because of the spread availability of clays in nature. Therefore, it is acceptable to consider that the chemical composition of brick fragments is a good indicator of their provenance.

The comparison of the chemical composition of one fragment with the typical chemical composition of a certain production centre allows to determine, by appropriate statistical calculations, the probability for the case where the fragment is from that provenance. Due to the significant amount of data gathered, and its dispersion, the results obtained from old bricks were treated using sophisticated calculations. This

analysis was made using a statistical methodology developed previously, Castro *et al.* (1997) and Castro (1999), and is based on the comparison between the Euclidean distances of the "chemical composition" vectors of different groups. It is mainly composed by two main phases, each one having the following sequential steps:

1. Preliminary calculations:

- a. Elements like sodium and chloride (possible contamination due to culinary uses), phosphor and sulphur (possible contamination due to conditions of preservation or archaeological samples), lead and tin (present in glazes) and lime (present in mortar and plaster) are excluded from the analysis;
- b. The rough concentrations are normalized for a 100 % basis, to eliminate differences on loss of ignition values due to different types of clay materials, firing conditions, moisture contents and organic material contents. This calculation allows to eliminate the eventual effects that different degrees of contamination would have on the results.

2. Cluster analysis:

a. A variable reduction is performed in order to make all elements equally important for the statistical analysis and to avoid that variations in the major constituents, like silica or alumina, are much more strongly considered than variation in minor elements, like manganese, titanium, zirconium, strontium, rubidium, etc. The reduced variable Z_i is calculated through the following formula:

$$Z_i = (C_i - \underline{C_i})/\sigma_i$$

where C_i is the content of the element i in the sample, $\underline{C_i}$ is the average of the contents of that element in all the samples considered and σ_i is the corresponding standard deviation (of the element i).

- b. A clustering method is then employed, which is *the nearest neighbour method* applied to the Euclidean distance between samples and group of samples.
- c. A multivariate test (Hotelling t² test) is performed to compare two clusters taking into account the average value for each element and the standard variation of the values. This allows to estimate the probability that two clusters are dissimilar.
- d. With the formed clusters, a Principal Components analysis was performed to evidence the main components responsible for the variance of the sample and hence the differentiation between clusters.

Therefore, the specimens have been grouped according to their chemical similitude, being the groups characterized by the average values and standard deviation for the various determined chemical elements, always with bricks from the same origin. The purpose was to determine if the chemical composition was uniform among each brick of different origin, which could highlight the presence of bricks that resulted from different raw materials or bricks that experienced contamination, especially coming from lime mortars. It must be noted that the clustering method used in this work takes the closest chemical components, regardless of the value. Therefore, a Euclidean distance of 2.0 or less was used as additional criterion to consider the sample similar to the other one. Experiments performed so far have shown that, for a Euclidean distance of 2.0 or less, corresponds a probability of 90 %, or more, of two samples being similar. For a Euclidean distance much higher than 2.0, the sample is considered dissimilar and, consequently, not grouped. This methodology resulted in the formation of fourteen groups for old bricks and two groups for the new bricks, all presented in Table 4.

From the 146 old bricks analyzed, only 20 could not be grouped. These 20 bricks exhibit a significant distance from the other groups and were discarded from subsequent analysis (Euclidean distance between 5 and 10). The bricks from most locations were divided into two or more groups, exhibiting a rather large variability of chemical properties between groups. The exceptions are the bricks from SA and TO, which could be grouped in one large group, each of them. This means that nearly all bricks from groups SA and TO have similar chemical properties, further corroborating the homogeneity of these two groups. Table 5 exhibits the Euclidean distances between the groups formed with this statistical analysis.

The Principal Components analysis of each group revealed that the component SiO₂ (silica), which is the largest component in clays, contributes very little to the distinction between old samples and cannot be used to distinguish any particular characteristic of bricks. In fact, no single component was found to strongly influence a particular group of bricks, meaning that statistical grouping is a result from all chemical constituents. Actually, silica only explains about 25 % of the variance of the sample, while Al₂O₃ and Fe₂O₃ explain 10-14 % of the variance of the sample. Eventually, no brick or group of bricks stands out due to a particular element.

With the above information, it is possible to make a comparison of the brick groups found with groups present in a database containing the chemical constituents of archaeological and ethnographical ceramics coming from diverse points of Portugal, Castro (1999). It was found that the bricks from PO and TI exhibit a strong similarity (Euclidean distance inferior to 3, see Table 5) and present the typical composition of clays from ethnographical ceramic samples found in the area of Prado, Braga and Guimarães. This corresponds to the construction sites of the referred monuments, namely, Monastery of Tibães in Braga and Monastery of Pombeiro in Felgueiras, which

are within a range of 20 km from Guimarães. The Euclidean distance between OU1 and TA2 also reveals a value below 3 but, due to the distance between their locations, it seems to be statistical coincidence. For the remaining groups, it was not possible to determine any type of correspondence because they present a very high Euclidean distance from the values in the database (Euclidean distance superior to 5). Therefore, the chemical composition of the bricks is distinct from those present in the database, and strongly suggests that the raw clays used in the manufacture of bricks were obtained locally. Moreover, the groups of bricks from distant regions are usually closer to each other than to the chemical compositions available in the archaeological database. This indicates that archaeological ceramics cannot be compared with old building bricks, as they seem to be manufactured with rather distinct materials.

3.4 Comparison with the new handmade clay bricks

With respect to the new bricks, only four specimens were tested for each brick origin, as the variability exhibited by the tested specimens was lower than 20 %, see Table 3. In Table 4, oxides SiO₂ and Al₂O₃ exhibit low coefficients of variation, ranging between 3 and 16 %. The chemical composition of the new bricks was found to be in the range of the values found for old bricks, with a lower amount of Al₂O₃. The bricks from PC exhibit rather large values for CaO. The most reasonable explanation is that those bricks were manufactured using unclean clays that had a high amount of lime in their raw composition. In addition, these bricks contain very high amounts of MgO (magnesium oxide), Sr (strontium) and Mn (manganese), which indicate that those bricks were manufactured with rather heterogeneous clays.

The statistical treatment of the new handmade brick's data resulted in the formation of two distinct groups, with the bricks from each location showing a relatively large distance between their chemical characteristics, despite the relative proximity of manufacture sites. These bricks were then compared to the old clay bricks and with the database or archaeological and ethnographical ceramics. The results, presented in Table 5, showed that the new handmade clay bricks have, in general, a different constitution than old clay bricks and are fairly different from archaeological ceramics, exhibiting an average Euclidean distance of 11.

4 Assessment of the physical and mechanical properties

The porosity and water absorption of bricks were calculated according to LUMA4, Rilem (1991). The mechanical property retrieved from the test specimens was solely the peak compressive strength f_c , obtained according to ASTM C67-07a (ASTM, 2007).

4.1 Absorption testing procedure

The determination of the bulk mass, water absorption and porosity of both old and new clay bricks was carried out on full size specimens. Working with material with very heterogeneous properties makes grinding full bricks into smaller specimens inadequate for the assessment of these parameters. In fact, if the bricks were ground, the relative porosity would be increased by opening closed pores and voids, which is undesirable. Due to the large number of specimens, the tests have been performed along different periods of the year. The air conditions (temperature and relative humidity)

were monitored and resulted in the following values: 18-22° C for air temperature and 40-60 % for relative humidity. It should be noted that the variation of these parameters is only moderate, meaning that their influence in the results is limited.

The procedure described is based on the results from water absorption by immersion. Firstly, the specimens were dried in an oven at a temperature of $100 \pm 5^{\circ}$ C until constant mass was reached, i.e. when the difference between the masses of the dried bricks weighed every 2 hours, after a first 24 hour drying period, was lower than 0.1 %. Then, the bricks were cooled down at room temperature and their dry mass was determined. Afterwards, they were immersed in water and saturation was carried during a period of 48 hours, which was generally necessary to achieve constant mass, although the standard only indicates a 24 hour period. The temperature of the water was continuously monitored and maintained a temperature around 14.5 and 15° C during the testing period. Then, the specimens were weighed and their saturated mass was calculated. Finally, the hydrostatic mass of the specimens that corresponds to the weight of the water moved by the immersion of the saturated brick in a known volume of water was calculated.

4.2 Compression testing procedure

For the compression test, the specimens were cut from the bricks in the moulding direction, perpendicular to bed joints (see Figure 4). Generally, very small specimens were obtained, with typical cross sections of $30 \times 30 \text{ mm}^2$, and 30 to 40 mm of height. All specimens were machine ground, so that the loading faces were properly aligned. Due to the heterogeneity of the samples and the small height of the bricks, it was not possible to obtain specimens with a height/width ratio adequate to neglect the

boundary effects, i.e. such that the height/width ratio should be larger than two, Neville (1963). The presence of inclusions/voids also affects the results. Here, the smallest dimension accepted for testing after surface rectification was 20 mm, as suggested in Binda *et al.* (1996). This criterion resulted in the rejection of some specimens because the minimum dimension was not met, or the distortion was so large that it was impossible to obtain a regular specimen for testing.

In order to minimize the effect of the friction effects imposed on the specimen by the machine platens and in order to secure a uniform stress distribution at least in the centre of the specimen, a pair of friction-reducing Teflon sheets with a thin layer of mineral oil in between was placed between the surfaces of the platens and the specimen.

The test set-up for the compression tests is composed by a steel frame equipped with a compression load cell with a maximum capacity of 25 kN and connected to an appropriate control system, see Figure 5. The steel platens were rectified in order to provide a flat surface. The lower platen has a spherical seat made of tempered steel that allows the initial alignment and accommodation of the specimen, thus, facilitating the alignment of the applied load with the centre of the specimen as well as preventing any other unfavorable effect due to geometrical imperfection of the specimen. Finally, a steel cube with $100\times100\times100~\text{mm}^3$ provided additional height for the specimens. Because of the rather small dimensions of the brick specimens, besides the control displacement transducer, no displacement measuring device was mounted in the system. The variable selected as feedback signal was the vertical displacement of the actuator, provided by an external Linear Variable Displacement Transducer (*LVDT*) with a linear field of $\pm 2.5~\text{mm}$, mounted in the actuator body. The test procedure started in load control until the specimen was loaded with a force of 0.25~kN. Then, the procedure was carried out entirely under displacement control by means of the vertical *LVDT* at a

displacement rate of 3 μ m.s⁻¹. The compressive strength f_c of the test specimens was calculated by dividing the maximum compressive load on the specimen by the initial cross-sectional area.

4.3 Results of physical tests

The results of the bulk mass, porosity and water absorption by immersion in cold water for the entire sample of old clay bricks are presented in Table 7, as well as the coefficient of variation (*COV*) obtained for the total number of specimens of the group of bricks from a given origin. The average values for the three parameters considering the entire sample are: 1750 kg/m³ for the bulk mass with a *COV* around 3 %, 29 % for the porosity with a *COV* around 18 % and 17 % for the water absorption by immersion with a *COV* of 24 %. These values show that the average properties of the Portuguese old clay bricks from the 12th-19th centuries present a moderate variability, and thus, the average values can be adopted as reference for current engineering applications involving the diagnosis and retrofit of historical structures.

Table 7 presents also the results for the new handmade bricks with the number of specimens considered for testing between brackets. Although the bricks were manufactured in different locations, although rather close geographically, and present visually important differences related to their color and texture, the dispersion of the results is globally very low, with a *COV* smaller than 1 % in the case of bulk mass and an average of 3-3.5 % in the case of porosity and water absorption. The values obtained for these new bricks are very close, showing only a marginal difference of 0.9-1.7 % between average results.

The difference of bulk mass between new and old clay bricks is negligible, as shown in Table 7. However, the new bricks have a much lower porosity and absorption rate, when compared with old bricks, which is an important improvement in terms of durability. The porosity of the new bricks is 29 % lower than old bricks, while for water absorption, the difference reaches 37 %.

Because the porosity plays a very important role in the durability of bricks, an additional characterization of the porosity was carried out. Figure 6 illustrates the distribution of porosity among old and new bricks. It can be observed that the most frequent range of values for old bricks is about 20-35 %, which is rather large and indicates a considerable scatter. In contrast, modern bricks exhibited a much lower range of values (15-25 %). In addition, 90 % of the new brick specimens exhibit 19-22 % of porosity, which indicates a homogeneous sample and an improved manufacturing process, although traditional.

4.4 Results of mechanical tests

The average compressive strength f_c for the complete sample as well as the dispersion of the data is illustrated in Table 7. The number of specimens considered for testing in each group was: OU (8), PO (28), SA (27), TA (18), TI (23) and TO (16). Generally, a large variability on the compressive strength was obtained, with coefficients of variation up to 50 %. It is possible to observe that the bricks with lower f_c exhibit also a higher dispersion. The wide range of strengths found, is between 6.7 and 21.8 N/mm², with an average of 11.6 N/mm² considering the total sample and 8.3 N/mm² considering the four weakest bricks (origin). In particular, the strength of bricks from Tomar (TO) and Salzedas (SA) differ significantly from the other four groups of

bricks. Their high compressive strength could be related to the technological process available at the time of manufacture, as the bricks from Tomar (TO) and Salzedas (SA) are more recent than the other bricks.

The different locations in the building from where the bricks were sampled must also be considered. Clay bricks from Outeiro (OU), Pombeiro (PO), Tarouca (TA) and Tibães (TI) were mainly collected from buried remainings, soil deposits and infill material, while the clay bricks from Tomar (TO) and Salzedas (SA) were obtained from building elements. Therefore, different environmental actions and deterioration could also have contributed to the results obtained.

With respect to the new handmade bricks, a total of fifteen specimens were considered for testing in both cases. Their average compressive strength is presented in Table 7, where it is clear that bricks from Galveias (GA) are stronger than bricks from S. Pedro de Corval (PC). The lighter brown color of the bricks from S. Pedro de Corval might indicate a lower firing temperature relatively to those from Galveias. The group GA has a total average dispersion of 11 %, with results ranging from 8.1 to 14.0 N/mm², which is lower than the 17 % obtained for the group PC, whose values range between 4.2 and 8.8 N/mm².

Table 7 shows, additionally, that the new bricks have comparable compressive strength to the old bricks. In fact, if the high values of compressive strength from SA and TO are ignored, the results for new bricks are within the range of compressive strength values found for old bricks. This result strongly emphasizes the role of the firing process for the final compressive strength and that particular care is needed when replacing bricks. Therefore, the new handmade clay bricks are not compatible, from a mechanical perspective, with the strong SA and TO bricks.

5 Discussion on the compatibility of old and new bricks

In order to choose the most appropriate material for a particular intervention, it is necessary to have a certain number of parameters that will guide towards the materials to choose. These parameters should be straightforward to obtain. That is why the physical and mechanical properties of the final brick and chemical properties of the raw clay discussed in this paper were chosen.

The chemical characteristics of the raw clay used in new and old bricks are fairly different. Additional analysis showed that it influences little the final properties of the bricks, as it was not found a particular element that influences a particular characteristic of the final fired brick. In fact, the chemical information was mostly used and is most useful in determining the source of the raw clay, which is then compared to known clay pits. Obtaining similar raw materials is one parameter that can help manufacturing suitable bricks as long as the manufacturing process (traditional method) allows to obtain bricks with the other necessary properties.

Relatively to the mechanical and physical properties, the new handmade bricks were found fairly suitable for the replacement of most of the old bricks, as the mean values of the compressive strength are quite close to each other and the values for porosity and water absorption are much better. The exceptions are the bricks from SA and TO, which exhibit a much larger compressive strength. In this case, it should be necessary to study another manufacturing process that could make stronger bricks, in a traditional way.

6 Conclusions

Typical chemical, physical and mechanical properties of handmade bricks from different places and periods have been characterised. The mechanical and physical characterization of old clay bricks from Portuguese monuments from the 12th to 19th century presents a large variability, typically with high porosity (29%), high water absorption (17%) and low strength (11.5 N/mm²). The results of the statistical analysis based in the comparison of the chemical constituents of the clay bricks sample and of other archaeological ceramics showed that clay bricks for construction are much more different than typical archaeological ceramic ware. The analysis of the chemical constituents indicates that the raw clay for these materials was most probably obtained in local clay pits associated with the building location. This conclusion is compatible with the rather large difference in chemical composition of all clay bricks found by the authors. Therefore, it seems hardly possible to make chemically compatible replacements of missing parts in conservation works.

The creation of a database for clay bricks would be of interest for the purpose of identifying the provenance and for the selection of clays. This is a long lasting and costly project, taking into consideration that the database of Portuguese archaeological ceramics described here was initiated more than 10 years ago and encompasses more than 2000 samples.

Regarding new handmade bricks, the results showed that they have mechanical properties of the same order of magnitude, but much lower for porosity and water absorption. This indicates that these new bricks are more durable than the old ones tested, and have similar compressive strength. In terms of chemical characteristics, the raw clay used is different from the old bricks adopted in this study, due to the different

geographical location, and also different from the archaeological ceramic ware present in the comparison database. Additionally, the chemical characteristics of these bricks were rather different too, despite the fact that the raw clay sites were relatively close geographically.

It seems that the new handmade bricks can be suitable for the substitution of most of the old bricks tested in this work. The chemical constituents have a fairly similar base, although rather different due to the fact of being in clearly different type of soil, and they exhibit a very similar compressive strength. The new handmade bricks are also more durable, as the potential of suffering from deterioration caused by water infiltration decreased significantly.

However, the properties of a particular brick cannot be entirely determined solely through the characteristics studied in this article, as additional information might be necessary to further understand the manufacturing process, such as the firing temperature. The latter requires other types of analyses not carried out within the framework of this study. Together with the chemical analysis, which can allow to find the most adequate raw clay to make adequate replacement bricks, one should be able to further increased the appropriateness of replacing bricks.

The compatibility within chemical, physical and mechanical properties can be possibly established by several aspects, namely geometrical aspects, manufacturing aspects and building technology, vulnerability towards degradation (freeze-thaw resistance or pore size distribution) and compressive strength (providing possibly an lower and upper boundary).

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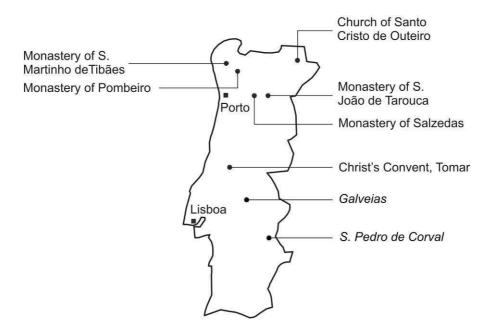


Figure 1 – Portuguese map showing the location of the brick sample (new handmade bricks locations are in italic).



Figure 2 – Typical examples of the collected samples for – old bricks: (a) Salzedas (SA); (b) Outeiro (OU); (c) Pombeiro (PO); (d) Tarouca (TA); (e) Tibães (TI); (f) Tomar (TO) – and new handmade bricks; (g) Galveias (GA); (h) S. Pedro do Corval (PC).



Figure 3 – Analysis equipment, X-Ray spectrometer, and preparation of the ceramic sample for spectrometry measurements. (b) Aspect of the sample extracted from bricks and (c) plastic tablet with the ceramic material ready for testing

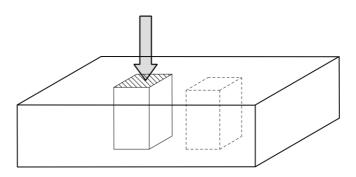


Figure 4 – Schematics showing how the specimens were cut from the bricks and the direction of compression tests.

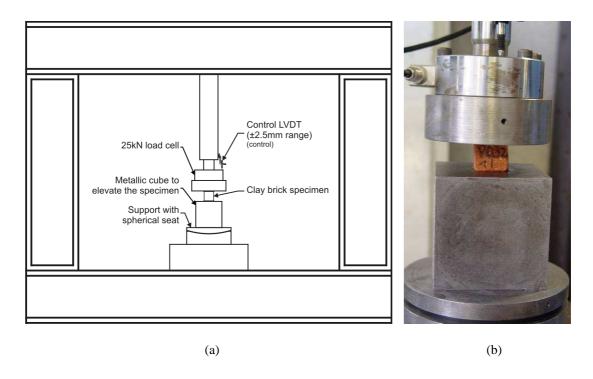


Figure 5 – Steel frame equipped with a 25~kN load cell for compression tests on brick specimens:

(a) general view; (b) detail of specimen and load cell.

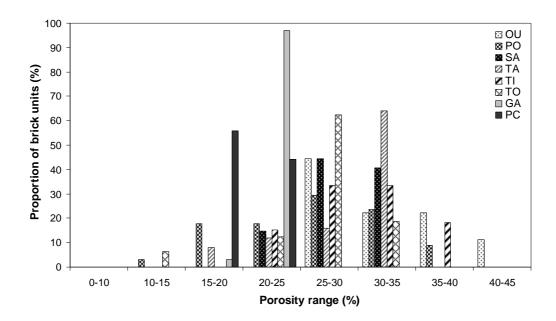


Figure 6 – Distribution of porosity for the complete sample.

Table 1 – Description of the origin and number of specimens of the sampled old material. The coefficients of variation (COV) for the dimensions are given between square brackets.

Monument	Construction period (century)	Location	Dimensions (cm [COV]×cm[COV]×cm[COV])	Acronym (no. specimens)
Church of Santo Cristo do Outeiro	17 th	Outeiro (Bragança)	14.6[14%]×10.5[23%]×4.0[11%]	OU (10)
Monastery of Pombeiro	12 th to 16 th	Pombeiro (Felgueiras)	15.7[24%]×11.6[13%]×3.6[22%]	PO (34)
Monastery of Salzedas	12 th to 18 th	Salzedas (Tarouca)	25.8[7%]×13.0[9%]×3.2[12%]	SA (27)
Monastery of São João de Tarouca	17 fo 17		13.9[26%]×10.4[23%]×3.7[18%]	TA (28)
Monastery of São Martinho de Tibães			20.4[23%]×15.0[22%]×3.8[16%]	TI (32)
Christ's Cloister	18 th to 19 th	São João Baptista (Tomar)	19.1[31%]×12.9[4%]×3.5[13%]	TO (16)

Table 2 – Description of the origin, number of specimens and dimensions of the sampled new material.

The coefficients of variation (COV) are based on the ration between the volume from the declared dimensions and the real volume calculated from the immersion in cold water procedure described within this paper.

Manufacturer	Location	Dir (cm×cm×cm)	Acronym (specimens)		
Fernando Casaca	Galveias	20×10×6 25×14×6	33 32	5.4 %	GA
	Sur / Clus	30×15×4	30		(95)
	São	30×15×6	15		DC
Luís Dias	Pedro do Corval	30×15×4	30	13.3 %	PC (61)
		25×10×5	16		(01)

Table 3 – Average chemical composition of old and new clay bricks with the number of specimens between brackets and the coefficients of variation between square brackets.

	SiO ₂	Al_2O_3	Fe ₂ O ₃	K ₂ O	Na ₂ O	TiO ₂	CaO	MgO	Ba	Zr	Sr	Rb	Mn
				(%)			l			I .	(ppm)	1	
	Old bricks												
OU (10)	56.2	25.3	11.4	3.5	0.5	1.0	0.3	1.5	698	285	75	195	758
	[9%]	[5%]	[41%]	[14%]	[40%]	[10%]	[47%]	[29%]	[23%]	[18%]	[25%]	[12%]	[28%]
PO (34)	57.5	25.1	8.4	4.9	0.5	1.3	0.4	1.6	986	392	147	302	496
	[5%]	[10%]	[18%]	[12%]	[33%]	[10%]	[106%]	[19%]	[34%]	[28%]	[50%]	[15%]	[35%]
SA (27)	54.4	32.2	4.1	5.1	2.0	0.3	0.8	0.9	352	102	99	319	313
	[5%]	[8%]	[61%]	[17%]	[35%]	[87%]	[66%]	[41%]	[50%]	[75%]	[66%]	[18%]	[37%]
TA (28)	55.6	30.9	4.1	5.0	1.9	0.4	0.9	1.0	440	147	121	317	335
	[7%]	[13%]	[22%]	[15%]	[41%]	[26%]	[84%]	[29%]	[57%]	[59%]	[86%]	[27%]	[29%]
TI (32)	53.8	29.4	8.1	4.4	0.5	1.2	0.9	1.4	856	406	116	282	474
	[6%]	[9%]	[18%]	[11%]	[31%]	[8%]	[60%]	[20%]	[20%]	[28%]	[58%]	[19%]	[20%]
TO (16)	60.8	21.6	7.0	3.6	0.4	0.8	3.6	2.2	410	113	40	139	328
	[4%]	[10%]	[9%]	[22%]	[23%]	[12%]	[81%]	[15%]	[14%]	[35%]	[27%]	[11%]	[27%]
Range	54-61	22-32	4.1-11.4	3.5-5.1	0.4-2.0	0.3-1.3	0.3-3.6	0.9-2.2	353-986	101-406	40-147	139-314	318- 758
Average	56.4	27.4	7.2	4.4	1.0	0.8	1.2	1.4	624	241	100	259	451
						New bri	cks						
GA (4)	67.5	18.8	5.5	4.2	0.7	1.3	0.6	1.3	532	307	81	164	893
	[3%]	[4%]	[14%]	[3%]	[14%]	[13%]	[13%]	[13%]	[8%]	[10%]	[13%]	[6%]	[22%]
PC (4)	53.8	19.6	10.7	3.6	1.7	1.2	4.6	4.5	506	234	275	119	1368
	[2%]	[3%]	[11%]	[6%]	[16%]	[9%]	[11%]	[4%]	[13%]	[17%]	[13%]	[14%]	[11%]
Range	52.7-69.1	18.3-20.2	4.7-11.8	3.4-4.3	0.7-2.0	1.1-1.5	0.5-5.3	1.1-4.6	443-589	193-349	73-307	104-173	723- 1544
Average	60.6	19.2	8.1	3.9	1.2	1.3	2.6	2.9	519.3	270.8	178.2	141.5	1130.6

Table 4 – Groups formed by chemical similarity between specimens of the same origin, with the number of specimens between brackets, the average of each chemical component and correspondent standard deviation between square brackets.

Groups	SiO_2	Al_2O_3	Fe ₂ O ₃	K ₂ O	Na ₂ O	TiO ₂	CaO	MgO	Ba	Zr	Sr	Rb	Mn
(samples)	(%)								(ppm)				
OU1 (4)	52.8	25.6	14.3	3.6	0.5	1.0	0.4	1.7	637	240	71	188	676
	[1.2]	[0.5]	[0.7]	[0.1]	[0.1]	[0.0]	[0.1]	[0.3]	[87]	[18]	[9]	[2]	[7]
OU2 (3)	59.2	24.5	9.1	3.6	0.6	1.0	0.4	1.5	838	312	82	212	928
	[1.3]	[1.7]	[0.2]	[0.3]	[0.1]	[0.1]	[0.2]	[0.4]	[40]	[13]	[28]	[16]	[139]
PO1 (5)	56.8	26.7	8.5	4.4	0.4	1.1	0.2	1.8	702	237	64	238	468
	[2.0]	[1.0]	[1.0]	[0.1]	[0.1]	[0.1]	[0.1]	[0.2]	[54]	[50]	[24]	[22]	[50]
PO2 (6)	53.5	27.8	9.9	4.4	0.4	1.3	0.7	1.9	906	305	162	255	432
	[1.0]	[1.0]	[0.7]	[0.3]	[0.1]	[0.1]	[0.4]	[0.2]	[141]	[96]	[32]	[17]	[153]
PO3 (18)	58.0	24.1	9.3	4.9	0.6	1.2	0.3	1.5	955	413	173	303	447
	[1.5]	[1.5]	[0.8]	[0.5]	[0.2]	[0.1]	[0.2]	[0.2]	[78]	[73]	[18]	[22]	[49]
SA (24)	54.9	32.4	3.8	5.1	2.1	0.2	0.7	0.8	325	80	105	312	269
	[1.6]	[2.2]	[1.0]	[0.6]	[0.6]	[0.1]	[0.3]	[0.1]	[66]	[40]	[37]	[34]	[50]
TA1 (4)	57.6	23.3	11.5	3.24	1.0	0.9	1.0	1.3	450	170	128	150	669
	[1.4]	[1.1]	[1.1]	[0.2]	[0.1]	[0.1]	[0.1]	[0.1]	[111]	[68]	[21]	[15]	[157]
TA2 (10)	52.0	26.9	13.5	3.8	0.6	1.0	0.4	1.7	546	113	59	169	578
	[1.6]	[1.2]	[1.5]	[0.2]	[0.1]	[0.1]	[0.2]	[0.2]	[44]	[64]	[11]	[20]	[75]
TA3 (4)	50.4	31.6	12.0	2.8	0.5	0.9	0.2	1.4	435	162	42	202	522
	[1.0]	[1.2]	[1.4]	[0.1]	[0.1]	[0.1]	[0.0]	[0.2]	[61]	[89]	[22]	[16]	[65]
TA4 (5)	52.5	32.14	9.1	3.6	1.1	0.6	0.1	0.8	347	87	51	256	304
	[2.0]	[1.6]	[1.1]	[0.2]	[0.2]	[0.0]	[0.0]	[0.1]	[103]	[43]	[21]	[22]	[39]
TI1 (20)	52.4	30.5	8.6	4.2	0.5	1.2	1.0	1.4	770	351	109	261	468
	[2.1]	[1.8]	[1.3]	[0.3]	[0.1]	[0.1]	[0.5]	[0.2]	[134]	[62]	[29]	[47]	[97]
TI2 (4)	52.2	28.9	8.7	4.8	0.7	1.3	1.2	1.8	1058	444	225	312	407
	[0.7]	[1.1]	[1.5]	[0.2]	[0.2]	[0.1]	[0.3]	[0.2]	[75]	[94]	[17]	[32]	[61]
TI3 (3)	56.7	29.2	7.2	3.7	0.3	1.2	0.3	1.2	814	603	144	277	455
	[0.7]	[1.8]	[0.8]	[0.2]	[0.0]	[0.0]	[0.1]	[0.2]	[103]	[161]	[19]	[28]	[112]
TO (16)	60.8	21.6	7.0	3.6	0.4	0.8	3.6	2.2	410	113	40	139	328
	[2.3]	[2.1]	[0.6]	[0.8]	[0.1]	[0.1]	[2.9]	[0.3]	[58]	[40]	[11]	[15]	[87]
GA (4)	67.5	18.8	5.5	4.2	0.8	1.3	0.6	1.3	532	307	81	164	893
	[2.0]	[0.8]	[0.8]	[0.1]	[0.1]	[0.2]	[0.1]	[0.2]	[40]	[32]	[11]	[10]	[197]
PC (4)	53.8	19.6	10.7	3.6	1.7	1.3	4.6	4.5	506	234	275	119	1368
	[1.3]	[0.5]	[1.2]	[0.2]	[0.3]	[0.1]	[0.5]	[0.2]	[66]	[40]	[35]	[17]	[154]

Table 5 – Euclidean distances between groups.

	OU1	OU2	PO1	PO2	PO3	SA	TA1	TA2	TA3	TA4	TI1	TI2	TI3	TO	GA	PC
OU1																
OU2	7.51															
PO1	5.86	4.94														
PO2	6.44	4.58	3.11													
PO3	8.52	4.77	4.29	3.15												
SA	11.19	10.07	7.85	8.9	8.73											
TA1	5.21	4.94	7.23	6.52	6.61	8.08										
TA2	2.61	6.17	4.48	5.08	7.47	7.78	4.51									
TA3	5.96	7.34	9.92	6.74	8.32	7.48	7.35	7.47								
TA4	10.67	8.01	7.84	9.15	8.64	4.57	8.71	6.61	5.77							
TI1	5.08	3.91	2.96	2.23	3.35	7.33	5.09	4.17	5.43	7.24						
TI2	10.96	7.27	6.47	2.5	4.09	9.3	8.33	9.13	10.26	10.44	3.53					
TI3	8.69	4.53	4.46	4.27	3.25	9.97	7.11	6.36	7.66	13.3	2.78	5.94				
TO	8.03	7.54	5.23	7.45	8.77	6.8	4.89	5.07	6.01	6.33	5.92	10.65	7.77			
GA	10	4	7.6	9.23	8.2	9.25	6.68	7.85	12.4	10.86	6.79	11.71	8.04	5.53		
PC	12.93	10.35	13.86	11.8	12.56	16.02	12.49	12.64	15.41	19.66	10.87	12.23	13.47	9.06	13.33	

Table 6 – Air conditions of the room during water absorption experiments.

Temperature	Relative humidity	Water temperature				
18-22 °C	40-60 %	14.5-15 °C				

Table 7 – Average porosity, bulk mass, water absorption by immersion and compressive strength for the old bricks and the modern bricks using the traditional manufacturing process, with the number of specimens considered during the experiments between brackets and the coefficients of variation between square brackets.

Specimens	Porosity	Bulk mass	Water absorption	Specimens	Compressive strength f_c						
	(%)	(kg/m^3)	(%)		(N/mm^2)						
Old bricks											
OU (9)	33.0 [13.9%]	1742 [1.7%]	21.3 [21.6%]	OU (8)	8.5 [28%]						
PO (34)	26.3 [25.5%]	1754 [2.2%]	15.8 [33.4%]	PO (28)	9.2 [54%]						
SA (27)	28.2 [10.6%]	1800 [1.9%]	17.6 [13.4%]	SA (27)	14.5 [32%]						
TA (25)	29.2 [14.5%]	1747 [1.8%]	17.8 [17.7%]	TA (18)	8.7 [41%]						
TI (33)	30.4 [14.7%]	1739 [1.5%]	18.8 [19.8%]	TI (23)	6.7 [55%]						
TO (16)	27.5 [14.2%]	1656 [3.0%]	15.0 [15.5%]	TO (16)	21.8 [31%]						
Average	29.1	1740	17.7	Average	11.6						
	New bricks										
GA (4)	20.9 [3.2%]	1735 [0.8%]	11.2 [4.2%]	GA (15)	9.2 [11%]						
PC (4)	20.5 [2.8%]	1750 [0.5%]	11.1 [3.3%]	PC (15)	5.8 [17%]						
Average	20.7	1743	11.2	Average	7.5						