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Characterization of Imitated Marble Used in Historical Buildings in Cairo - Egypt

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AMANY MOHAMED BAKR, MONA FOUAD ALI,
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Characterization of Imitated Marble Used in Historical Buildings in Cairo – Egypt

THE architectural heritage of Mohamed Ali's family period (1805-1952) in Egypt shows strong European influences in decorative styles. Using artificial decorative materials that simulate the natural stones is one of these influences. From the fifteenth century up to the second half of the twentieth century nearly all the European architecture was characterized by the presence of relief decorations made of artificial stone.¹ The material defined as "artificial stone", frequently used on the façades of the late 19th and early 20th century buildings, is a mixture of binder, aggregate and other additives and may either be applied directly as a coating on wall surfaces or precast in moulds and then attached to façades as decorative architectural elements.² In the early 20th century the terms "concrete stone", "cast stone", and "cut cast stone" replaced the term "artificial stone" that was commonly used in the 19th century. Various materials and techniques were used to produce the cast stone that simulate natural stone. Natural cements, Portland cements and sodium silicate cements were all used as binding agents. The differences in the resulted products reflected the different stone aggregates, binding agents, methods of manufacture, curing and systems of surface finishing that were used to produce them.³

The foremost natural decorative stone that inspired the artisanal attention was marble. Luxury aesthetic appearance of marble attracted the artisan either to use it for decoration or to imitate it when the real stone was impractical for many reasons (e.g. high costs and shortage

1. Pecchioni, Malesani, Bellucci, Fratini, "Artificial Stones", p. 227-233.
2. Ersen, Gürdal, Güleç, Yöney, Pekmezci, "An Evaluation of Binders and Aggregates", p. 207-221.
3. Pieper, *42nd Preservation Brief the Maintenance*, p. 1-16.

of some types of marble). Prudon and Benhamou,⁴ reported some techniques and materials that were developed and applied as marble-imitating wall decoration during 19th and 20th century in Europe.

Several terms were used to describe marble-imitating which was used for casing the interior and exterior wall. Stucco marble or artificial marble is the term used for a mixture of gypsum, pigment, water and animal glue utilized to imitate the natural marble for the decoration of building interiors.⁵ Scagliola is the Italian definition of stucco marble that is an inlay work of various colored, smoothed and polished stucco pastes. In addition to recreating decorative drawings using the technique of scagliola, it was used for the decorating the large architectural areas to imitate the natural stone.⁶ This technique originated in Italy in the 17th century and was popular as a relatively cheap substitute for marble in Europe during the 18th century.⁷

In Egypt, different materials and techniques were used for casing the exterior and interior walls to imitate marble during the 19th and 20th centuries. The resulted imitated marble in some cases was not as skillful as those produced in Europe. Whatever the composition of the imitated marble, it commonly exhibits deterioration that is caused by the interaction between intrinsic factors (nature and form of the artifact) and extrinsic factors (the environment at which the artifact is exposed). The restoration of historic imitated marble requires knowledge that enables to reproduce materials with physico-chemical and mechanical properties similar to the original ones. The historical literatures dated back to this period of time didn't contain sufficient information about the materials and techniques used for imitating marble. This study aims to characterize the imitated marble materials and techniques used for wall casing in two historical building dating back to Mohamed Ali period in Egypt: *sabīl* Umm 'Abbās and Sa'īd Ḥalīm palace. The main objective is to identify the mineral composition of the mixture used and to shed light on the applying process.

Materials and Methods

Materials

Sabīl Umm 'Abbās

The Arabic word *sabīl* stands for public drinking fountain. This *sabīl* was built in 1867 by Khedive 'Abbās's mother. The left and right sides of the main octagonal façade are decorated with marble-imitating wall casing that simulate the white veined marble (fig. 1a and 1b). The imitated marble casing shows various deterioration phenomena such as exfoliation, blackening and cracks in different sizes and shapes (fig. 1b). Separation of the facing layer from the stone background is dispersed all over the wall casing. Three representative samples were collected.

4. Prudon, "Simulating Stone, 1860-1940: Artificial Marble, Artificial Stone, and Cast Stone", p. 79-91; Benhamou, "Imitation in the Decorative Arts", p. 1-13.

5. Wittenburg, "Introduction to the Project, in: Baroque Artificial Marble", p. 7-8.

6. Berner, Weber, "Stucco Marble", p. 11-20.

7. Fawcett, *Historic Floors*.

The first sample represents the stratigraphic structure of the stucco used to case the wall. The second sample represents the outer white surface layer, which contains white area and white area veined with black color. The third sample represents the subsurface layer (middle layer).

Sa'īd Ḥalīm Palace

This palace was built during the period from 1899 to 1901. As attested by the superimposed monograms "SH" on the many decorative pillars, the owner was prince Sa'īd Ḥalīm Pasha, a grandson of the great Wali of Egypt Mohamed Ali. The palace was designed and constructed by the Italian architect Antonio Lasciac. It presented novelty in style and magnificence in external decoration of fake marble. Most of the decorative elements used in this palace were imported from Europe⁸ (fig. 2a). Red tiles veined with white and black colors, which are imitated the natural veined red marble, were used for casing the exterior wall of the palace façades. The tiles are rectangular and their dimensions are about 25 cm × 50 cm. At some places in which the tiles are missing more information could be obtained. The total thickness of these tiles is about 3 cm. Two main layers can be recognized, the facing imitated marble-texture layer (its thickness is ranging from 5 mm. to 8 mm) and the backing gray plane coarse layer. The common deterioration phenomenon of the red veined tiles is the disfiguration caused by the accumulation of dust and the airborne particles. Separation of the facing layer of the veined red tiles from the backing layer is seen in some area of the palace façades (fig. 2b). Reticulate cracks are seen in some tiles occupied the lower part of the façade. A representative sample was collected from the veined red imitated marble-texture layer of the wall casing tiles (fig. 2c).

Analytical Methods

Various analytical techniques were applied in order to obtain an exhaustive description of the studied samples' composition.

Optical Microscopy

Preliminary morphological observation of the raw surface and polished cross section of the *sabil* and palace samples were carried out using a Zeiss optical light microscope. Thin sections of the samples were examined under a Nikon polarized transmitted light microscope (model opti photo x23, equipped with photo camera S23, under 10x and 20x magnification in plane-polarized light).

Scanning Electron Microscopy

Scanning electron microscopy (SEM) investigations of raw samples and polished cross sections were carried out by a Philips microscope (model XL30), equipped with an EDX micro-analytical system by which information on the total element contents in samples was obtained. Images were acquired in backscattered mode (BSE).

8. Negm, *Palaces of Princes and Pashas of the Mohamed Ali Family.*

X-Ray Diffraction

Fine powders of the samples were analyzed with a Philips diffractometer (model PW 3071, CuK α 40 kV, 30 mA). The scanned 2 θ range was 5 to 60 degrees.

Fourier Transformed Infrared Spectroscopy

The samples were analyzed as KBr pellets by a JASCO FT\IR spectrometer (model 460, 400 – 4000cm⁻¹, 4cm⁻¹ resolution), in the transmission mode.

Results and Discussion

Sabīl Umm ‘Abbās

The preliminary observation of the collected samples showed the stratigraphic structure of the marble-imitating stucco. The polished cross section examination revealed the presence of three main layers (fig. 3a): inner, middle and outer layer (finishing layer). Secondary layers are seen within the middle layer of some samples.

The PLM examination of the thin cross section showed the very compact fine grains structure of the layers. The outer layer is light colored and transparent under the plane polarized light (PPL). The middle layer is characterized by the dark opaque color and high density. The gradually alternation between low density light colored and high density dark colored is well observed within the contact area between this layer and the outer layer (fig. 3b). The high density of the middle layer is most probably due to the high content of glue that was used as an organic binder in this layer (corroborated by FTIR analysis). The outer layer is less dense than the middle layer, which indicates the low content of organic binder. Aggregates of secondary gypsum are seen within the outer and middle layers. The presence of secondary gypsum is most probably the result of remobilization of the primary gypsum or weathering alteration of calcite that are both intentionally used in the execution of the layers.

The polished cross section of the studied sample was examined by SEM-EDX. The back-scattered electrons (BSE) image (fig. 4a) displayed the stratigraphic structure and the inhomogeneous elemental composition of the sample. The SEM examination of the sample representing the outer white surface layer veined with black color showed the disintegrated cracked surface of sample (fig. 4b). The thickness of this layer ranges between 75 to 183 μ m. The total EDX analysis of the samples showed that zinc (Zn), carbon (C) and lead (Pb) are the dominant elements in the white area of the surface layer (fig. 4c); their values are ranging between 15% to 20%, 14% to 16% and 5% to 15% respectively. Small amount of sulfur (S), calcium (Ca) and silicon (Si) (their values are ranging between 4% to 5%, 2% to 5% and 1% to 2% respectively) were recorded. Carbon (C) is the dominant element in the white area veined with black color sample, its value is 53%. Iron (Fe) is recorded in some area of black veins. The EDX analysis of the subsurface layer (the middle layer) (fig. 4d) showed a wide variety of the elements amounts from area to another within this layer, due to the interference between the outer, middle and inner layer. Carbon (C), lead (Pb), calcium (Ca), zinc (Zn), sulfur (S) and

barium (Ba) are the dominant elements (their highest values are 50%, 31%, 24%, 22%, 19% and 18% respectively). The inner layer which contacts directly with the limestone background wall, seen in the polished cross section (fig. 4a3), is composed mainly of calcium (Ca), sulfur (S) and carbon (C), (their values are 24%, 19% and 16%, respectively). Small amounts of Na, and Cl, were recorded all over the samples.

The XRD analysis was performed on the detached samples (Table 1). The XRD analysis of the white surface layer veined with black color showed that the major minerals are zincite (ZnO) and cerussite (Pb CO₃) and the minor mineral is gypsum (CaSO₄.2H₂O). The XRD analysis of the subsurface layer indicated that the major minerals are barite (BaSO₄), calcite (CaCO₃) and gypsum (CaSO₄.2H₂O). Small amount of zincite (ZnO) and cerussite (Pb CO₃) were also detected in this layer. The XRD of the inner layer in contact directly with the limestone wall showed the presence of calcite and gypsum.

The FTIR analysis of the collected samples was carried out to check the presence of an organic binding media in the paint layers. The FTIR spectra of the outer and the middle layers' samples showed a very broad band at 3405 cm⁻¹ due to intermolecular hydrogen bonding O-H stretching. This band overlapped N-H stretching. The bands at range of 2925-2853 cm⁻¹ correspond to the saturated hydrocarbon chains (C-H) stretching vibration. The broad band at 1428 cm⁻¹ is due to combination between C-H bending vibration of amide III and carbonate group (CO₃) stretching band. The band at 1619 cm⁻¹ corresponds to the carbonyl (C=O) stretching vibration of amide I. The band at 1550 cm⁻¹ corresponds to the combination between (C-N) stretching and (N-H) bending of amide II. All these bands are comparable with those of animal glue.⁹ All the function groups of sulfates, related to gypsum, barite, and carbonates, and/or related to calcite and cerussite, were also detected.

The mineralogical compositions of the studied samples showed that the used stucco consists of different strata. The outer finishing layer, white paint, composed mainly of zincite (ZnO) known as zinc white. Zinc white (ZnO) was manufactured and first introduced as an artist's pigment in France in the 1830s.¹⁰ Cerussite was added to this layer to improve the covering power of zinc white and to accelerate its drying process.¹¹ Subsurface layer composes of barite, calcite and gypsum. Calcite and gypsum were used as ground before applying the primer layer which is composed mainly of barite. It is known that barite (BaSO₄) (also known as "blanc fixe") was used as an extender and filler for paint.¹² The detection of Fe with large amount of carbon in the white surface layer veined with black color indicates that a mixture of amorphous carbon black and iron-oxide black was used to obtain the black veins. This kind of mixture was used to obtain black solid paint layer.¹³ From the examination of the studied samples the used technique was concluded. A thin ground layer composed of calcite and gypsum was applied directly on

9. Michele, Dusan, James, *Infrared Spectroscopy in Conservation Science*.

10. Welsh, "Identification of 1850s Brown Zinc Paint", p. 17-30.

11. Gettens, Kuhn, Chase, "Lead White", p. 67-78.

12. Laar, Burnstock, "With Paint from Claus & Fritz", p. 1-16; Franquelo, Duran, Herrera, Jimenez de Haro, Perez-Rodriguez, "Comparison Between micro-Raman and micro-FTIR Spectroscopy Techniques", p. 404-412.

13. Garcia, Florez, "The Nasrid Plasterwork at 'Qubba Dar Al-Manjara L-Kubra' in Granada", p. 75-89.

the limestone wall, then the primer layer which is composed of barite was applied. The white finishing layer composed mainly of zincite and cerussite was applied above the previous layers. The animal glue was used as a binding media in the middle layer and in the outer layer. The content of glue used in the middle layer is higher than that used in the outer layer. The tile-imitating black border and black veins were drawn on the white finishing layer to imitate the veined white marble.

Sa'id Halim Palace

The collected samples are characterized by the strength and light weight. The surface of the imitated marble-textured layer is very dense and smooth while the back side is rough. The interference of red, black and white colors that form the veins of the tiles is seen on the surface and within the cross section of marble-textured layer. Significant amounts of spherical and irregular air voids in different sizes can be seen with naked eye within the cross section (fig. 2c).

The examination of the veined red with black and white colors thin section under the polarizing light microscope displayed the gel formation (colloform) texture of very finely microcrystalline matrix composed mainly of carbonate (calcite) and silicates (fig. 5a). The matrix that represents the red area is stained with red iron oxides (hematite). Coarser crystals (100µm to 200 µm) of unstained calcite are seen in the matrix indicating that hematite was mixed with unstained powder of limestone to prepare the red mixture. Thin veinlets built of secondary gypsum are dispersed within the matrix. Spherical air voids (ranging from 50µm to 200µm) are also seen in the matrix. The contact area between the red, white and black color are characterized by wide zigzag form. The presence of these textures indicates that colloid solution was used in the execution of the veined red imitated marble-texture layer. The gradational texture of the dark red and light red observed in the veined red thin section is most probably the effect of the gradually addition of the various hues of red colloid solution. Dark opaque angular grains of magnetite (ranging from 50µm to 300µm) are dispersed within gray matrix that represents the black veins in the veined red tile (fig. 5b).

The polished cross-section of the veined red samples was examined by SEM-EDX to distinguish between the elemental compositions of different hues of colors. The back-scattered electrons (BSE) images shows fine-grain matrix with numerous coarse grains of iron oxides. Rounded dark-grey grains, composed mainly of Ca, Si, C and O, represent probably calcium silicate hydrate phase, are seen within the lighter area composed mainly of Ca, C and O that represents carbonate phase (fig. 6a, b). The results of microanalysis in different areas of the studied sample indicated that carbon (C), calcium (Ca), iron (Fe) and silicon (Si) are the dominant elements in the samples. Their values are ranging between 10% to 80%, 2% to 54%, 2% to 59% and 2% to 9% respectively. Their ratios varied significantly between different areas of the samples depending on the interference of various hues of colors (red, black and white). Small amounts of, sulphur (S), aluminum (Al) and magnesium (Mg) were recorded throughout the samples (their values are ranging between 0.5% to 4%, 1% to 5% and 0.5% to 2% respectively. Small amount of chlorine (Cl) and sodium (Na) were also recorded.

The X-ray powder diffraction pattern (fig. 7) of the studied sample shows the presence of the following mineral phases: calcite (CaCO_3), as a prevalent mineral, quartz (SiO_2),

Hematite (Fe_2O_3), gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and Tobermorite [Calcium Silicate Hydrate ($4\text{CaO} \cdot 5\text{SiO}_2 \cdot 5\text{H}_2\text{O}$)]. Tobermorite is one of the hydraulic mineral phases that are usually identified in Portland cement.^{14, 15} The FTIR analysis of the studied samples showed the absence of any organic matter. Only the functional groups of carbonates, related to calcite, and silicate, most probably related to Tobermorite, were detected.

On the bases of the mineral composition and the microscopic examination, it is suggested that a mixture of very fine powder of limestone (unstained and stained with red iron oxides), very fine sand and white cement colored with red and black iron oxides pigments was used to execute the red veined imitated marble layer of the tiles. The gypsum detected in the sample is most probably related to the white cement used. It is known that gypsum is added to the cement clinker before grinding during production in order to control the setting of the cement mortars.¹⁶ The air voids in different shapes (spherical and irregular) and sizes usually indicate to the high content of water used in the preparation of the mixture.¹⁷ The gel formation (colloform) texture noticed in the petrographic examination confirms the use of colloid solution (high content of water) in the execution of the red veined layer. Depending on the petrographic features and the mineral composition it can be suggested that the red veined imitated marble tiles are manufactured by the wet cast process or molding process and it most probably belongs to the decorative cement tiles elements. The cement tiles were invented during the second half of the 19th century in Europe. During this period of time the diffusion of the new hydraulic binders, such as white cement, made it possible to manufacture elements of particular hardness and durability with the aim to imitate the stone.¹⁸ The manufacture of the veined red layer was based on the pouring of hydraulic colored mixture into a main mold that it was in the same size of the tile. The mixture composed of fine powder of limestone, very fine sand and white cement was mixed with hematite to get the red color or was mixed with magnetite to get the black color. Random patterns of swirls with black and white mixtures were poured within the red mixture to create the black and white veins. Each tile has its own pattern of white and black veins. The visual examination of the tile showed that under the veined red layer there is a base layer composed of normal gray Portland cement mortar. The shiny surface seen in some studies samples indicate that the surface of tiles were perfectly polished without using organic matter since no organic substances were identified with the FTIR analysis. The mineralogical-geochemical study of the sample reveals the presence of some mineral composition related to the degradation process. The presence of secondary gypsum veinlets is most probably the result of remobilization of the primary gypsum used in the white cement or weathering alteration of calcite that is represented the main component of the tiles. The volume changes between the mineral phases could lead to disintegration and formation of cracks, degrading the tiles.

14. Charola, and Henriques, "Hydraulicity in Lime Mortars Revisited", p. 95-104.

15. Miroslava, Dalibor, Rudolf, Ludmila, *Proceedings of the 2nd Historic Mortars Conference*, p. 175-185.

16. Ersen *et al.*, "An Evaluation of Binders and Aggregates", p. 207-221.

17. Walker, *Petrographic Methods of Examining Hardened Concrete*.

18. Fartini, and Elena, "The Artificial Stone in Florence Between the XIX and XX Centuries", p. 295.

Conclusions

The analytical investigation of the studied samples supplied all the information about the artistic technique, i.e. the raw materials and the application process used in the *sabīl* Umm ‘Abbās and Sa‘īd Ḥalīm palace. In the white veined imitated marble of *sabīl* Umm ‘Abbās, Zinc white (ZnO) and cerussite were used to obtain the white paint of the surface layer. Barite, calcite and gypsum were used, in various portions, for priming and ground layers. Black veins were drawn on the surface using mixture of carbon black and black iron oxides. Animal glue was used as a binding media in the paint layers. The mineral composition of Sa‘īd Ḥalīm palace samples indicates that a mixture of very fine powder of limestone, very fine sand and white cement colored with hematite and magnetite oxides (to get the red and black colors respectively) was used to execute the veined red imitated marble layer of the tiles used for casing the façades walls of Sa‘īd Ḥalīm palace. The tiles were manufactured by pouring the hydraulic colored mixture into a main mold that it was in the same size of the tile. The white and black veins were created by mixing white and black mixtures within the main red mixture using freehand technique. The body of the tile is composed of normal gray Portland cement mortars. The materials and techniques used in the manufacture of the tiles are comparable with cement tiles that were invented during the second half of the 19th century in Europe. Identification of the main components of the marble-imitating materials is important for their conservation both in terms of diagnosis of deterioration problems, and to aid conservators in the selection of suitable materials for their conservation.

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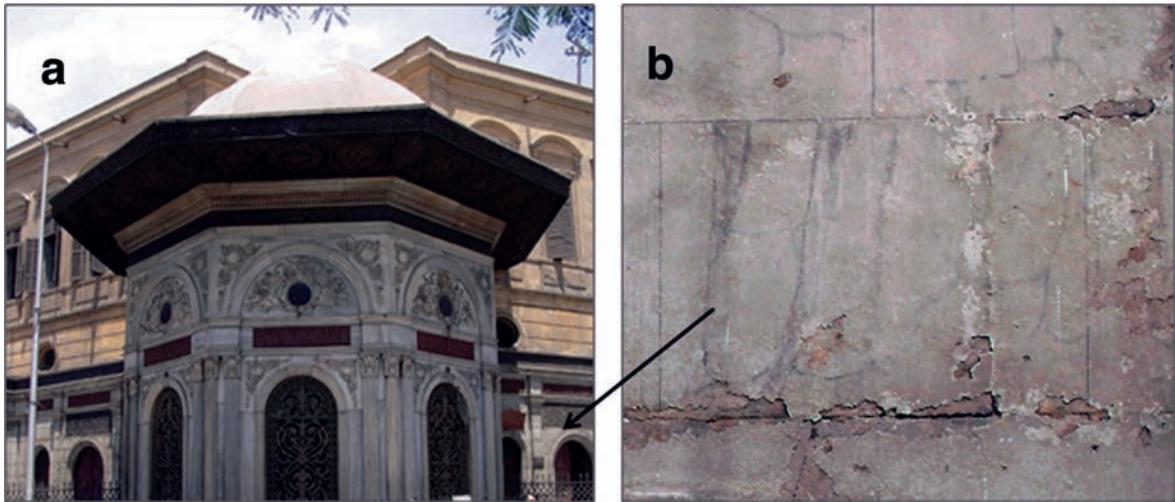


Fig. 1a. The façade of *sabil* Umm 'Abbās.

Fig. 1b. The white veined imitated marble and its deterioration phenomena.

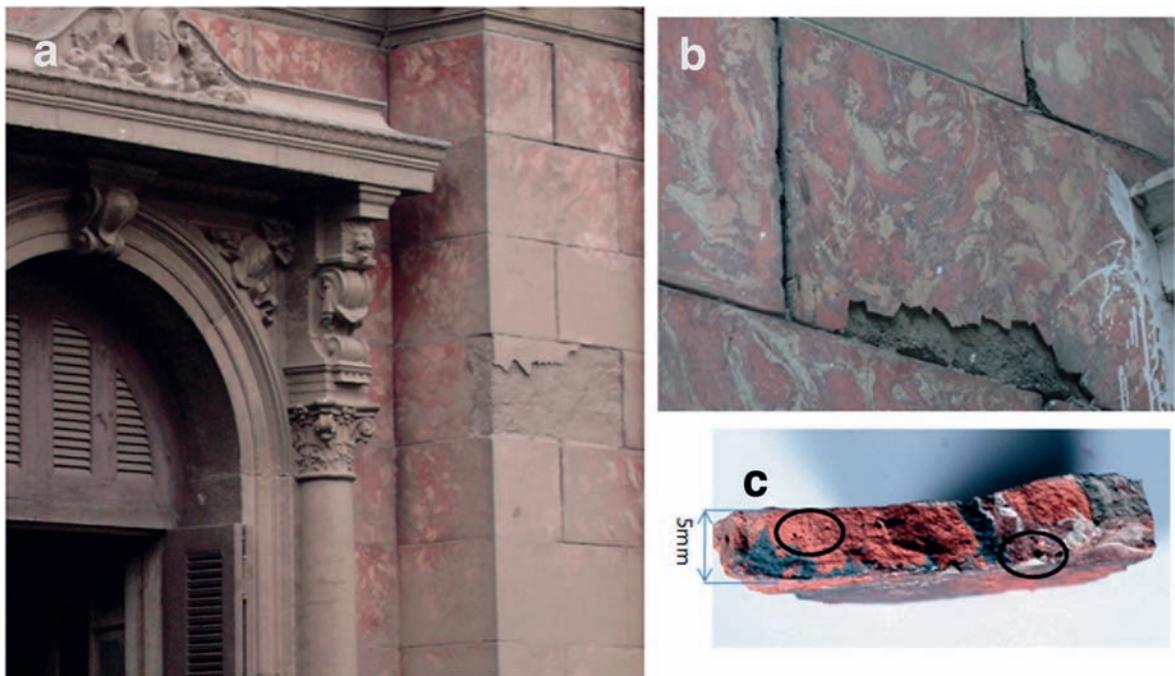


Fig. 2a. Part of the façade of Sa'īd Ḥalīm Palace.

Fig. 2b. The disfiguration and separation of the facing layer of the veined red tiles.

Fig. 2c. The collected sample, the circles are marked the spherical and irregular air voids seen within the cross section.

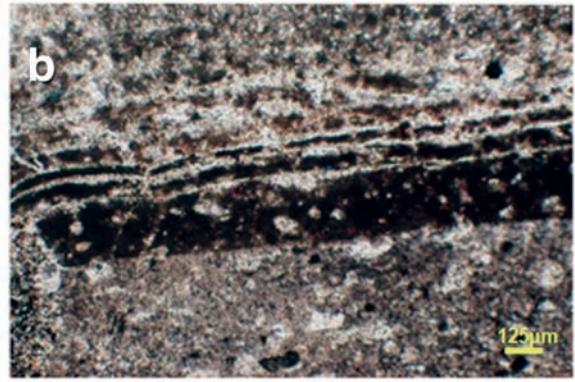


Fig. 3a. The polished cross section of the *sabīl's* sample.

Fig. 3b. Thin cross section image under plane polarised light $\times 20$.

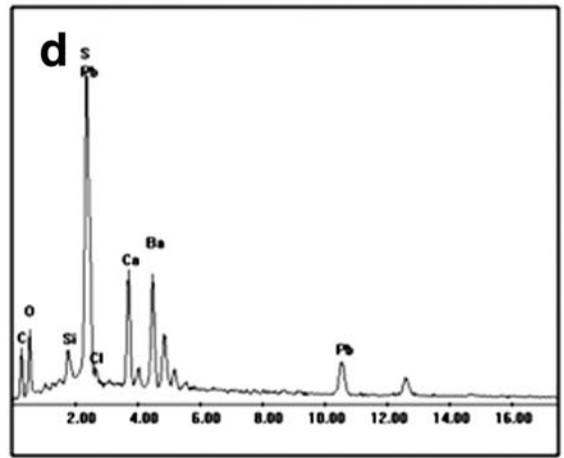
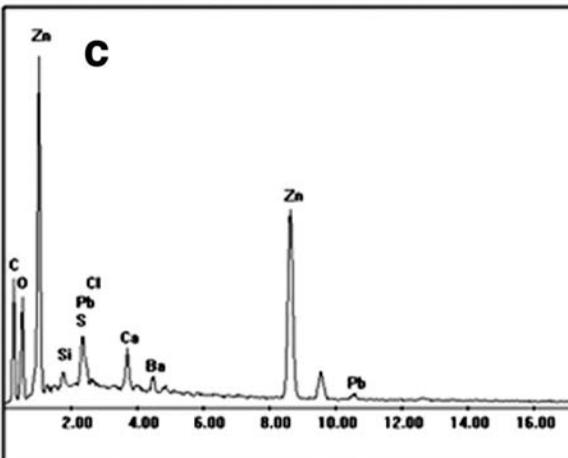
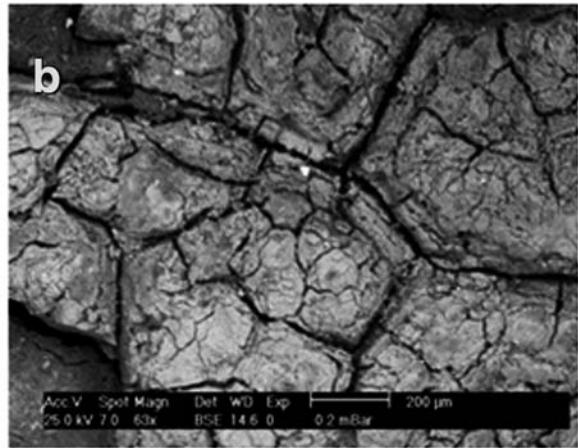
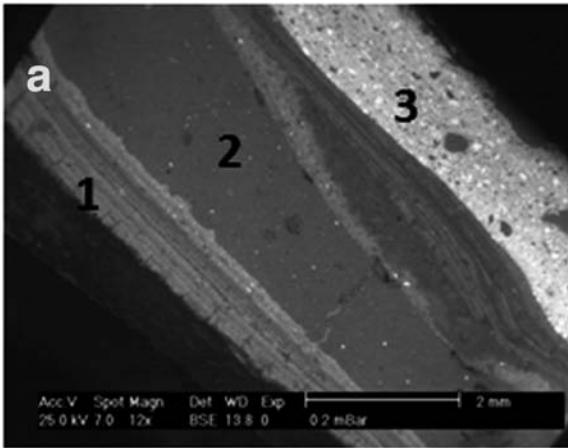


Fig. 4a. BSE image of the polished cross section of the *sabīl's* sample, nos. 1, 2 and 3 point to the outer, middle and inner layers respectively of the imitated marble.

Fig. 4b. BSE image of the outer white surface layer veined with black colour.

Fig. 4c. The elemental analysis (EDX) of the outer white surface layer.

Fig. 4d. The elemental analysis (EDX) of the subsurface layer (the middle layer).

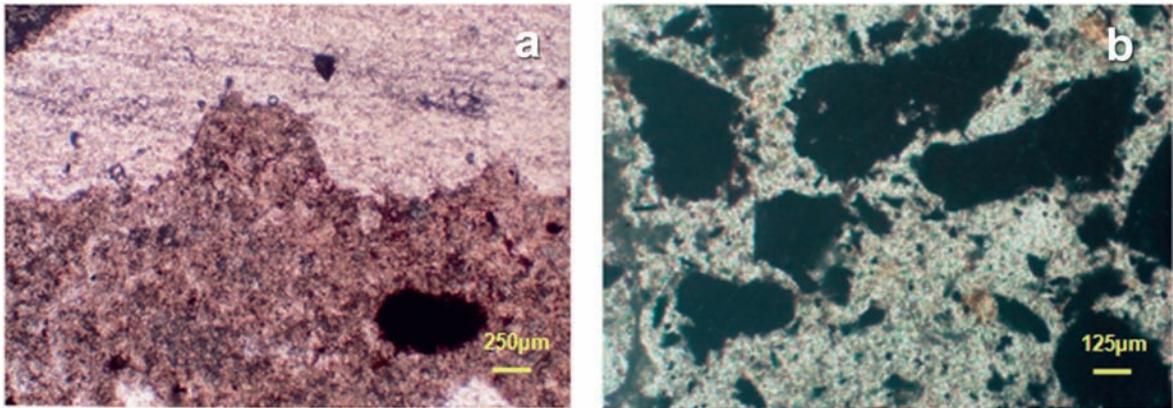


Fig. 5a. The thin cross section image of the veined red sample of Sa'īd Ḥalīm Palace under plane polarised light × 10.
 Fig. 5b. The thin cross section image of the black veins in the veined red tile under plane polarised light × 20.

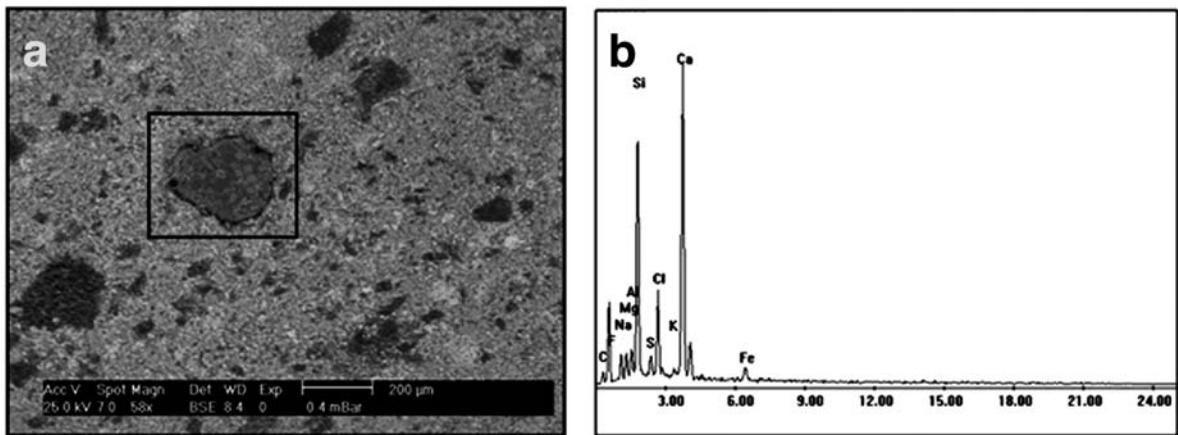


Fig. 6a. BSE image of veined red sample, the square illustrates the grain represented calcium silicate hydrate phase.
 Fig. 6b. Elemental analysis (EDX) of the marked area in the (a) photo.

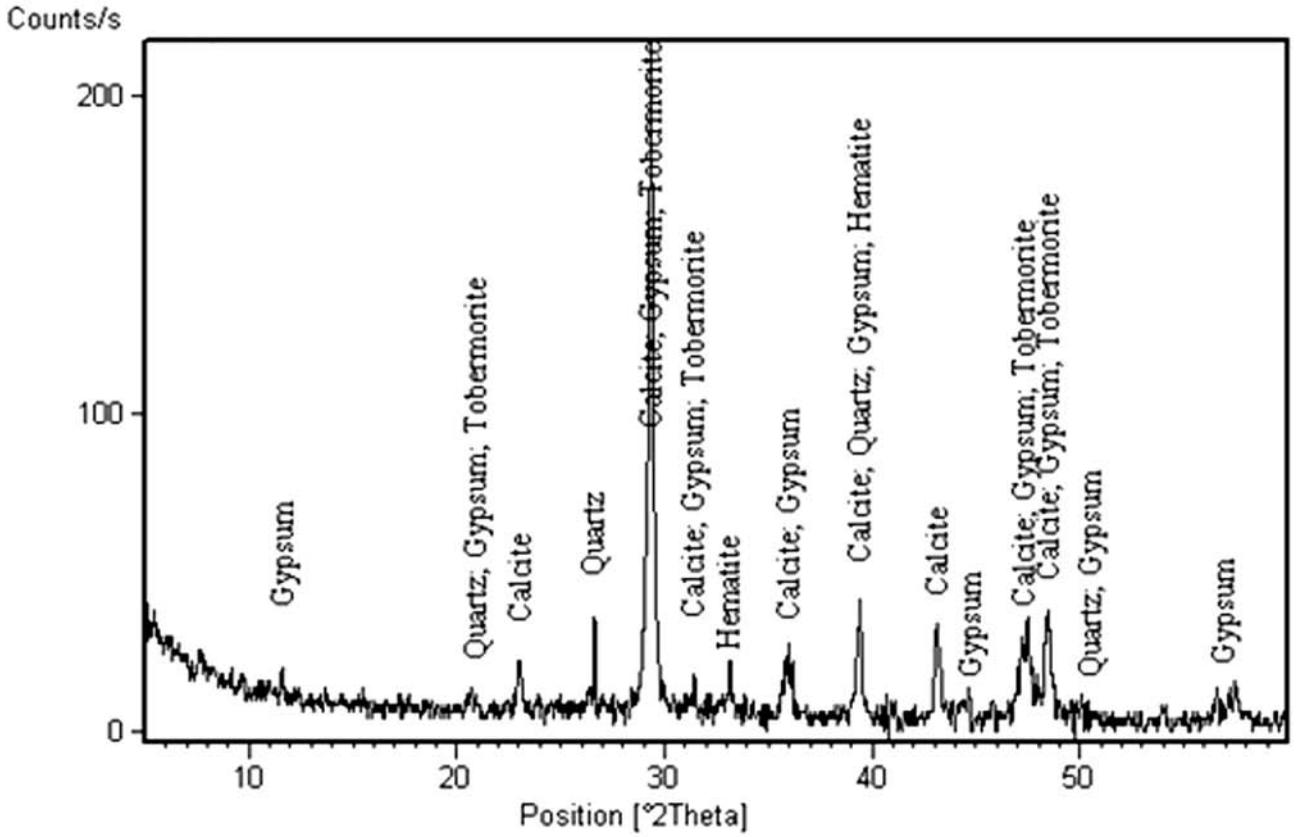


Fig. 7. The XRD pattern of the veined red sample of Sa'īd Ḥalīm palace.

