

CONSOLIDATION OF ARCHAEOLOGICAL BASALT STONE: A NEW EXPERIMENTAL PROTOCOL BY USING DIFFERENT DISPERSIONS FORMULATION

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Abstract

In this study, a new protocol of consolidation for basaltic stone artifacts was investigated. A considerable series of samples of deteriorated basaltic artifacts were collected from the Rujm el-kursi archaeological site in Jordan. Colloidal dispersion was prepared to act as a consolidant solution for the selected basalt artifacts. The phase behavior for a system composed of Water /Calcium hydroxide/ Isopropyl alcohol/ Tetraethyl orthosilicate (TEOS)/ Paraloid B72 was investigated. The resulting ternary phase diagram was used to prepare the optimum formulation for consolidant dispersion. A series of laboratory experiments were conducted to evaluate the efficiency of the prepared dispersion. Experiments included capillarity water absorption, penetration of water, water drop absorption, salts movement and accelerated artificial aging test. Treated samples were investigated using scanning electron microscope (SEM), X-ray diffraction (XRD), X-ray fluorescent (XRF) and polarizing microscope (PM). The results of laboratory experiments showed that the prepared colloidal dispersion expressed acceptable efficiency in strengthening the internal structure of stone. Obvious changes in the mineralogical composition of treated samples were not recorded. The treated basalt stone appeared to be water repellent; can decrease the penetration of water, which in turn led to the decrease of salts crystallization process inside basalt stone pores. The prepared dispersion appeared to be stable against accelerated aging factors and effective in decreasing the basalt stone decaying process.

Keywords: Basalt stone; Consolidant dispersion; Ternary phase diagram; Paraloid B72; SEM; XRF.

Introduction

In Jordan, consolidation treatments of archaeological stone have been performed for more than 20 years. However, very few studies of the employed products and methods have been done. Experts in the field of stone conservation have different opinions in regard to the effectiveness of consolidating treatments and the long-term effects thereof. These circumstances also apply to questions concerning the suitability of different consolidation products and treatments. Other controversial issues are the number of treatments needed to ensure a good result, and the risk of over-treating the stone [1, 2].

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Consolidation is the treatment of stone with a substance that restores the mechanical properties after they have been degraded by weathering. Stone consolidation can be a risky intervention, due to its irreversibility and the fact that it can cause harmful side effects of some stone types. Like ferruginous rock, sandstone and limestone, show an important color change after consolidation while shrinking of the consolidant during its polymerization is another important phenomenon, due to this shrinking, the consolidation of big pores is limited and the grain size of the materials seriously influences the efficiency of the treatment. The preparation of suitable consolidants will remain a challenge to many researchers applying them, observing and assessing their future effect. The consolidation process can prevent damage from salt crystallization, or probably it can make the surface of the artifact water-repellent [3-8].

According to *G. Ziegenbalg and E. Piaszczyński* [9] silicic acid esters (SAE), especially products based on tetra ethylorthosilicate (TEOS), are one of the most commonly applied consolidants for stone, mortar and plaster. The action of SAE bases on hydrolysis and condensation reactions occurring in the presence of humidity. Amorphous, gel like silicic acid is formed. The reactions are normally slow. The success of stone consolidation with SAE depends on the characteristics of the material to be treated as well as the climatic conditions (temperature, humidity, wind, etc.) during the application. Mineralogical composition, salt content as well the dimensions of the cracks, fissures or delamination that have to be stabilized are of great importance, too [10, 11]. Recently the sol-gel materials as a “stone consolidant” have been found to be successful in applications for the conservation and restoration of stone. However, a well-known drawback of these materials is their tendency to crack during drying inside the pores of the treated stone [2, 12]. So the main purpose of this study is to analyze physical property changes deteriorated basalt stone, after consolidation treatment with a new protocol of different dispersions formulation with the guidance of the four prepared ternary phase diagram of the system of $H_2O/Ca(OH)_2$ /isopropyl alcohol/TEOS.

Experimental

Basalt stone samples

A considerable collection of basalt stone blocks and artifacts were collected from Khirbet el-Kursi archeological site in Jordan (Fig. 1). According to archaeologists, this site is dated back to Byzantine period (4th to 7th A.D) [13]. For the physical testing, the samples were shaped as regular cubic of 5x5cm. then cleaned from soil deposits. The cleaning process was performed with distilled water and toothbrush. To ensure efficient cleaning, the samples were soaked in a water bath sonicator at 25°C for 5 minutes then dried using oven at 70°C for 24 hours.



Fig. 1. The basalt stone artifacts from Khirbet el-Kursi archaeological site

Materials

The following chemicals and materials were used without further purification: Calcium Hydroxide (SD fine chem. Limited SDFCL), tetraethylorthosilicate 98% assay (Sigma Aldrich), isopropyl alcohol 100% assay (VWR), sodium sulfate decahydrate (Panreac Quimica

SD), and Paraloid B72 (Pentraeth, Industrial Park, UK).

Ternary phase diagram

Nine samples for each of Ca(OH)₂/isopropyl alcohol, Ca(OH)₂/TEOS and TEOS/isopropyl alcohol combination were prepared for each ternary phase diagram in the range of (10-90)% of Ca(OH)₂ in isopropyl alcohol by mass. These masses were achieved by weighing a desired amount of Ca(OH)₂ and then adding isopropyl alcohol to get a final mass for each sample. Titration technique was used in this approach by adding water, drop by drop, to increase percentage mass of water across the phase diagram toward the water corner. After each increment, the samples were vortexed for 1.0min, followed by normal centrifugation for 15min at 2500turn/min. Visual observation was used for all samples after each addition to identify number of phases present regularly. The phase diagram for Ca(OH)₂/isopropyl alcohol/TEOS, was constructed by titration of Ca(OH)₂/isopropyl alcohol combination with TEOS.

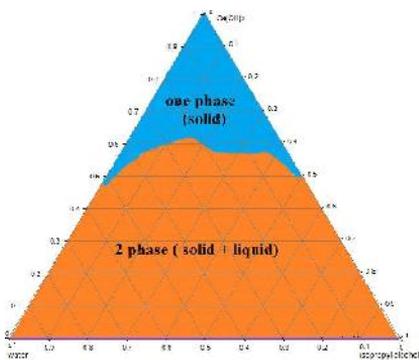


Fig. 2. Ternary phase diagram of water/Ca(OH)₂/isopropyl alcohol system

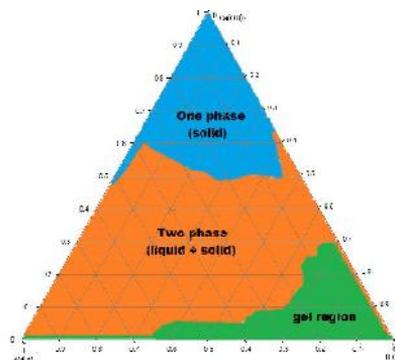


Figure 3. Ternary phase diagram of water/Ca(OH)₂/TEOS System 13

Dispersion solutions

Dispersion solutions were prepared with the guidance of the constructed ternary phase diagrams of H₂O/Ca(OH)₂/isopropyl alcohol/ TEOS and 3% of Paraloid B72. However, deep brushing and impregnation were the suitable methods used for application of the prepared solutions.

Instrumentation

The following instruments were used for the characterization of the basalt stone before and after consolidation treatment: Scanning electron microscopy (SEM) Shimadzu Corporation Superscan SSX-550 SEM-EDX (Tokyo, Japan), X-ray diffraction spectroscopy (XRD) 6000 Shimadzu for powdery samples with Cu_K radiation of 1.543 Å (Tokyo, Japan), X-ray fluorescence spectrometer (XRF) Shimadzu 1800 for powdery samples with Ultra-fast Scanning of 300°/min (Tokyo, Japan), polarizing microscopy MEIJI with(10X).

Methods of analysis and investigation

To characterize the chemical composition of the basalt stone samples and evaluate the effectiveness of the consolidation process and efficiency of dispersions used various analytical and examination techniques were used. SEM was used to examine the morphological texture of basalt stone samples. XRD was used to determine the mineralogical composition of stone material based on studying the crystalline structure of the materials [14, 15]. The elemental composition of the stone samples was determined by XRF. Petro-graphic examination was performed using polarizing microscopy to identify the rock-forming minerals of a stone and its texture; also to quantify its mineralogical composition; all this information allows a preliminary prediction of the physical behavior of the stone as well as its potential decay processes; it is also essential for establishing its petro-graphic classification.

Laboratory tests

Physical and mechanical tests such as penetration of water, capillary of water absorption, salt movement and compressive strength test, for the treated and untreated samples were performed according to international scientific standard to measure the ratio of physical change of the treated samples in comparison with their original state before treatment [16]. Furthermore, accelerated artificial aging test was carried out to investigate the stability of treated samples against the effect of different weathering factors such as relative humidity, temperature and radiation.

Results and discussion

Phase diagram determination

The phase diagram of Water/ $\text{Ca}(\text{OH})_2$ /isopropyl alcohol behavior system (Fig. 2) shows that $\text{Ca}(\text{OH})_2$ and isopropyl alcohol are not soluble in each other. However, the solubility of isopropyl alcohol in $\text{Ca}(\text{OH})_2$ reached to 53% (by mass). Water and isopropyl alcohol were totally miscible in each other (one phase). While $\text{Ca}(\text{OH})_2$ solubility in water was insignificant, the solubility of water in $\text{Ca}(\text{OH})_2$ reached to 48%. For $\text{H}_2\text{O}/\text{Ca}(\text{OH})_2/\text{TEOS}$ system (Fig. 3), $\text{Ca}(\text{OH})_2$ solubility in water was insignificant, but the solubility of water in $\text{Ca}(\text{OH})_2$ reached 48% (by mass). TEOS and water were totally soluble in each other, a gel region was determined along $\text{H}_2\text{O}/\text{TEOS}$ edge. Two phase (solid + liquid) area was formed when titrating with water. The phase diagram of $\text{Ca}(\text{OH})_2$ /isopropyl alcohol/TEOS system (Fig. 4), show that TEOS and isopropyl alcohol are totally miscible in each other. Two phase (solid and liquid) region was formed for about 70% (by mass) between isopropyl alcohol and $\text{Ca}(\text{OH})_2$ and between TEOS and $\text{Ca}(\text{OH})_2$. One phase solid was determined from 70% per mass and above of $\text{Ca}(\text{OH})_2$ from each edges with TEOS and isopropyl alcohol. $\text{H}_2\text{O}/\text{TEOS}/\text{isopropyl alcohol}$ system (Fig.5.) showed that along isopropyl alcohol and TEOS line, a one phase area was determined and they are totally miscible in each other, while titrating with water the two phase (liquid + liquid) was formed then the gel region was determined at 47% (by mass). Taking into consideration the behavior of the four constructed ternary phase diagrams, selected areas were tested to be used in preparing suitable dispersion for consolidation treatment which should be far away from the gel region. The percentages used in this treatment for consolidation was $\text{H}_2\text{O}/\text{Ca}(\text{OH})_2/\text{TEOS}/\text{isopropyl alcohol}$ 9%, 14%, 6%, and 61% respectively.

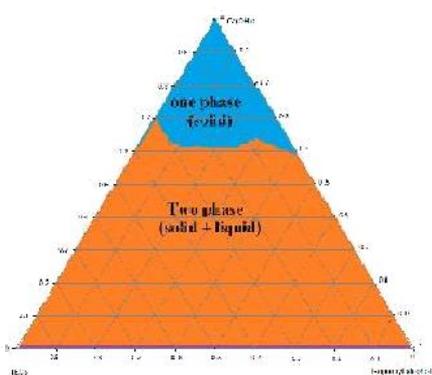


Fig. 4. Ternary phase diagram of $\text{TEOS}/\text{Ca}(\text{OH})_2/\text{isopropyl alcohol}$ system

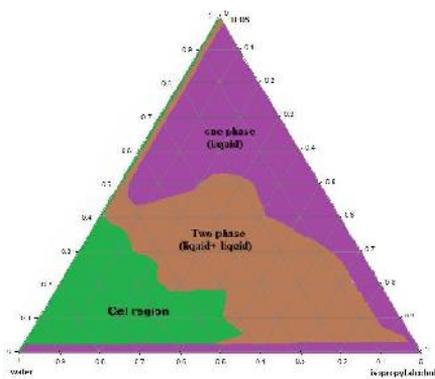


Fig. 5. Ternary phase diagram of water $/\text{TEOS}/\text{isopropyl alcohol}$ system

Chemical composition determination

Polarizing microscope identification

A polarizing microscope investigation was carried out to accurately identify the chemical composition of basalt stone samples [17, 18]. The samples were prepared as perpendicular thin sections to be examined. Figures 6 and 7 show the mineralogical composition obtained before and after consolidation treatment. Obvious mineralogical changes after the consolidation treatment were not observed, where, plagioclase and quartz, are the basic minerals in the basalt stone, which proved the efficiency of treatment. The photo of polarizing microscope was taken by 13 Megapixels Samsung Camera Resolution.

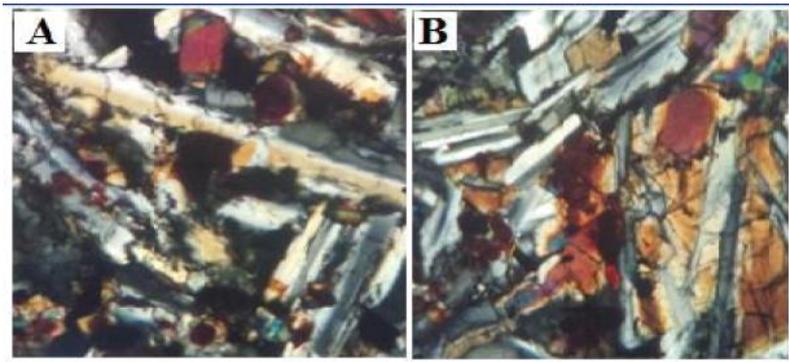


Fig. 6. Polarizing microscope images show the mineralogical composition of basalt stone before consolidation: A - sample 1, B - Sample

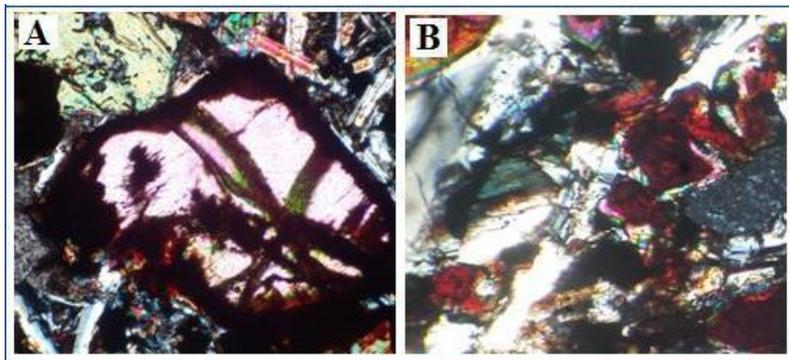


Fig. 7. Polarizing microscope images show the mineralogical composition of basalt stone after consolidation: A - sample 1, B - Sample

Scanning electron microscopy observation

Addition to the visual examination of stone objects, small samples of stones were investigated by scanning electron microscopy (SEM), which was operated in a secondary electron mode to examine the surface morphology and body structure of the stone material and consolidant material [19]. It can be observed that the particles are irregularly shaped and varied in sizes. Whereas other aspects of fractured surface and highly fissured nature of decayed surfaces were observed (Fig. 8) Furthermore, SEM examination was carried out on cross-sections of the same stone samples to examine the structure morphology of inner core of every sample. SEM images showed how the inner body differs from the outer surfaces especially the grain size and surface texture. The scanning electron micrographs reveal that the particles were

heterogeneously shaped and less-vitrified with a lot of incisions. Besides, there is a slightly evident structural and compositional continuity between the surface and the bulk. Whereas the micrographs show the masses are more compact, nearly heterogeneous and fairly vitrified. However, only slight dissimilarities in color and texture were observed during examining all the stone sections. However SEM images of the treated samples with a dispersion of $H_2O/Ca(OH)_2/TEOS/isopropyl\ alcohol$ (14%, 9%, 6%, and 61%) reveal that the formed film is very thin and homogeneous. Moreover, it obviously spreads to cover the entire stone surface without altering stone color or texture. The most important advantage of the formed film is that it is glossy without luster appearance.

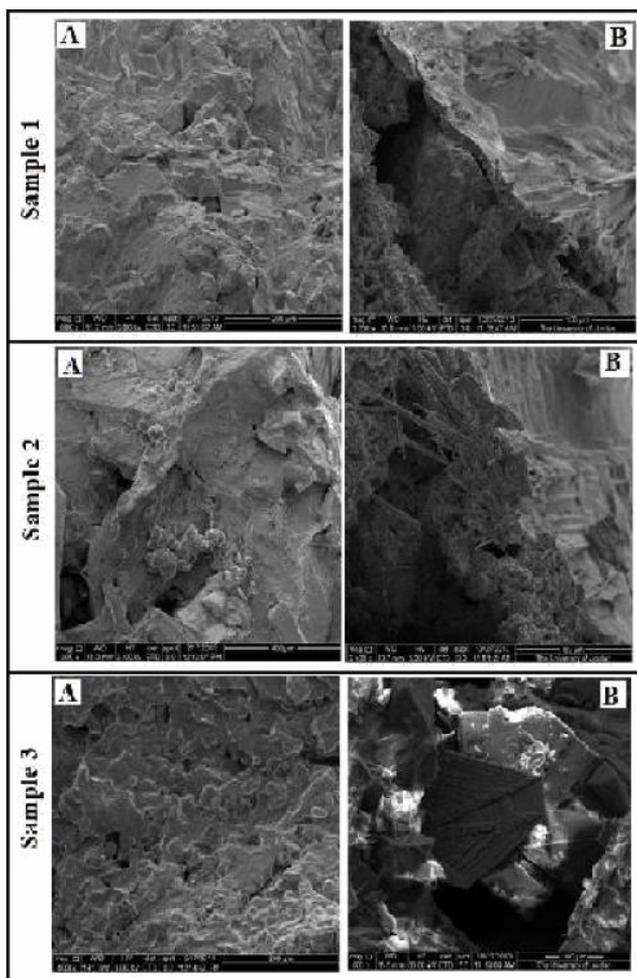


Fig. 8. SEM images of basalt stone samples (A) before and (B) after consolidation treatment

XRF spectroscopy results

Table 1 shows the chemical composition of the basalt stone samples before and after treatment. It was observed that silica (SiO_2) content increased after treatment due to TEOS presence.

Table 1. The chemical composition of the basalt stone samples before and after consolidation treatment obtained by XRF

Oxides %	Before consolidation			After Consolidation		
	Sample 1	Sample 2	Sample 3	Sample 1	Sample 2	Sample 3
Fe ₂ O ₃	10.72	12.79	12.89	12.59	12.18	10.90
TiO ₂	1.90	2.26	2.28	1.73	3.55	2.03
CaO	10.69	9.81	9.41	9.26	10.36	10.53
K ₂ O	1.02	0.78	0.82	0.78	0.51	0.77
P ₂ O ₅	0.43	0.31	0.32	0.10	0.09	0.25
SiO ₂	44.43	44.78	44.71	46.46	44.16	46.78
Al ₂ O ₃	14.02	14.18	14.26	14.18	13.97	14.50
MgO	5.62	6.24	6.26	6.97	5.72	5.23

XRD spectroscopy results

XRD patterns (Fig. 9) show that the samples are mainly composed of plagioclase [(Na,Ca)(Si,Al)₄O₈], pyroxene [CaAl₂SiO₆], olivine [Mg₂SiO₄], in addition to some trace phases resulting from some effect of physical and chemical weathering. The identifications reveal that the major minerals of basaltic stone samples are Augite [(Ca,Na)(Mg,Fe,Al)(Si,Al)₂O₆] and Anorthite (CaAl₂SiO₈) (belong to the plagioclase feldspar group, an isomorphs solid solution series composed of calcium aluminum silicate). May contain some sodium replacing the calcium, but that amount must be less than 10 percent to be strictly anorthite. XRD results indicated that Augite was the new mineral formed after treatment. Augite appeared because of the consolidation treatment which contain Ca(OH)₂ salt, although it shows that no changes on the mineralogical compositions of the stone.

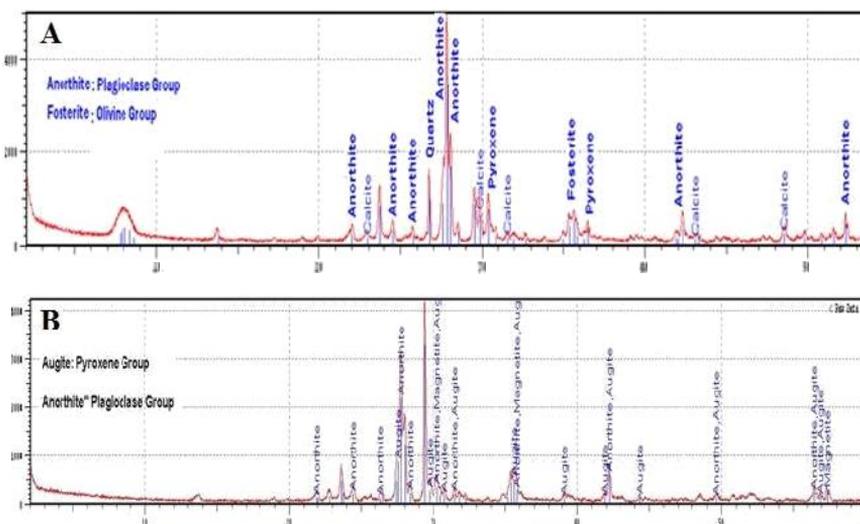


Fig. 9. XRD patterns of basalt stone artifact; (A) before and (B) after consolidation treatment.

Physical properties determination

Capillary water absorption

The amount of absorbed water M_i , at time t_i per surface unit (0.001) m^2 (S), is defined as follows:

$$Q = (M_i - M_0) / S \tag{1}$$

The Q values are plotted against the square root of time ($t^{1/2}$) according to the general adopted equation $Q = K \cdot t^{1/2}$. As shown in Figure 10, the mass of basalt stone artifact samples before and after consolidation treatment taken at different time intervals, which show the efficiency of treatment of decreasing the quantity of water absorbed to the stone by capillary forces. The water absorption at different time intervals for the three samples before and after the consolidation treatment are present in Figures 10 and 11 which show that the saturated coefficient of the stone is decreased for the treated sample and it also determined the maximum amount of TEOS the sample can absorb by capillarity which is helpful in determining the amount of consolidant needed for the entire stone. According to the linear relationship between saturation coefficient Q and $t^{1/2}$, the slope of this relationship show the efficiency of the consolidation treatment (K) which applied in the three basalt stone artifact samples. The efficiency as calculated is high because (as shown in Table 2) the percentage of water absorption by capillarity forces after consolidation treatment decreased [1].

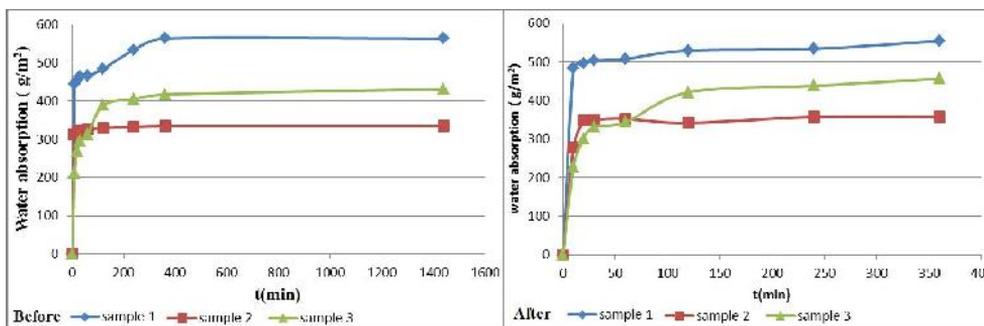


Fig. 10. Graphic representation of capillary water absorption for three basaltic samples before and after the consolidation treatment as a function of time (min)

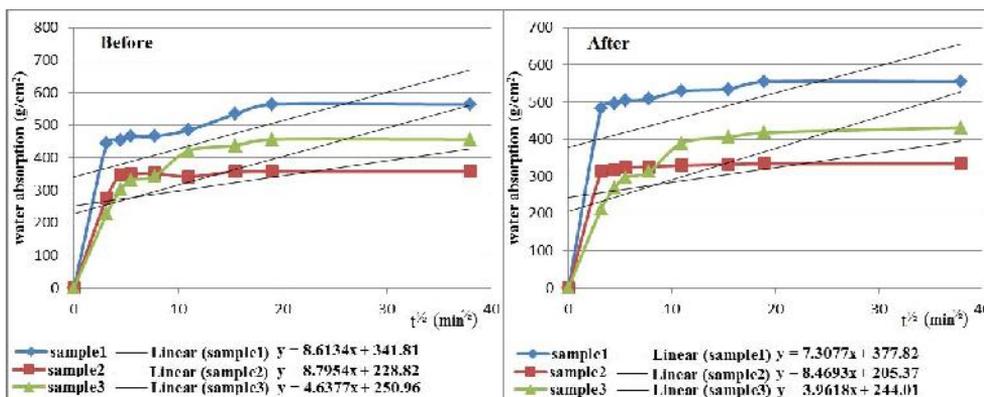


Fig. 11. Graphic representation of capillary water absorption (Q) for three basaltic samples before and after consolidation treatment, as a function of the square root of time ($min^{1/2}$)

Table 2. The comparison between efficiency of treatment before and after consolidation treatment

Sample no.	K before	K after
1	8.61	7.31
2	4.64	3.96
3	8.75	8.47

Penetration of water test

The results of penetration of water test show the treatment efficacy for the basalt stone artifact samples is applied and the rate of penetration is decrease. This proved that repellency of water to the stone is lowered as shown in Figures 12 which compare the rate of penetration of water between three different basalt stone artifact samples, after and before the consolidation treatment. As shown in Figure 12 and after calculating the slope of the linear relationship (Table 3), where slope represents the rate of penetration, so finding that it decreased for each sample after treatment suggests the success of consolidation process from the penetration point of view.

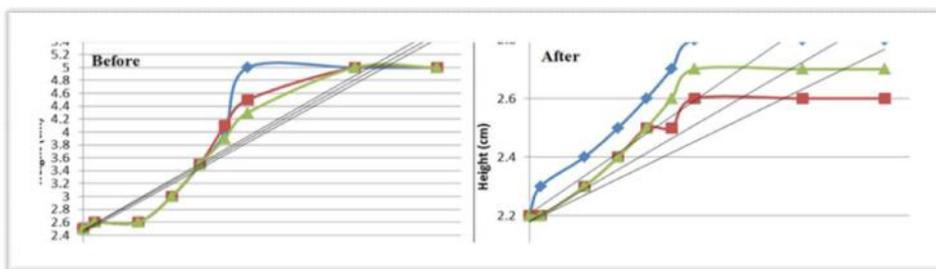


Fig. 12. Height vs $t^{1/2}$ for penetration test of three samples before and after consolidation treatment and the rate of penetration of water

Table 3. The rate of penetration of water before and after consolidation treatment

	Rate of penetration of water		
	Sample 1	Sample 2	Sample 3
Before consolidation treatment	0.42	0.41	0.40
After consolidation treatment	0.11	0.07	0.09

Water drop absorption test

The time taken for the total absorption of water for: Treated sample is (tx). Untreated sample is (tn). Whereas determining the evaporation time (te) for water by dropping 1.0mL of water in a glass trough [20]. The following equation is useful to calculate the *water drop absorption* (WA) as in Tables 4 and 5:

$$WA\% = [1 - (tx - tn)/(te - tn)(te/tx)]100 \tag{2}$$

Water repellency may be expressed by using the following equation:

$$WR(\%) = 100 - WA.$$

The result of the test shows that the treated samples exhibit longer absorption times compared with untreated samples; this indicates a reduction of stone porosity by the consolidation treatment as shown in Tables 4 and 5.

Table 4. The needed time for the water drop absorption and evaporation for the treated and untreated samples

Sample no.	tn (min)	tx (min)	te (min)
1	25.35	25.28	20.15
2	25.46	25.40	20.15
3	25.24	25.22	20.15

Table 5. The water drop absorption percentage for treated and untreated samples

Sample no.	WA%	WR%
1	98.93	1.07
2	99.10	0.90
3	99.69	0.31

Movement of salts test

The movement of salt test was made for the soluble salts in water, so the crystallization process of salt inside the pores of basalt stone artifact result from the evaporation process of water, the mass of absorbed salt will be calculated form the following equation: *Mass of salts absorbed* = ($M_i - M_0$). Figure 13 shows the salt movement test for treated and untreated samples, the comparison between treated and untreated samples proved that the quantity of salt crystallization inside the pores after consolidation treatment is less than before consolidation treatment [20-22].

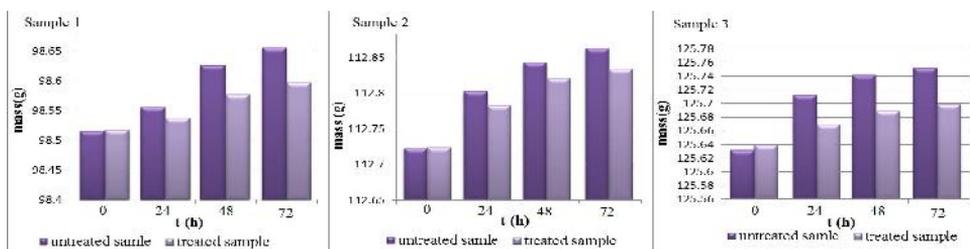


Fig. 13. Mass vs. time for the movement of salts test before and after consolidation treatment

Accelerated aging test results

After 120 hour of a constant exposure to 110% relative humidity, 105°C and 25nm UV light, the treated basalt stone samples appeared to be relatively stable since no obvious colorimetric or morphological changes were optically observed on the stone surface. The microscopic investigation carried out by optical microscope confirmed these observations also except a slight darkness of the resulted film is observed.

Compressive strength resistance

Table 6 records the values of compressive strength of three stone samples (cubic 5X5 cm) before and after consolidation treatment. However the resistance to compressive strength of the treated samples relatively increased which reflect the effectiveness of the consolidant used [1, 23, 24] .

Table 6. Compressive strength values of stone samples before and after consolidation treatment.

	Compressive strength value(Kg/cm ³)		
	Sample 1	Sample 2	Sample 3
Before consolidation treatment	231.4	233.02	229.2
After consolidation treatment	231.6	233.08	229.5

Conclusions

The consolidation treatment of the basalt stone samples collected from Khirbet el-Kursi archaeological site was successfully evaluated, in order to select the appropriate material for conservation of historical basalt stone. A series of basalt stone artifacts and blocks were selected and treated with the dispersion solution which was prepared with the guidance of

ternary phase diagram $H_2O/Ca(OH)_2/TEOS/isopropyl\ alcohol$. Similar types of dispersions were previously applied on different groups of limestone and sandstone, and have acceptable result in consolidation of decayed stones. A 3% of Paraloid B72 dissolved in acetone was successfully used as a fixed material of the dispersion on the stone surface. The capillarity absorption test indicates that the treatment decreased the ratio of water absorbed inside the stone which can decrease the ratio of salt crystallization inside the pores. Penetration of water and water drop absorption tests also show a positive result of decreasing the ratio of water penetration appearance inside the pore of basalt stone artifact.

The physical examinations showed the treatment does not obviously affect the stone surface and the stone surface was homogeneously coated by the dispersion. The chemical analysis showed the chemical and mineralogical composition of stone was as it is before the treatment without any obvious changing. Furthermore the treated samples appeared to be relatively stable against accelerated aging test. The overall conclusion is that the consolidation treatment was successfully applied without any physical or chemical alteration of the basalt stone artifact. The formed consolidation film effectively acts also as a protective coating of stone surface.

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