Egyptian Journal of Archaeological and Restoration Studies
An international peer-reviewed journal published bi-annually
www.ejars.sohag-univ.edu.eg

Original article

THE STONE BLEEDING PHENOMENON AFFECTING SOME SANDSTONE INSCRIPTIONS IN THE KARNAK TEMPLES

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Article info.
EJARS – Vol. 12 (2) – Dec. 2022: 175-186

Article history:
Received: 21-12-2021
Accepted: 1-7-2022
Doi: 10.21608/ejars.2022.276151

Keywords:
Stone bleeding
Sandstone
Loss of the binding material
Karnak temples

***Abstract**:*
Stone bleeding affecting the sandstone artifacts is considered one of the most deterioration forms that dominated the Karnak temples complex. This symptom is a serious deterioration form resulting essentially due to losing the binding material of the stone as a result of the synergistic deterioration factors and their related mechanisms (witting/drying & heating/cooling) cycles. This form is composed of an external hard crust with weak interior components that can be loosed by scratching, causing the bleeding of quartz grains. Some analytical techniques were used, such as SEM-EDX, XRD, and AAS, to define the main essence of this symptom and understand the chemical and mineralogical components. The main results of this study showed that stone bleeding is formed essentially by environmental deterioration factors, especially saline groundwater that causes the weakening of the interior components of the stone and dissolving the binding of its materials.

1. Introduction

In Egypt, many monumental sites and stone buildings have several extrinsic and intrinsic factors that negatively affect the deterioration processes, including physicals, chemicals, and biological. [1]. The bleeding of stone is one of the severest deterioration phenomena, resulting in the complete loss of the monument over time. It is evident in monuments made of sandstone, such as the Nuba, Luxor, Edfu temples, and many places in Karnak temples. It results from the loss of a part of the hard crust resulting from the interaction of the compounds that bond the stone granules and the medium [2]. These compounds moving from inside to the surface deposit with a percentage of accumulating dust, creating a highly adhesive and hard layer. In contrast, the back parts are weak due to the continuing loss of the binder. Thus, any scratch of the surface crust makes the sub-layers prone to continuous bleeding of granules. This means the loss of natural stone features due to the weak internal structure resulting from the poor bonding or disintegration of the granules of the surface layer. This process results from dissolving or rupturing the granules' binder because of stress caused by salt crystallization or ruffle of mud metals in the natural composition of the stone.

Deterioration or weakness manifestations also include crust that dislocates one layer after another due to the bumps of florescent salts or heat as a damage factor. Several methods have been utilized to handle this phenomenon [3].

2. Definition of the Phenomenon

A hard crust appears because of water movement via the pores and interacts with the binder, and the outcomes deposit on the surface and may mix with dust, dirt, and aerosols, forming the crust. Below the crust is a weak layer resulting from the continuous loss of the binder between the granules. This phenomenon is called stone bleeding [4], fig.(1).

Figure (1) Shows **a.** a gate of Amenhotep Temple (middle), **b.** a block in the entrance of the Obet Temple

2.1. *Reasons for occurrence*

Deterioration factors causing stone bleeding to weaken the internal structure and separate the binder of the stone, they include:

***)** Incoherent construction materials and different mineral structures of the stone cause different expansion and shrinkage ratios [5], weaken the bonding between granules, and decay the stone compounds with the help of deterioration factors [6], fig.(2-a).

***)** Salt crystallization causes the disintegration of granules, loss of the construction materials, cracks, and rupture [7]. Salts resulting from the use of black cement in the reinforcement operations, salts in the wall of the monument, and the interaction with the surrounding environment [8], including heat and humidity in the case of rains rich in salt, cause stone bleeding because of water evaporation and the concentration of the salt solution in the stone, causing salt crystals with increased size in the pores. Consequently, the internal pressure affects the stone components and the binding material. Droppings of birds [9] and animals loaded with salt motivate stone bleeding. Furthermore, groundwater salts play a significant role in stone deterioration, especially in the Karnak Temple [10]. Deterioration resulting from salt crystallization depends largely on the salt type, stone resistance, and pores. Physical weathering, including salt crystallization, mainly affects the sandstone's degradation, causing differences in the texture and petrophysical properties of the stone, which affect the stone and result in stone bleeding in the hard outer crust and the inner components [11]. Moreover, compressive strength, porosity weight loss, salt crystallization, mineral structure, and type of the binding material affect the sandstone [12]. Heat variations and pressures on the stone result, in addition to, the recrystallization of fine salt layers may cause shear stress, which can cause stone corrosion [13], fig. (2-b).

***)** Groundwater is an important factor in affecting stone bleeding because it causes size expansion and bristle of mud minerals [14], resulting in breaking the bonds between the granules and the decay of the stone components. It comes from many sources, such as agriculture, rains, and fluids [15], especially sewage rich in salts with differences in the rates of heat and humidity. Water rises due to the capillary system, with salts, including C, NO_3, SO_4, Mg, Ca, K, and Na, fig.(2-c).

*) Relative humidity dissolves salts in the stone and moves the salt solution to the surface, causing water evaporation and salt efflorescence, which, in turn, causes pressures, decay of the stone, and forming a hard crust because of the dissolution of the binder. The pores are filled with water, which is taken to the surface. Frequent evaporation causes a tight layer and the loss of the binder in the walls. Scratching this layer causes the bleeding of stone, fig. (2-d).

*) Temperature fluctuations cause changes from dry to wet conditions, causing severe damage, disintegrating granules [16], and stone bleeding. Moreover, the color of the mineral affects the deterioration operations because dark minerals absorb more heat than the light minerals, causing granular disintegration [17], fig. (2-e).

*) Improper restoration works [18], deteriorate the monument because of using improper materials and black cement, which have a harmful effect because of being saturated with saline water and reinforcement materials that have not penetrated the pores, deposited on the surface, and formed a layer. When this layer separates, it takes the patina layer and causes stone bleeding or completely disintegrates when they exposed to in witting followed by drying cycles [19], fig. (2-f). Some stones have high porosity, while others have low porosity. Sandstones with high porosity are affected by deterioration because porosity is one of the most important features affecting sandstones' deterioration [20].

Figure (2) Shows some occurring reasons of stone bleeding affected the study areas **a.** depth of preparation layer of incoherent construction materials, **b.** salt efflorescence in some doorsteps, **c.** groundwater in the wall of the port below the entrance of Karnak temples, **d.** humidity and its effect on the Taharqa temple, **f.** improper restoration using black cement in the southern part of Hall two of the Holy of Holies, Temple of Ramesses III, Karnak.

2.2. *Mechanism of occurrence*

The mechanism of this symptom is occurs when the surface layers are exposed to direct sunlight absorb and store high heat energy because the stones cannot do thermal conductivity [21]. Over hours a large part of the stored heat slowly leaks to the inner layers. At night, with the lack of a heat source, temperature drops, and the outer layers become colder because they lose heat quickly due to direct contact with the cold air. In other words, the surfaces' outer layer interaction with temperature fluctuations differs from the inner layers completely [22]. Successive and irregular movements occur in the surface layer of the walls exposed to the atmosphere and sun rays due to differences in the expansion and shrinkage coefficient of the mineral granules, causing the deterioration and disintegration of the binders. The walls' surface layers differ in thermal treatment, i.e., storage or loss of heat [23], from the layers below. They have successive movements, causing the deterioration of the binder between the granules and the following layers. As a result, the surface layers separate in the form of crusts, and the internal structure of the disintegrated granules erodes. Exposing the stone to frequent heating and cooling eventually causes stone deterioration. Expansion coefficients result from minerals in the stone. Therefore, sandstone has high expansion values because of quartz [24]. Based on diameter, the pores are affected as they play a significant role in the decomposition processes such as rock degradation [25]. When rain falls, it leaks into the stone through cracks or pores, where it crystallizes in the case of drought and disintegrates the surface layers due to crystallization pressure. Consequently, archaeological inscriptions and signs are lost [26]. Figure (3) shows that stone pores have different forms, which affect deterioration and water behavior [27] in the sandstone. Fragmentation of the sandstone into different sizes and the separation into fine layers parallel to mortar are notable deterioration patterns [28]. The hydrological behavior of the most sandstone types' was studied in detail. It was found that several stones contain clay, and a distinctive relationship can be established between the small pores and increased humidity. Many minerals found in building stones are thermodynamically unstable on the earth's surface. So, when having contact between the unsaturated minerals and natural water, e.g., rainwater, the building components dissolve, associated acidic water interact, and ions attack the mineral components, causing deterioration forms according to the following formulas:

- $2KAlSi_3O_8 + 2H^+ + 9H_2O \rightarrow 2K^+ + Al_2Si_2O_5(OH)_4 + 4Si(OH)_4$
- $Mg_5Al_2Si_3O10(OH)_8 + 10H^+ \rightarrow 5Mg^{2+} + Al_2Si_2O_5(OH)_4 + 4Si(OH)_4 + 5H_2O$
- $Fe_2SiO_4 + 1/2O_2 + 3H_2O \rightarrow 2FeOOH + Si(OH)_4$ [24].

The atmosphere is an important source of the disposition of salts in the building materials. Based on field observations, El-Gohary [29] demonstrated different forms of weather factors and related manifestations that affect the stone surface. A hard surface layer due to the effect of iron oxides and a hard crust (red, brown, and sometimes black) are formed. The surface crust is hard outside, covering crumbly and unsuitable layers known as stone bleeding. Rocks containing a large number of clay minerals that are more susceptible to decomposition, cracking, and disintegration when exposed to weathering cycles of hydration and drying [30].

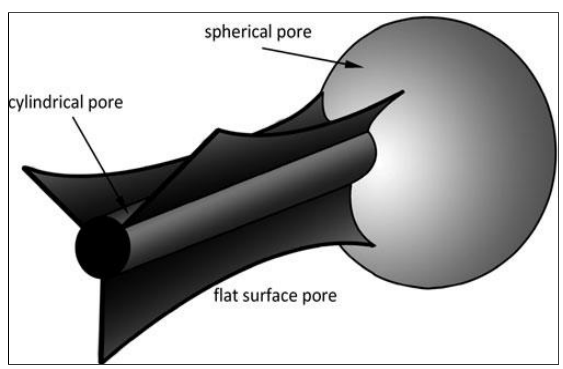


Figure (3) Shows the basic geometric shapes of the pores, especially in sandstone, (*After, Bernabe, 1991*) [31].

2.3. *Forms*

A hard crust is formed. Over time, a thick layer is formed, which deteriorates and deforms the aesthetic shape of the monument, making the stone below is very decayed, fragile, then, susceptible to disintegration decomposition processes. When exposed to scratching, continuous stone bleeding takes place in the layer in the form of disintegrated granules like sand powder, fig. (4).

Figure (4) Shows the development stages of the stone bleeding; **a.** crust created, **b.** granular disintegration, **c.** stone bleeding.

2.4. *Locations*

Stone bleeding was observed in several locations in the Karnak temples, mostly in the lower parts of the walls, because they are more prone to the effect of humidity or groundwater and salts (*the main causes of stone bleeding*). It also occurs near the inappropriate restoration works using very hard black cement, which contains a high percentage of salts, such as the gate of the *Ptah temple*, *Taharqa temple, the columns of compartment in the Khonsu temple,* and some parts of *the Obet temple walls*. In addition to other many places in *Karnak area*, fig. (5)

Figure (5) Shows some locations of the bleeding in the symptom in some parts of Karnak temples; **a.** *the entrance of Ptah temple*, **b.**, *Taharqa temple*, **c.** *eastern wall of the Obet temple*, **d.** the cornices of *Khonsu's temple hall*.

3. Investigation Methods

Different investigation types were used to evaluate the main cause of stone bleeding, and study the negative effects of the stone body on the collected samples. Scanning electron microscope (SEM) model *JEOL JSM-5400LV* attached with EDAX ZAF Quantification was used to examine the morphological features of the collected stone samples. Furthermore, it helped identify the chemical composition of the samples, especially the elements that resulted from weathering processes. *Analytical X, pert pro-PW 3040/60 X-ray diffraction* (XRD) analysis was adapted to identify the mineralogical components of the affected stone due to the bleeding process. Finally, *Perkin Elmer "AAS Analyst 400", Spectrophotometer Unico-1200"* technique was elevated for analyzing water samples to evaluate TDS and salts in water dominated in the study area.

4. Results

4.1. *SEM investigation*

SEM investigations of the samples showed that their morphology contains quartz and silica. It also illustrates the different forms of deterioration such as kaolinite particles as a main weathering products, in addition to halite and thenardite as salts that led to some cracks and micro-fissures that resulted as a direct effect of stone bleeding, fig. (6).

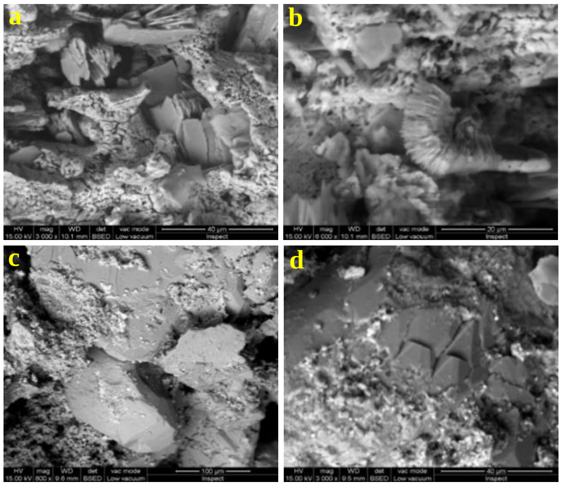
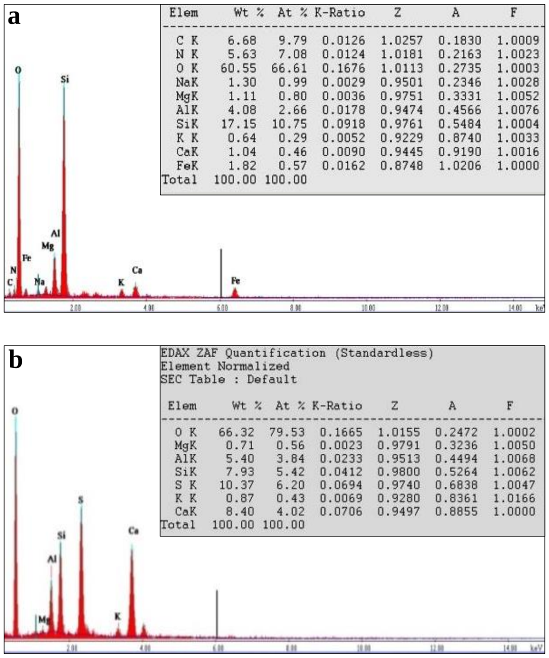


Figure (6) Shows SEM photomicrograph; **a.** deterioration forms and kaolinite (3000x), **b.** NaCl within the cracks (6000x), **c.** Na_2SO_4, granule disintegration, **d.** some accumulated particles of weathering products (3000x).

4.2. *EDX analysis*

EDX analytical results of *Khonsu temple* sample showed a high percentage of **O** (60.55%), followed by **Si** (17.15%), C (6.68%), **N** (5.63%), **Fe** (1.82%), **Na** (1.30%), **Mg** (1.11%), and **Ca** (1.04%). It also showed secondary elements, such as K, fig. (7-a). The sample related to the *open courtyard* showed a high percentage of **O** (66.32%), followed by **S** (10.37%), **Ca** (8.40%), **Si** (٧.٩٣%), and **Al** (5.40%). It also showed secondary elements, such as **K** and **Mg**. Finally it illustrated that silicon is the main component of sandstone, fig. (7-b).



Elem	Wt %	At %	K-Ratio	Z	A	F
C K	6.68	9.79	0.0126	1.0257	0.1830	1.0009
N K	5.63	7.08	0.0124	1.0181	0.2163	1.0023
O K	60.55	66.61	0.1676	1.0113	0.2735	1.0003
NaK	1.30	0.99	0.0029	0.9501	0.2346	1.0028
MgK	1.11	0.80	0.0036	0.9751	0.3331	1.0052
AlK	4.08	2.66	0.0178	0.9474	0.4566	1.0076
SiK	17.15	10.75	0.0918	0.9761	0.5484	1.0004
K K	0.64	0.29	0.0052	0.9229	0.8740	1.0033
CaK	1.04	0.46	0.0090	0.9445	0.9190	1.0016
FeK	1.82	0.57	0.0162	0.8748	1.0206	1.0000
Total	100.00	100.00				

EDAX ZAF Quantification (Standardless)
Element Normalized
SEC Table : Default

Elem	Wt %	At %	K-Ratio	Z	A	F
O K	66.32	79.53	0.1665	1.0155	0.2472	1.0002
MgK	0.71	0.56	0.0023	0.9791	0.3236	1.0050
AlK	5.40	3.84	0.0233	0.9513	0.4494	1.0068
SiK	7.93	5.42	0.0412	0.9800	0.5264	1.0062
S K	10.37	6.20	0.0694	0.9740	0.6838	1.0047
K K	0.87	0.43	0.0069	0.9280	0.8361	1.0166
CaK	8.40	4.02	0.0706	0.9497	0.8855	1.0000
Total	100.00	100.00				

Figure (7) Shows EDX analysis of some affected samples in the study area; **a.** Khonsu temple, **b.** eastern wall of the Obet temple

4.3. *XRD Analysis*

XRD results of the sample of the wall of Amenhotep II temple of showed that *quartz* is the main component, whereas *aphthitalite* and *thenardite* are impurities, fig. (8-a). Moreover, analytical results of the sample behind the great hypostyle hall illustrated that *quartz* as the main component, *microlite* as a secondary component, and *kaolinite* as an impurity, tab. (4) & fig. (8-b).

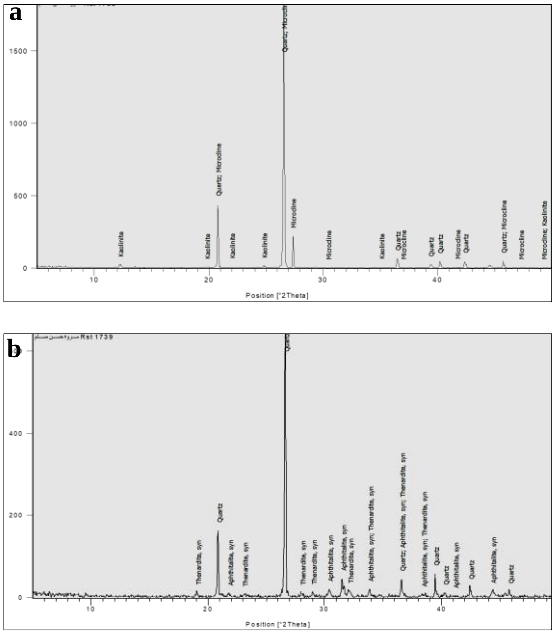


Figure (8) Shows the XRD pattern of samples; **a.** Amenhotep II Temple, **b.** great hypostyle hall.

4.4. *AAS results*

The results of analyzed samples taken from different points in the temples; **1)** open courtyard eastern wall, **2)** Amenhotep II temple and **3)** great hypostyle hall, all of these results are listed in tab. (1)

Table (1) AAS analytical results of the samples taken from some points in Karnak temples

Sample No.	Unit mg/L	Cations					Sum Cations	Anions			Sum Anions	TDS
		Fe	K^+	Na^+	Mg^{++}	Ca^{++}		Cl^-	SO_4^-	HCO_3^-		
1		3.157	1240	1970	31	166.6	**3410.8**	2545	510	354	**3409**	7170
2		0.065	2274	1140	45.26	18.1	**3477.4**	315	827	2347	**3489**	8830
3		0.654	6.551	22.4	11.24	34.01	**74.86**	18	28	34	**80**	167

5. Discussion

Groundwater affects sandstones, especially those with high porosity. The water content leaking into the stones is a dangerous factor that causes several forms of deterioration. This water moves to stones through the capillary feature with a high percentage of salts that form salt crusts rich in C, NO_3, SO_4, Na, Mg, Ca, and K on stones' surface or within the pores, causing internal stress, which, in turn, cause stone bleeding [12]. By evaluating the results of AAS, tab (1), it can be noted that the main components of salt type (Na^+ & Cl^-) were absent in XRD data due to the continuous leaching of salts by the effects of ambient groundwater or the partial evaporation of Na^+, and Cl^- emission [32]. The wall of Amenhotep II temple contains the highest level of K^+, Na^+ and TDS values due to the precipitation of *aphthitalite* and *thenardite* within the pores of the sandstone. Great stone damage and durability problems can occur especially with alternative wetting/drying cycles that caused the transformation from thenardite to mirabilite, and vice versa especially below 32°C [33,34]. Furthermore, halite is recognized in the open courtyard eastern wall which encountered the highest values of Na^+ and Cl^- in pore-water. Halite, gypsum sylvite, barium sulfate, and sodium nitrate are common desert salts that have a higher expansion potential than rock minerals. The volumetric expansions of sodium nitrate, halite, and potassium chloride are more than three times that of granite [35]. The crypto-efflorescence of these salts within the pores of the stone exposed surface can cause some aggressive deterioration symptoms like spalling and powdering [36] will be occurred after scratching the surface hard crusts. These hard crusts are composed of salts, dust, dirt and aerosols due to interactions with the binder. Temperature fluctuations expose the monument to wet and dry cycles cause different expansion and shrinkage ratios, which harm the stones and cause granule disintegration symptom known by "*sanding disintegration*" [37]. Moreover, the high change of temperature and the successive and irregular movements in the surface crust of the walls exposed to the atmosphere and sunrays cause differences in the coefficient of expansion and shrinkage of the mineral granules and collapsing of the binder between the granules and the following layers [27]. Within the same context, salt crystallization, as a physical weathering mechanism results mainly in the deterioration of sandstone and cause differences in its texture and petro-physical properties, which, in turn, causes disintegration of the stone due to pore sizes increment and stone bleeding with a hard crust from the outside and powder or sanding from the inside [38]. Based on the XRD analytical results, the non-affected sandstone in the study area composed essentially of pure Quartz (SiO_2). Generally, sandstone contained about 96% of quartz belongs to quartz arenite [39]. The wall of Amenhotep II temple contains aphthitalite and thenardite as the sandstone subflorescences impurities in the pore space of the sandstone. The non-hydrated sulfate of potassium and sodium were detected, where, aphthitalite ($(KNa)_2SO_4$), and thenardite (Na_2SO_4) showed variable compositions with the potassium oxide to sodium oxide ratio of three to one) [40]. Increase of the temperature and a decrease of the humidity and grades transform mirabilite Na_2SO_4 $10H_2O$ into thenardite [41]. The minor K-bearing and Na-bearing feldspars and clay minerals of the sandstone are insufficient to explain the variety of the salts determined. The sample from the great hypostyle hall contains microlite $(Na,Ca)_2Ta_2O_6(O,OH,F)$ as a secondary component and kaolinite ($Al_2Si_2H_4O_9$) as an impurity that represent traces of clay minerals associated within the sandstone upon alteration [42]. Fine salt layers recrystallization may cause shear stress, and consequently stone corrosion [13]. The source of these salts comes from the black cement used in the reinforcement [8], salts in the wall of the monument, and the groundwater. When sandstone is exposed to weathering especially through water-rock interaction process, iron concentration increases, and the concentrations of both silicon and aluminum decrease with the replacement of kaolin inside the stone with iron due to the hydrolysis process [12], causing corrosion and stone bleeding due to etching of Qz crystals and other minerals present [43]. Furthermore, the pores are affected as they play a significant role in the decomposition processes, such as rock degradation. Additionally, incoherent construction materials and different mineral structure of the stone cause different expansion and shrinkage ratios, weaken the bonding between granules and decay the stone components with the help of the dominated deterioration factors [6]. Studying the deterioration factors in the Karnak region proved that the deterioration of sandstone results from the interaction between the internal structure, the dominant weathering factors and strength as attested previously by Friolo, et al [44] in their case study. The deterioration of the internal structure of stones results essentially from the heterogeneous stone structure, salts and clay minerals, and evaporation of the salt solution and forming salt crystals that increase within the pores and cause internal stress, which may also cause bleeding process. Furthermore, the decay of sandstone does not happen in isolation, but it results from the interaction among several factors include, moisture [45], wind especially in very cold weather. [46] and predominantly anthropogenic environmental air pollutants, whose concentrations have changed dramatically during the last centuries [47] that cause several deterioration phenomena, including cracks, micro fissures, peeling, stress, granule disintegration and separation, that finally lead to stone bleeding process. This process that was assured through all analyses and investigations mentioned above where, SEM investigation of the sandstone surface illustrated severe corrosion features in cement layer that attributed to the effect of thenardite as a one of the more sever salts dominated in the study area, which will be transition to mirabilite as a hydrated film on the crystal surface [48]. In addition, erosion and disintegration of quartz grains, especially in their edges (*changed from angular to rounded grains*) due to the migration and dissolving of the binder. **Finally, it can be claimed that "Stone bleeding" defined as:** "*A symptom, which is initially forming beneath the stone hard crust through affecting synergistic deterioration agents that alter the aesthetic shape of the monument and etching the cement material over time. Below the hard crust a very fragile stone developed and susceptible to be disintegrated and decomposed. If the surface is scratched, uninterrupted stone bleeding occurred in the disintegrated grains like sand powder".*

6. Conclusion

The experiments in this study indicate that there are several deterioration factors affecting the sandstones in the Karnak area, including salts, moisture, temperature variation, groundwater, wind, and rain that cause the phenomena of damage, such as cracks, separations, peeling, powdering, pressures and stresses of the pores of the stone. All of these symptoms played a synergistic role. They finally led to the occurrence of the phenomenon known as stone bleeding due to the dissolving of the cement material and disaggregated Qz grains beneath the surface hard crust. It was also proven through examinations and analyses using the techniques mentioned above the presence of quartz as the main component of the stone in our cases. Furthermore, the presence of halite and thenardite as sources of salts dominated the study area, and kaolinite was a weathering product.

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