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Non-destructive portable X-ray Fluorescence (pXRF) method for the characterization of Islamic architectural ceramic: Example of Saadian tombs and El Badi palace ceramics (Marrakech, Morocco)

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ABSTRACT

Archeological decorated ceramics from the Saadian tombs and El Badi palace sites (Marrakech, Morocco) have reached an advanced deterioration phase; the glazes have been increasingly weakened due to human and environmental impacts over time. Portable X-ray Fluorescence (pXRF) was performed in situ and on samples selected from these two monuments, in order to define the chemical agents responsible for the color of the studied glazed ceramics and to determine their evolution over time to help find answers and link between degradations and chemical compositions of different type of glazes. The results show that all samples are lead-silica type glazes with 25–59 wt% of PbO and 51 wt% of SiO_2. The coloring agents used for the original glaze are conventional, copper (Cu^{2+}) for the green color, iron (Fe^{3+}) and manganese (Mn^{2+}) for the yellow and black glaze. Phosphorus (P_2O_5), comes from carbonate mineral phases, is responsible for the blue opalescence of glazes. The study reveals that the ceramic industry has evolved recently in Morocco; elements such as calcium and potassium are currently used in small quantities while lead is increasingly used as flux in the glaze mixture. Iron and copper are still used for black, yellow and green colors, while the use of phosphorus has been replaced by other elements such as cobalt and copper.

1. Introduction

The history of mankind has been accompanied by the use of ceramics for buildings, monuments and art objects. In the course of time, all ceramics are affected by weathering. The interaction between ceramic clayey materials and natural or anthropogenic weathering factors controls the type and extent of ceramic damages. Utilization of the monuments, insufficient maintenance or inappropriate restoration activities may have contributed to alarming ceramic damage. Due to the increasing awareness and respect for our built heritage, preservation of ceramic monuments has become an important public and political concern (Fitzner et al., 2002; Avrami et al., 2008).

In Morocco, historical Islamic buildings of Marrakech impress by their filigree architecture and colorful facades and many of them therefore are inscribed to the UNESCO world heritage list as having cultural and historical significance. Among the most fascinating historical sites of Marrakech, Saadian Tombs and El Badi Palace date to the Saadian dynasty (1549–1959) (Paccard, 1981). These two monuments are characterized by their carefully arranged and gorgeous decorative glazed ceramics, also called Zellige, which give the buildings their typical and imposing appearance. The art of Zellige is present in Morocco and in western Mediterranean regions since the early dynasties; it appears under the control of the Almohads (1130–1269) and the Merinides (1244–1465) in Fes (Morocco), the Zayanids (1236–1554) in Tlemcen (Algeria), the Nasrids (1232–1492) in Granada (Spain) and the Hafsides (1229–1574) in Tunis (Tunisia) (Erzini, 1993; Hattstein and Delius, 2000). However the shapes and colors used have evolved over time, the manufacturing technology varies from one period to another with the introduction of new chemical elements in the glaze and the ceramic shards to improve their physicochemical properties and ensure their stability against different degradation factors.

The inspection of historical monuments in the region of Marrakech ensure the conservation and restoration of Saadian Zellige, several catering companies were made to allow visitors to admire the monuments without changing the original architecture of the building. Despite these efforts, the original decorated ceramics are reached advanced degradation stat, which requires a conservation-restoration strategies based on scientific and technical studies.

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The Saadian tombs as well as the El Badi palace contain three categories of Zellige spaced out over three periods; the original Zellige dating from the 16th century, the first restoration period occurred in the first half of 20th century, and the recent restoration started in 2015 and made by Zellige from Fez (first site of manufacturing Zellige in Morocco). The need for having valid and useful heritage documentation is therefore evident and should constitute a priority action in any type of restoration (Almagro, 2013). Unfortunately, in these studied monuments, the availability of proper documentation is very limited (Gradmann et al., 2015; El Halim et al., 2018), as is the case in most historical monuments of emerging or developing countries in the South and East Mediterranean areas. To this end, the aim of this study is to characterize the chemical composition of the different glazes ceramics and to emphasize their evolution over time. The study allows to define the chemical agents responsible for the color of the studied ceramics and the reformulation of certain hypotheses on the manufacturing technology of these materials. This study will also help to distinguish between the different types of materials depending on their chemical compositions.

2. Materials and methods

The first site (Saadian tombs) contains the tombs of the Saadian kings and their caliphs, and consists of a garden cemetery, on either side of it there are two mausoleums known as “The Kouba” and “ The mausoleum principal”. Walls and tombs were decorated by the mosaics of the original and the restored Zellige. The second site (El Badi palace) was built by the monarch Ahmed Al-Mansour at the end of 16th century using very precious materials, the general layout of the palace and its decoration shows the beauty and perfection of Moroccan architecture at that time, the building contains four large pavilions, six basins, a large garden and guest houses.

For these architectural buildings, there is a classical arrangement of materials typically founded in Islamic historical monuments. Terracotta tiles and marble for floors, mosaic of Zellige for walls for an overall high of one to two meters, a frieze of chilled Zellige between these mosaics and stucco carved in lace, and then ceilings are generally covered with polychrome cedar wood and green tiles (Fig. 1).

In these monuments, Zellige patterns generally consist of an eight-pointed star called Khatem and sixteen-pointed star called Sett-Acher, surrounded by more than twenty different patterns with various color and shape (Benamara et al., 2003). The colors used to adorn the building are green, blue, yellow, white and black, the same colors used for the current decorative ceramics in the most palaces and buildings of Marrakech. A total of 122 samples of Zellige glazes were analyzed; these samples includes 38 samples of the original ceramic, 53 samples of the ceramic used in the first restoration (the most widely distributed in the site) and 31 samples of the current decorative ceramic used in the second restoration phase. An average of three to five measurements was made for each sample using a Bruker AXS portative analyzer instrument. Light elements have been detected using 50 kV with 20 μA for optimum excitation and without a filter in the X-ray beam path leading to the sample. The lighter elements Mg and Al have higher relative errors than the heavier ones but with a vacuum and He-flush system, which reduces the absorbing air between sample surface and detector, the precision of the measurements is improved and even Na can be detected. The abundant elements in Islamic glazes such as Mg, Al, Si, P, Pb, K, Ca, Ti, Fe, Mn, Cu and Zn are all well detected except Co excluded on this list because of the interference with Fe. Iron and cobalt have very closed peaks so if one of them is present in significant concentrations an interference results (Bonvin, 2000).

The instrument provides the Universal mode, bringing together the benefits of Empirical and FP calibrations, automatic Method detection, and Dual Energy excitation. In the first second of a measurement, the analyzer determines the best excitation voltage, anode current, and beam filter based primarily on the matrix element (Ti, Al or Cu), and automatically switch to it. In the next five seconds, the algorithm determines if the measurement would benefit from a second analysis at a lower energy (Dual Mode). Typically, at the end of a 10–15 s assay, the appropriate calibration will be chosen (Empirical or FP) and results will display.

Another non destructive pXRF analyze was made on 23 samples of the Saadien tombs building using a pXRF Thermo Fisher Scientific Niton XL3t GOLDD instrument, the results of this study was used to have reference data for evaluation and comparison. The obtained results were also compared with those obtained by Gradmann (2016) on several Saadian building using a pXRF Thermo Fisher Scientific XL3 Hybrid device non destructive instrument (Table 1).

Portable X-rays Fluorescence (pXRF) represents one of the most effective tools for in situ, non-destructive elemental analysis (Schackley, 2011; Frahm & Doonam, 2013; Shugar & Mass, 2014; Ceccarelli, 2016).

The statistical analysis and the visualization of results are served using the functions provided by the two packages FactoMineR and

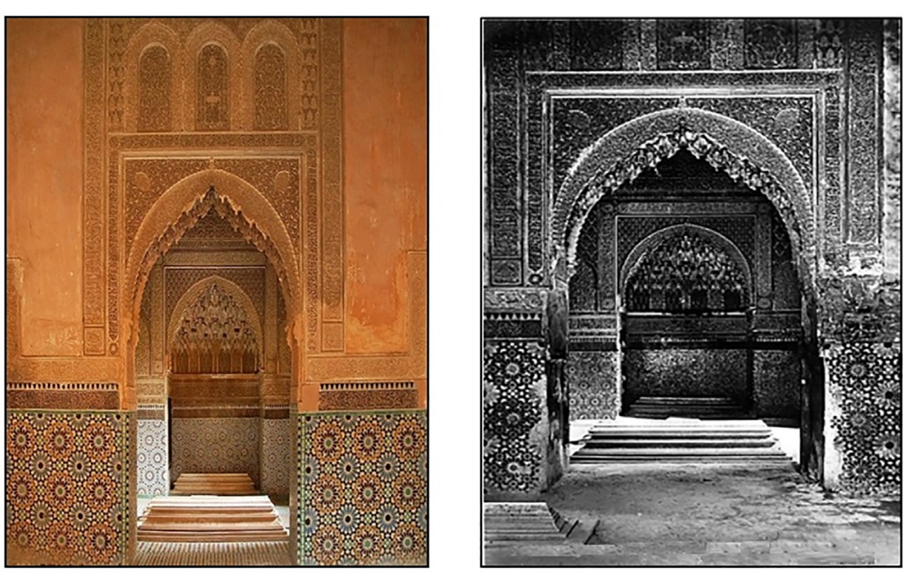

Fig. 1. The Kouba of the Saadian tombs in 1917 (left) and 2018 (right).

Table 1
Overview of the historical classification, pXRF instrument, ceramic body and number of glazed studied samples.

pXRF instrument	Century	Number of glazed ceramic studied	Ceramic body
Bruker AXS portative analyzer instrument	Original glaze (16th)	38	Quartz
	First restoration (20th)	53	Quartz
	Second restoration (21th)	31	Quartz
Thermo Fisher Scientific Niton XL3t GOLDD instrument	Original glaze (16th)	11	Quartz
	First restoration (20th)	7	Quartz
	Second restoration (21th)	5	Quartz
Thermo Fisher Scientific XL3 Hybrid device (Gradmann, 2016)	Original glaze (16th)	10	Quartz

Factoextra under the R Software. The statistical methods used are the principal component analysis (PCA) and Hierarchical Classification on Main Components (HCMC) (Husson et al., 2010).

3. Results and discussion

3.1. Degradation state of Saadian glazed ceramics

Despite the efforts made during the restoration period, most of the archaeological glazed ceramic are affected by different kinds of soluble salts, as well as by structural problems endangering their integrity. The original and the Zellige used in the first restoration period are suffering from a glaze detachment for the majority of the colors (Fig. 2), this phenomenon generally appears when the glaze is applied without slip layer which will ensure its adhesion to the ceramic shard and when the pieces have not reached the optimum threshold of firing. This type of degradation is characterized by a loss of enamel that leave the ceramic paste exposed particularly at the edges and extremities of pieces.

The original and the Zellige currently used in the restoration are very affected by cracks; this type of degradation comes generally from a phenomenon of expansion and contraction of the non-proportionally vitreous coating to the Zellige ceramic support (Tite et al., 1998; Atkin, 2015). The samples are increasingly affected by cracks when the glazes are relatively rich in alkaline with lacking lead content (Henshaw et al., 2007). These cracks may be more or less wide and they can appear right out of the kiln or later.

Fractures, gaps, abrasions and scratches are other alterations often found in all type of Zellige of the two sites, they can affect the glaze and also the ceramic paste. They are caused by the frequent passage of visitors, by the movement and the friction of pieces. These alterations can lead to the total or partial disappearance of the glaze.

In the absence of an archiving policy and documents in these historical sites, it is difficult in some places to distinguish between the three types of Zellige or to identify the location of original materials in order to create a link between the type of materials and the degradation factors. The majority of alterations and defects identified are mainly related to external factors, so they cannot serve as a tool of distinction between the Zellige of different periods. The chemical composition of the glaze is therefore a specific element to each type of material by which the distinction between the Zellige of the Saadian tombs will be possible.

3.2. Identification of the glaze components

Binary diagrams of CaO-PbO, Fe_2O_3-PbO, K_2O-PbO and SiO_2-PbO (Fig. 3) show that glazes contain an average of 34 wt% of lead, this element ranges between 9 wt% and 59 wt%. White glaze represents the highest levels of lead (25–59 wt%). Iron and lead are positively correlated for the most part, especially in black and yellow glazes. Lime shows negative correlation while other oxides do not correlate very closely with the lead oxide. Several studies show that ancient ceramists often use lead mixed with tin as opacified agent (Hochuli-Gysel, 1977; Hatcher et al., 1994; Marraki, 1998; Molera et al., 2001; Gliozzo et al., 2015). Lead oxide is also used in glazes as a flux. Results present fairly consistent compositions for silica and lead, SiO_2 ranges between 35 wt % and 68 wt% with an average of 51.5 wt%, the same results was obtained by Gradmann et al. (2015) for some samples collected from the same building (Fig. 4). The use of lead as flux in association with alkalis promotes the expansion properties during firing and increases the hardness of the shard and makes the color more light (Henshaw et al., 2007).

3.3. Identification of the colored glazes

Green glazed Saadian Zellige is a lead-glazed earthenware; lead oxide was the principal flux in the glaze often mixed with quartz. The

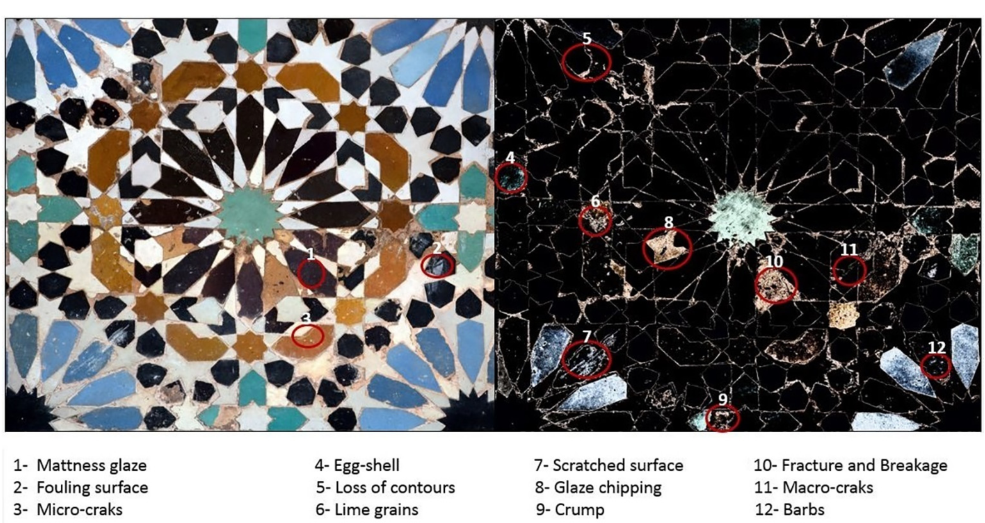

Fig. 2. Defects and alterations of archaeological Zellige in the Saadian Tombs site (Natural picture on the left and image in panchromatic mode on the right side).

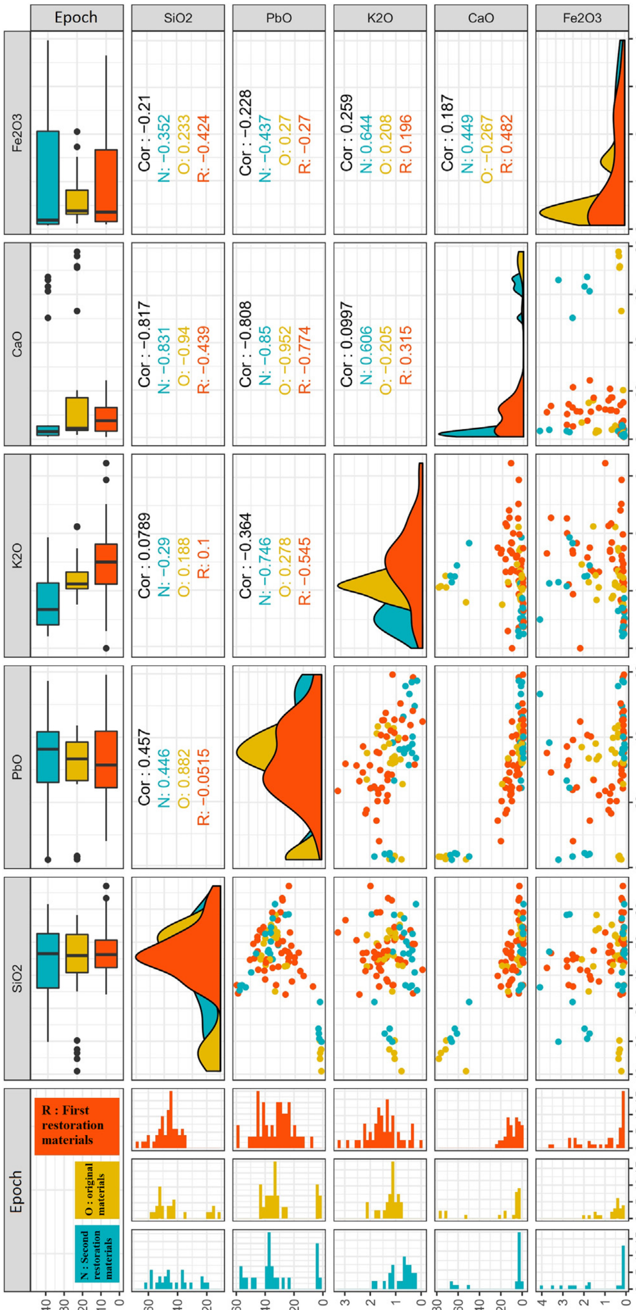

Fig. 3. Binary diagrams showing the degree of correlation between the main chemical elements components of glazes.

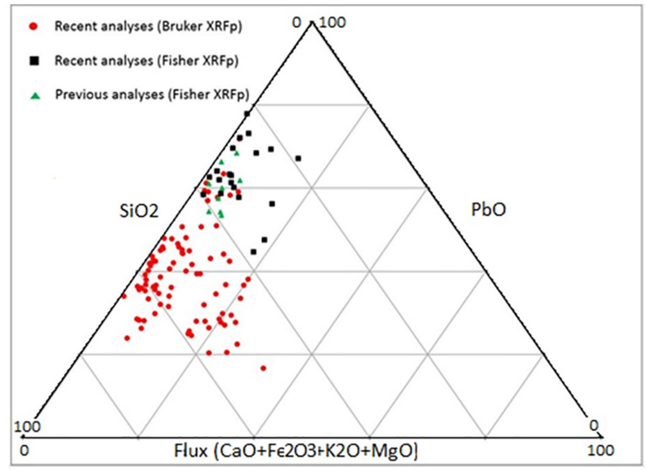

Fig. 4. Ternary diagram PbO-SiO_2-flux (CaO-Fe_2O_3-MgO-K_2O) indicating the high amount of lead and silica in Saadian glazes.

polychrome effect was obtained by using as coloring agent copper (Cu) (Fig. 5A). Copper oxide (CuO) and carbonate ($CuCO_3$) are used to give green in oxidizing firing conditions for the most islamic glazes from Morocco, Iraq, Iran, Egypt and Syria (Tite et al., 2008; O'Kane, 2016). The copper content in the green glazed Zellige ranges between 1.5 wt% and 12 wt%. The Cu^{2+} ion colors the alkali-rich glass light blue, whereas in a lead glass with more than 30 wt% of PbO, it turns into green (Weyl, 1967; Wedepohl, 2003). Glazes with a copper content lower than 5 wt% have a bright color. Above 5 wt%, the surface becomes dark or metallic. The copper oxide begins to volatilize from 1225 °C, this temperature is never reached for Moroccan glazes because the entire ceramic shard will be melt. Cu^{2+} ions escape from the glaze as a vapor that can influence the color of the other parts in the kiln. In reduction, it gives the famous red of copper (Rhodes, 1978).

Yellow glazed Saadian Zellige are iron-rich, this coloration was obtained by Fe^{3+} ions. Iron oxide is one of the most important of the coloring oxides (Fig. 5B). The transparent glazes placed on the clay with iron in the oxide state, reveal colors brown, reddish brown or yellow, the great variety of nuances depends on the quantity of iron oxide containing in the clay, the type of glaze, atmosphere and degree of firing. The iron content in these samples ranges between 1 wt% and 5 wt%. About 1 wt% iron oxide is enough to fade a glaze faintly, 3 wt% gives a medium shade, 5 wt% sustained tones and more than 7 wt% dark browns and blacks. With glazes containing a high among of Pb the iron crystals can be brightly colored in yellow (Gill *et al.*, 2014). As mentioned before, the Pb amount acts as flux, but also has a coloring agent in the copper-colored green glazes and the yellow glazes above 50 wt% of PbO (Wedepohl et al., 1995; Nassau 2001).

Black glazed Saadian Zellige is primarily colored with MnO_2 and additional Fe2O3 (Fig. 5C). The blend of iron oxide with manganese gives a black shade whatever the firing atmosphere with beautiful slightly matt or crystalline surfaces. About 2 wt% of the iron oxide is enough to give black color with most lead glazes. MnO_2 used can be explained with the geographical proximity to the manganese deposits near to Marrakech.

White glazed Saadian Zellige shows no trace of metal oxides; none of the common coloring ions of Mn, Cu, or Fe are present in these samples. White glazes contain only calcium, silica and lead-rich precipitates. Previous studies show that the white glaze in the Saadian monuments (e.g. El Badi Palace) is obtained by cassiterite (SnO_2) mixed with lead and silica (Gradmann et al, 2015; El Halim et al., 2018), but tin oxide cannot be detected by the Bruker AXS handheld analyzer instrument used (device detection limit). CaO can be used as whitening agents like SnO_2 but the higher lime contents observed in some white, black, yellow and green glazes (> 5 wt% of CaO) could also be due to efflorescence of the surface when the shard was made from calcareous clays (Walton and Tite, 2010).

Blue glazed Saadian Zellige shows moderate content of phosphorus P_2O_5 especially in the recent (2.5 wt% to 4.9 wt%) and original Zellige (6.5 wt% to 9.8 wt%), the phosphorus content in the Zellige of the first restoration is between 0.5 wt% and 2.5 wt%. oxides of phosphorus were known to give milky opalescence in glazes, and also to encourage the blue-turquoise color (Fig. 5D). In the Saadian Zellige of El Badi Palace, the analysis of blue glazes using a Panalytical Axios XRF spectrometer shows that phosphorus is the main agent responsible for this color with a content around 7.5 wt% (El Halim et al., 2018). The contents of silica and lead in these samples range respectively from 42.8 wt% to 62.6 wt % and 25.5 wt% to 50.2 wt%. Generally, the distinction between the two possible origins of P_2O_5, namely plant ash and mineral phases (often carbonated) is based on the values of MgO, K_2O and CaO (Gulzar

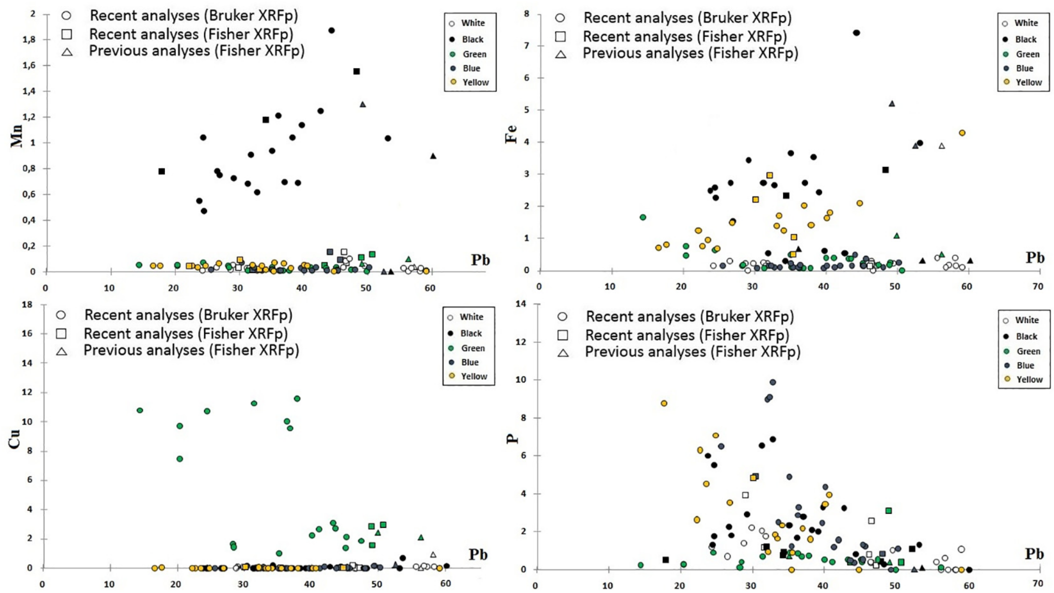

Fig. 5. The main chemical agents responsible for the colors of the Saadian glazes (A-green, B-yellow, C-black and D-blue).

et al., 2013). The ashes contain high MgO and K_2O and variable contents of P_2O_5. The MgO and K2O contents in the Saadian glazes does not exceed 2 wt%, the probable source of the phosphorus is therefore mineral phases such as apatite or other carbonate minerals very widespread in the high Atlas of Marrakech.

3.4. Evolution of the coloring agents of glazes over time.

The principal component analysis (PCA) for each color of the studied glaze samples revealed different groups according to their chemical composition (Fig. 6):

The white glazes can be individualized in three classes:

- The Class 1(W3, W4, W5 and W6) is characterized by high values of the variables P_2O_5, SiO_2 and K_2O and low values of the variable MnO.
- The Class 2 (W7, W16, W17, W18 and W19) is characterized by high values of the variables Ni, Cu and PbO and low values of the variable K_2O.
- The Class 3 (W10, W11, W12, W13 and W14) is characterized by high values of CaO, Al_2O_3, TiO_2, MnO, Fe_2O_3, Cr and MgO variables, and low values of the PbO, SiO_2, Ni, P_2O_5 and Cu variables.
- The results show that the contribution of certain elements in the white glaze has changed over time; the original glazes are much richer in calcium, aluminum and iron compared to silica and lead, which gives the Zellige a matte and bland appearance. The Zellige of the first restoration is rich in silica, potassium and phosphorus with moderate levels of lead. Content of calcium has decreased between the two periods, resulting in a glossy glaze with Arctic white color. The use of lead oxide has increased in the glazes currently used in the restoration, with a significant contribution of silica; this is related to the firing temperature used in current kilns.
- The green glaze is composed of two classes:
- The Class 1 (G15, G16, G17, G18 and G19) is characterized by strong values of the Cu and P_2O_5 variables and low values for the variables K_2O and MnO.
- The Class 2 is composed of individuals sharing and characterized by high values of the variable SiO_2, CaO, K_2O and PbO.
- Class 1 corresponds to the Zellige currently used in the restoration of Saadian historical buildings. The Cu content in the green glaze increased 3 times more than the value found in the original glaze. The general appearance of glazes that belongs to this class is a glossy dark green glaze. The calcium oxide does not exceed 1.6 wt% in most of these samples. The second class includes the old and the first restoration Zellige; these samples are relatively rich in lead, silica and potassium. The copper concentration used was around 3 wt%. This class is subdivided into subclasses according to CaO and K_2O contents as shown in the Cluster Dendrogram (Fig. 6B).
- The blue glaze can be individualized in three classes:
- The Class 1 characterized by high PbO values and low P_2O_5, CaO and K_2O contents.
- The Class 2 (B15, B17) characterized by high and variable SiO_2, moderate values of the P_2O_5 and low values for the variable CaO.
- The Class 3 is composed of individuals such as B11 and B12, this group is characterized by high values for the variables P_2O_5 and CaO, and low values for the variable PbO.

For the blue glaze, phosphorus was used in high concentration by Saadian artisans in 16th century, the calcium oxide content is still relatively strong in these glazes, and the result is a sky blue matte Zellige. In the 20th century, the phosphorus values detected in the glaze do not exceed 1.25 wt% (Class 1), whereas lead is present in high concentration. The recent Zellige contains moderate levels of phosphorus around 3.5 wt% (Class 2) with high silica value, the obtained blue is different with a dark and shiny appearance.

The black glaze is composed of two classes:

- The Class 1 is composed of individuals such as D4, D6, D7, D8, D9 and D11. This group is characterized by high values of the variables Fe_2O_3, P_2O_5 and CaO, and low values for the variable PbO.

- The Class 2 (D12, D13 and D14) is characterized by high values for the variables SiO_2 and MnO.

Black restored glazes (Class 1) are described to have relatively low contents of MnO $<$ 1 wt% with 2–7 wt% high P_2O_5 values. The black color of these samples was obtained by a mixture of iron oxide 2–4 wt% and CaO 5–14 wt% as flux. The original black colored ceramics are silica and lead oxide dominated glazes with 42–55 wt% of SiO_2 and 27–42 wt% of PbO. The contribution of MnO oxide is greater than iron in the glaze, the CaO content does not exceed 5 wt% in the majority of these samples. The iron content in the Zellige currently used in the restoration ranges between 2 and 4 wt%, whereas the manganese content is around 1 wt%.

The yellow glazes can be individualized in three classes:

- The Class 1 is composed of individuals such as Y16, Y17, Y18, Y19

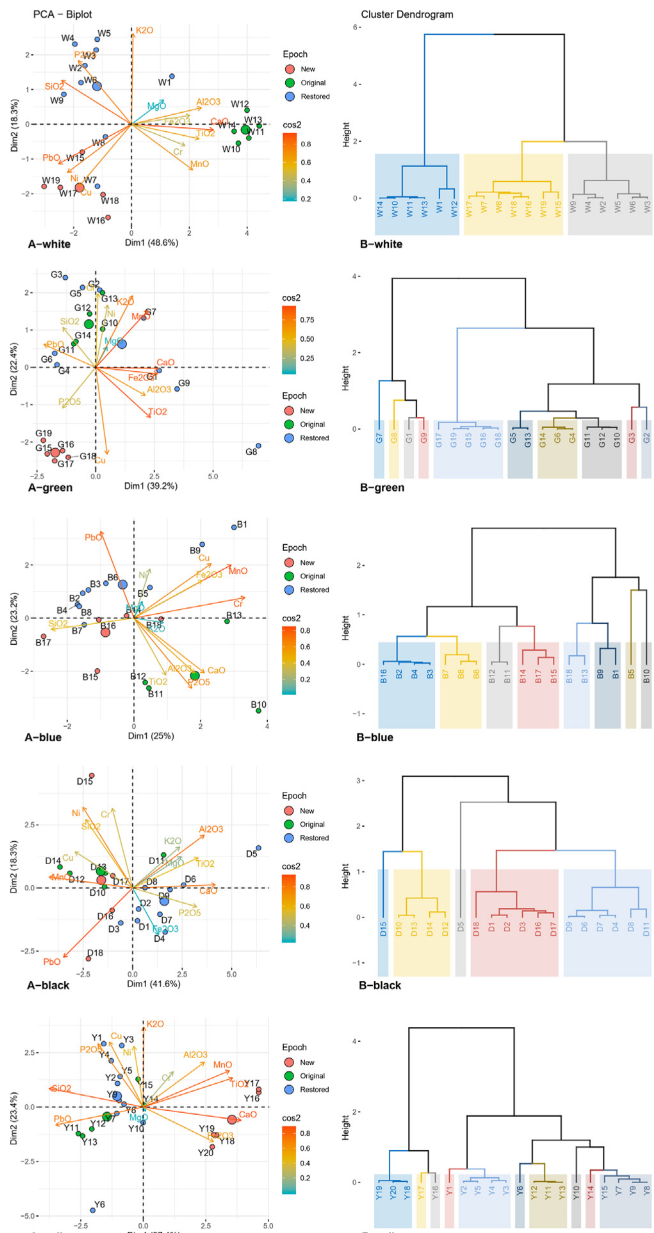

Fig. 6. Principal component analysis (A) and Cluster Dendrogram of the studied Saadian glazes (B).

and Y20. This group is characterized by strong values for the variable CaO and F_2O_3, and low values for the variables SiO_2, PbO and P_2O_5.

- The Class 2 (Y1, Y2, Y3, Y4, Y5 and Y6) is characterized by high values for the variables SiO_2 and P_2O_5, and low values for the variable Fe_2O_3.
- The Class 3 is characterized by high value for the variable PbO.

The yellow glaze in the all Saadian glazes is affected by Fe_2O_3 content, the Zellige of the first restoration period is the least rich in iron with values around 1.2 wt% (Class 2). The original Zellige has iron values of 1.6 wt% on average, while recent glazes have values around 2.2 wt%. The CaO content in these glazes (Class 1) is very high, which implies that calcium oxide was used as flux instead of lead in these samples. Calcium carbonates can be used as white colouring agent in these kinds of glazed ceramics (Rutherford et al., 1974).

4. Conclusion

The innovative methodology proposed in this study to follow the evolution of the chemical composition of the Saadian glazes is based on a statistical study using the principal component analysis (PCA) and the interpretation of the binary diagrams as well as the establishment of different correlations between the chemical oxides used in the glaze mixture. The results of the experimental work described here involve the following main conclusions:

- All samples are lead-type glazes, as is the case with most Islamic decorated ceramics since many centuries later.
- For original Zellige, lead, silica and calcium oxide were the main elements used as flux; the colors green, blue, yellow and black were obtained by ordinary oxides such as copper, phosphorus, iron and manganese.
- The development of the contemporary ceramic industry has allowed the reformulation of new recipes to obtain glazes with more bright and brilliant colors, this by reducing the CaO levels and the increase of coloring oxides content.

The results have important archaeological implications because the Moroccan ceramic industry is trying to create new formulations for the manufacture of glazes while retaining the appearance and mechanical properties of original Zellige in order to ensure good compatibility between the original and restoration parts.

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