

# Assessment of the Resistance of Dolomite Stone from Crac des Chevaliers Against Salt Crystallization

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Received: 28 November 2022, Accepted: 09 March 2023, Published online: 31 March 2023

## Abstract

This study was dedicated to evaluating the durability of the carbonatic stones of Crac des Chevaliers castle against salt crystallization. Crac des Chevaliers castle is a UNESCO world heritage site located in Syria and is considered one of the most important crusader castles built in the 12<sup>th</sup> century. Salt efflorescence is a common decay type detected on the stone blocks of this building, so six dolomite stone lithotypes with various structures tested under salt aging in three different salt solutions; sodium chloride, magnesium sulfate, and sodium sulfate. Among those lithotypes were two microbioclastic wackstone and four microcrystalline dolomite. The microbioclastic wackstone had low open porosity (18 and 11 v/v%) compared to the microcrystalline dolomites of about 28v/v% open porosity. Two of the microcrystalline dolomite lithotypes had bimodal pore networks and around 4 v/v% closed pores, however, the other two exhibited very small closed porosity (around 0.35 v/v%). The results revealed that even though the great effect of the open porosity on the durability of the specimens, the closed porosity also had a major role and the weathering form depended on the salt composition and the stone porosity properties. The primary decay type of the specimens in sodium sulfate solution was granular disintegration while pitted surfaces formed in sodium chloride solution, and the exfoliation of the stone surface into thin layers was observable after the aging with magnesium sulfate solution. The effect of NaCl crystallization on the specimen was not as aggressive as that of the sulfates.

## Keywords

dolomite, salt weathering, sodium sulfate, magnesium sulfate, sodium chloride, closed and open porosity

## 1 Introduction

Salt crystallization has always been reported as a typical stone deterioration case in many historical buildings [1–3]. Salts can be present inside the stone structure, coming from both external and internal sources. The external sources include the salts brought from the marine environment [4], deicing salts [5], sea spray effect [6], or rising ground damp [7]. The internal sources mean the salts result from the interaction between the stone components and inorganic materials, such as air pollution [8] or the use of some inappropriate conservation treatments (like cement mortar or cement plaster) that contribute to salt contaminations (calcium sulfates and sodium salts) [9]. The most common salts that are frequently found in building stone blocks are NaCl, KCl, CaCO<sub>3</sub>, MgCO<sub>3</sub>, KNO<sub>3</sub>, and NaNO<sub>3</sub> (all are simple salts); as well as Na<sub>2</sub>SO<sub>4</sub> and MgSO<sub>4</sub> (can have hydrated forms) [10].

Salts exercise physical stresses derived from two types of pressures: crystallization and hydration pressure (in the case of hydrated salts). The growing of salt crystals inside a pore can exert stress on the pore wall [11]. The damage induced by salt crystallization depends on the crystal shape, supersaturation degree, pore size distribution, and the presence of salt mixtures [12–14]. La Russa et al. [15] emphasized that the heterogeneity of pore size distribution generates several different stressed areas within the stone structure, which leads to more decay. The amount of the absorbed salt solution by the pore system is controlled by the open porosity [16–17] and viscosity of the salt solution [18]. Tiny pores affect the durability of the stone blocks, where the smaller pores will experience a higher crystallization pressure than large ones [19].

In addition to crystallization pressure, the molecular volume of hydrated salts increases by binding of water molecules, and therefore, the volume expansion causes a "hydration pressure" [20]. The most deteriorative common hydrated salts for stone blocks are  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  and  $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$  [21]. Due to the simultaneous appearance of the crystallization pressure and the hydration pressure, the hydrated salts are more deteriorative than simple salts [22]. However, Rodriguez-Navarro et al. [23] do not agree with that argument because they found that environmental conditions (relative humidity and temperature) define which pressure appears and induces damage to the stone. Many different experiments were completed to investigate the effect of salt crystallization on the durability of carbonate stones under various conditions, such as total immersion [24–26] or capillary absorption of salt solution [27].

Salt efflorescence is a common issue in Crac des Chevaliers castle [28], see Fig. 1. The field investigation revealed that salt crystallization at the castle could be caused by many sources such as rising damp, sea spray effect, or usage of inappropriate mortar [29]. Therefore, it is essential to evaluate the resistance of stone blocks against the crystallization aging test of different salts.

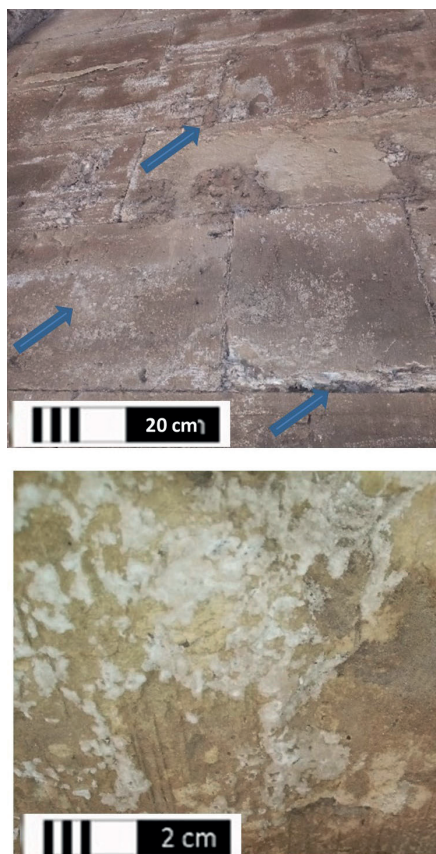


Fig. 1 Salt efflorescence on the castle walls

Crac des Chevaliers is one of the most important crusader castles in the Middle East and is the most distinctive and well-preserved medieval fortress in the world. This incredible historical structure was built during the medieval period in the 11<sup>th</sup> century and was later expanded with additional structures in the 13<sup>th</sup> century. These two building eras mirror the mixture of Gothic and Islamic architectures, so buildings with various functions: A Romanesque chapel, numerous residences and military facilities, various motifs, decorations, and inscriptions carved on the stones can be found in the castle. The castle is located in the northwest of Syria, about 35 km from the coast, as shown by Fig. 2. It is situated on a hill that rises around 660 m above sea level. The castle was built from dolomite, limestone, and basalt on a basaltic and clay foundation. The geological settings of the castle location attributed to different stone types that are recognized in the castle. The Neogene basalt is dominated in the eastern region that surrounds the castle [30], and it extends under the Cenomanian-Turonian (late cretaceous) to the west [31]. Cenomanian-Turonian mainly consists of well-bedded limestone, marls with chert nodules, and massive dolomitized limestone [32].

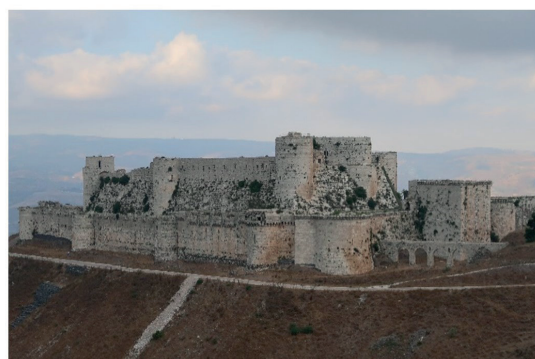


Fig. 2 The castle and its location

According to the classification of Köppen, the castle region has a dry Mediterranean, hot summer climate (CSA) [33]. Summers are warm to hot and dry (with monthly precipitation of less than 30 mm), while winters are mild-wet (with monthly precipitation of 160 mm and a monthly average temperature of 11°C) [34]. However, the temperature may drop to the freezing point in winter, so each winter day has at most one freeze-thaw cycle [28].

## 2 Materials

Six dolomite cylindrical stone cores with different textures were drilled from the castle walls (lithotypes D1-D6). Each core (5 cm in diameter) was divided into three specimens (each specimen to be examined under one salt attack).

D1 is a well-sorted micro bioclastic wackstone. It is slightly dolomitized, silicified, and the grain size is very fine. It contains limonitic spots, brownish micrite mottles, shell fragments and the pores are filled with calcite as shown by Fig. 3. The size of open pores ranges from 0.05 mm to 0.45 mm. D2 is a microcrystalline dolomite stone with many intercrystalline pores and a few large pores of 0.1 mm as shown by Fig. 3. D3 is a microcrystalline dolomite stone with ghosts of the original texture and crystals with clear rims and cloudy cores in their structure. It has a bimodal pore network, where the size of large pores ranges between 0.3–0.5 mm, and the micropores are about 0.02 mm as shown by Fig. 3. D4 is a highly recrystallized, homogenous, microcrystalline dolomite with about 0.005 mm crystal size. The pore size is around 0.2 mm, and it contains a lot of very fine pores as shown by Fig. 3. D5 is laminated dolomite with peloids and small intraclasts. It contains micritic and microsparitic layers with around 0.01–0.002 mm grain size. On the pore walls are observable sparitic crystals. The stone has a bimodal pore size; the large pores are about 0.3 mm, and the tiny ones range between 0.02–0.05 mm as shown by Fig. 3. D6 is a micro bioclastic wackstone with algal mat and fenestral pores. His bimodal pore distribution has two peaks in size of 0.4–0.6 mm and about 0.01 mm. The algal mat contains clast fenestral closed pores filled with sparitic calcite as shown by Fig. 3.

The chemical composition of the specimens was almost the same, consisting of about 30–40 w/w%  $\text{MgCO}_3$  and 60–70 w/w%  $\text{CaCO}_3$  (according to the differential thermal analysis /DTA), see Table 1. The physical properties of the specimens were different, see Table 1. D1 and D6 had the highest bulk density (2.2 and 2.4  $\text{g/cm}^3$ , respectively) compared to other specimens; and the smallest open porosity (18 and 11 v/v%, respectively). Even though D2, D3, D4,

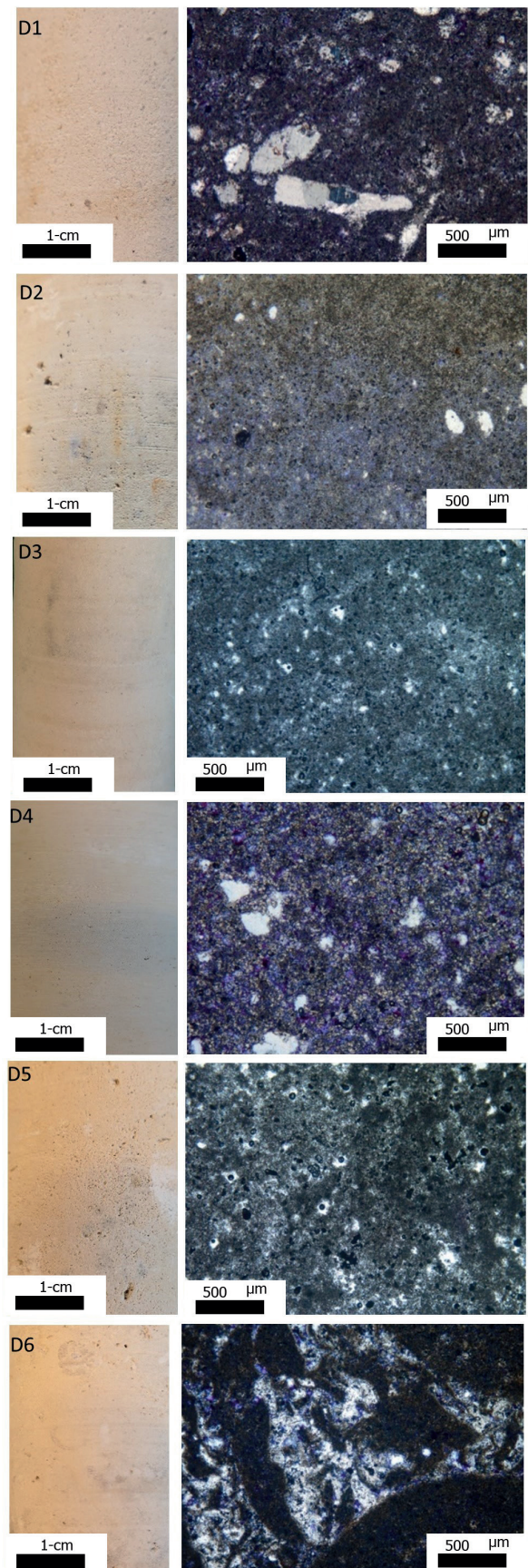


Fig. 3 Thin section photos of the tested specimens

**Table 1** Physical and chemical properties of the dolomite stone specimens

Specimen	Physical properties						Chemical properties	
	Bulk density g/cm <sup>3</sup>	Matrix density g/cm <sup>3</sup>	Total porosity % (v/v)	Open porosity % (v/v)	Capillary open porosity % (v/v)	Closed porosity % (v/v)	MgCO <sub>3</sub> % (m/m)	CaCO <sub>3</sub> % (m/m)
D1	2.204	2.751	19.9	18	13.8	1.9	29.50	67.89
D2	1.933	2.841	32.0	31.7	26.5	0.3	39.17	55.55
D3	1.943	2.879	32.5	28.1	20.7	4.5	39.75	56.77
D4	1.949	2.757	29.3	28.9	22.8	0.4	40.05	54.50
D5	1.902	2.834	32.9	28.8	22	4.1	38.53	57.82
D6	2.431	2.814	13.6	11.2	7.6	2.4	35.93	52.70

and D5 had approximately the same bulk density (around 1.9 g/cm<sup>3</sup>) and open porosity of about 30 v/v%; their closed porosity varies, see Table 1. D2 and D4 had the lowest closed porosity (around 0.3 v/v%); however, D3 and D5 had the highest closed porosity (about 4 v/v%).

Three salts were selected for the aging test: sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>), magnesium sulfate (MgSO<sub>4</sub>), and sodium chloride (NaCl). The choosing of salts for the experiment was based on the salts present in the castle. That is achieved by making a salt content test of stone crusts that were collected from the site. The results of salt content revealed that the major ions in those crusts were the chloride and sulfate.

As the castle is situated nearby the coast and is affected by the salt spray effect [35], it was essential to study the stones under NaCl solution. Sulfate salts were used, because Na<sub>2</sub>SO<sub>4</sub> and MgSO<sub>4</sub> are extremely damaging [26–27], and Na<sub>2</sub>SO<sub>4</sub> is considered the most aggressive salt used to assess the durability of the stone in the laboratory [36].

There are two crystallization phases of sodium sulfate Na<sub>2</sub>SO<sub>4</sub> (Thenardite) and Na<sub>2</sub>SO<sub>4</sub>·10H<sub>2</sub>O (Mirabilite). Sodium sulfate usually precipitates from a solution below 32.4°C and at high relative humidity (above 60%) in the form of Mirabilite. Above 32.4°C and according to the relative humidity, it crystallizes as Thenardite or will go into solution [23].

According to the test conditions, the temperature degree in the oven was under 32.4°C during the first two hours. Thus, the specimens were in a room above 32.4°C for the remaining 8 hours of the heating phase. The crystallization of Mirabilite could occur during the first two hours inside the oven, and the specimens mainly experienced the crystallization of Thenardite during the remaining period of the drying cycle. When the specimens were put in the salt solution, Na<sub>2</sub>SO<sub>4</sub> could undergo hydration to form Mirabilite. Therefore, the specimens suffered two pressures: i) hydration pressure when Na<sub>2</sub>SO<sub>4</sub> (present inside pores) transformed to Na<sub>2</sub>SO<sub>4</sub>·10H<sub>2</sub>O inside the solution,

ii) crystallization pressure of Na<sub>2</sub>SO<sub>4</sub>. In our experiment, the specimens were exposed to 50 aging salt cycles of sodium sulfate.

Magnesium sulfate can be found as monohydrate (MgSO<sub>4</sub>·H<sub>2</sub>O), hexahydrate (MgSO<sub>4</sub>·6H<sub>2</sub>O), or Epsomite (MgSO<sub>4</sub>·7H<sub>2</sub>O), depending on the temperature and relative humidity. In our experiment conditions, MgSO<sub>4</sub>·6H<sub>2</sub>O crystallized in the first hours when the water inside the oven still provided proper humidity. The humidity decreased when the water evaporated, MgSO<sub>4</sub>·6H<sub>2</sub>O dehydrated and crystallized as MgSO<sub>4</sub>·H<sub>2</sub>O. In the cooling period, the laboratory temperature was above 20 °C, at which the crystallization of Epsomite (MgSO<sub>4</sub>·7H<sub>2</sub>O) was not possible, instead, MgSO<sub>4</sub>·H<sub>2</sub>O crystals could crystallize. The weathering form of this salt was characterized by exfoliation and layer detachment from the specimen surface. 40 aging salt cycles of magnesium sulfate were performed in this study.

NaCl is one of the frequently found salts in stone blocks [10]. It is considered as a simple salt, i.e., its crystals do not contain water molecules [37]. As such, the crystallization of NaCl salt exercised only crystallization pressure to the pore walls. The durability of stone specimens was tested under 50 cycles of sodium chloride solution.

### 3 Methods

The behavior of the stone specimens was evaluated under aging with different salts (sodium sulfate, magnesium sulfate, and sodium chloride). The standard for determining the resistance against salt crystallization of Na<sub>2</sub>SO<sub>4</sub> (MSZ EN 12370 [38]) was followed. Against the recommendation of the standard (drying temperature of 105 °C), 70 °C was used as drying temperature in this test.

14 w/w% salt solutions were separately prepared with NaCl, Na<sub>2</sub>SO<sub>4</sub>·10H<sub>2</sub>O, and MgSO<sub>4</sub>·7H<sub>2</sub>O using distilled water. Before running the experiment, the specimens (6 × 3 = 18 stone specimens) were dried in the oven at 70 °C to reach the constant mass ( $m_d$ ). By the end of the cooling

period of each cycle, the mass of each specimen ( $m_i$ ) was recorded. Salt crystallization occurred inside the stone structure because of the repeated salt solution-drying cycles and caused at first, an increase in the specimen mass. However, when the crystallized salt led to grain loss stone degradation, the specimen mass decreased due to salt crystallization damage. The change in specimen mass in each cycle was calculated as a percentage of the initial dry mass [w/w%] as in

$$D_{salt} = \frac{(m_i - m_d)}{m_d} \times 100 \left[ \frac{w}{w} \% \right], \quad (1)$$

where;  $m_i$  is the successive mass in each salt cycle [g], and  $m_d$  is the dry mass [g].

50 cycles of NaCl and  $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$ , and 40 cycles of  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  were made. Three groups of specimens were prepared from the stone cores, and each group was tested in one salt solution. This test aimed to study the behavior and resistance of different stone types in different salt solutions.

#### 4 Results and discussion

In this study, the weathering form of the stone specimens depended on the salt composition and the stone porosity. The results show that the durability of the stone specimens is directly related to the open pore percentage. The effect of NaCl crystallization on the specimen was not as aggressive as that of  $\text{Na}_2\text{SO}_4$  and  $\text{MgSO}_4$ . All lithotypes experienced salt efflorescence on their surfaces, and some were damaged. It should be noted that the possibility of taking samples in this study was limited due to the particularly endangered monument.

The micro bioclastic wackstone dolomite lithotypes (D1 and D6) were the most resistant and did not exhibit mass loss under the influence of the salts. Due to their lowest open porosity (around 11 v/v%, and 13 v/v%, respectively), no damage could be seen visually on their surfaces. Their masses barely increased during the tests (on average 3.5% and 0.6%, respectively), as shown by Fig. 4, Fig. 5, and Fig. 6. No  $\text{Na}_2\text{SO}_4$  efflorescence was recognized on their surfaces. NaCl accumulated heavily on the D6 surface; however,  $\text{MgSO}_4$  appeared in a very fine salt layer on it. The efflorescence of NaCl and  $\text{MgSO}_4$  did not arise on D1.

The microcrystalline dolomites (D2, D3, D4, and D5) had the same open porosity (32 v/v%), but their durability was different. D2 and D4 had the lowest closed porosity (0.3 v/v% and 0.4 v/v%, respectively), so they showed different behaviors than the former specimens (D3 and D5). Under the crystallization of sodium sulfate, D4 and D2

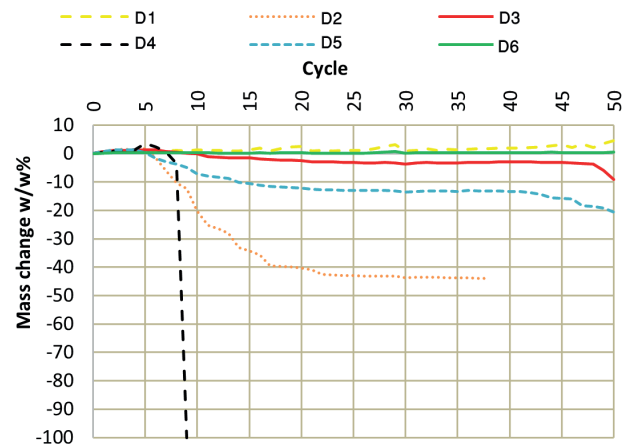


Fig. 4 The mass change as a percentage from the initial dry mass of the specimens in sodium sulfate solution

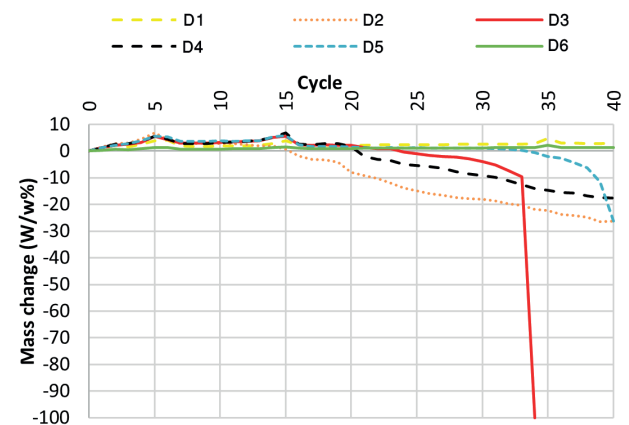


Fig. 5 The mass change as a percentage from the initial dry mass of the specimens in magnesium sulfate solution

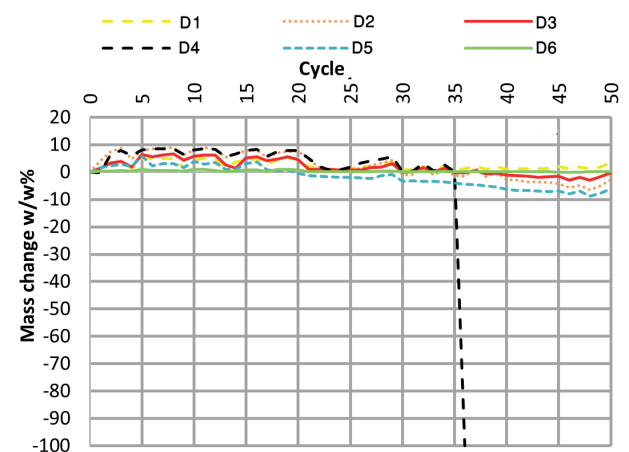


Fig. 6 The mass change as a percentage from the initial dry mass of the specimens in sodium chloride solution

were damaged and experienced a granular disintegration and a gradual loss in stone material (about 40% on average) as shown by Fig. 7. However, D4 suffered a rupture resulting in the specimen breaking in half at the 8<sup>th</sup>

cycle. D3 and D5 had the highest closed porosity (approximately 4 v/v%) and had approximately the same resistance against sodium sulfate, showing a loss in stone material. D5 showed up to 12.7 w/w% material loss from the 5<sup>th</sup> to the 21<sup>st</sup> cycle. Afterward, the specimen experienced a quiet stagnation period between the 22<sup>nd</sup> and the 41<sup>st</sup> cycle, and then it demonstrated a gradual material loss until arriving 20 w/w% by the end of the 50<sup>th</sup> cycle as shown by Fig. 4.

D2 and D4, which have the lowest closed porosity, exhibited similar behavior in the aging test with  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ , where both specimens started to lose their material at the 11<sup>th</sup> and 15<sup>th</sup> cycles, respectively. The specimens were weathered as a form of exfoliation. Very thin stone layers detached from the surface until the specimens had uniform surfaces as shown by Fig. 5 and Fig. 7. In the salt crystallization test with magnesium sulfate D3 and D5 (have the lowest closed porosity) showed different resistance behavior not only comparing to other specimens under the same salt, but comparing with D3 and D5 under the other two salts aging. D3 had a crack appeared on its surface. This crack expanded and developed along the height of the specimen until dividing it into two parts at the 34<sup>th</sup> cycle. The surface of the specimen also exhibited edge exfoliation on the top side (where the crack was present) from the 15<sup>th</sup> cycle as shown by Fig. 7. After that, its surface gradually exfoliated, and the material loss reached 10 w/w% before it broke. D5 increased its mass until the 15<sup>th</sup> cycle. After that, the edges started to detach, and the surface exfoliated, resulting in a gradual material loss (see Fig. 5 and Fig. 7). Between the 37<sup>th</sup> and the 40<sup>th</sup> cycle, the mass of the specimen dramatically decreased until the mass loss reached 26 w/w% as shown by Fig. 5.

The primary weathering phenomenon of the highly porous stone types caused by NaCl crystals was the forming of gradually pitted surfaces as shown by Fig. 6 and Fig. 7. D2 and D4, with the lowest closed porosity, increased their mass continuously until the 15–20<sup>th</sup> cycles, then started the mass loss. In addition, D4 was suddenly ruptured in the 36<sup>th</sup> cycle breaking in half. D3 had the highest percentage of closed pores (about 4.5 v/v%); therefore, it was very resistant. A mass increase was observed by D3 in the case of the NaCl aging test and it lasted until the 35<sup>th</sup> cycle. After that, this specimen lost its material slowly, whereas, by the 50<sup>th</sup> cycle, the mass was reduced by 3%. Under the same conditions, the mass of D5 increased to a lesser extent (around 0.8 w/w%) and after the 19<sup>th</sup> cycle it started to decrease in small percentages. By the end of the 50<sup>th</sup> cycle reached the mass loss 8 w/w% as shown by Fig. 6.

D3 and D5 were less durable in  $\text{MgSO}_4$  than in NaCl and  $\text{Na}_2\text{SO}_4$  solutions. Where D3 broke at the 34<sup>th</sup> cycle, and D5 lost around 30% of its stone material, which was close to the loss of D2 and D4. However, both (D3 and D5) about 15% (in average) loss in stone material under  $\text{Na}_2\text{SO}_4$ ; and around 3% (in average) under NaCl.

Salt crystallization can deteriorate the stone blocks, causing granular disintegration, alveolization, scaling (exfoliation), discoloration, and cracks [39]. The environmental conditions, the stone composition and the concentration of the presented salt define which type of decay patterns can appear on the stone [40]. At the laboratory scale, those decay patterns differ depending on the salt composition and the conditions of the salt aging experiment (whether the stone specimens are fully immersed or subjected to salt solution by capillary absorption). Ruiz-Agudo et al. [27] studied the difference between the effect of capillary absorption of sodium sulfate and magnesium sulfate solution on limestones. Their results demonstrated that the specimens exposed to the capillary absorption of sodium sulfate solution revealed exfoliation on the stone surface in very thin layers; however, the ones exposed to the capillary absorption of  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  solution, showed granular disintegration. In contrast to this investigation, in our study the specimens, which were fully immersed in the salt solution, exhibited the opposite weathering decay. The lithotypes with high porosity showed granular disintegration by weathering with sodium sulfate solution; and gradual exfoliation of stone surface in magnesium sulfate solution.

In case of the same porosity properties of the lithotypes, our specimens demonstrated during the aging test of one salt one distinct weathering form. Whereas other experiments exhibited that the different stone lithotypes suffered different weathering forms under the same salt solution.

Galanaki et al. [25] tested the durability of various stone lithotypes (biocalcarenes and calcitic sandstone) against sodium chloride crystallization by total immersion in the salt solution, where the stones demonstrated various decay forms such as granular disintegration and exfoliation of stone surface (multiple flaking).

Yu and Oguchi [12] and Jamshidi et al. [24] studies revealed that effective porosity considerably influenced the quantity of absorbed salt and hence the produced damage. Our study showed that specimens with similar effective porosity did not induce the same damage degree because the closed porosity played a significant role in the weathering processes. D2, D3, D4 and D5 had around

























Specimen	Initial dry phase	Na <sub>2</sub> SO <sub>4</sub>	NaCl	MgSO <sub>4</sub>
D1		 At 50 <sup>th</sup> cycle	 At 50 <sup>th</sup> cycle	 At 40 <sup>th</sup> cycle
D2		 At 38 <sup>th</sup> cycle	 At 50 <sup>th</sup> cycle	 At 40 <sup>th</sup> cycle
D3		 At 50 <sup>th</sup> cycle	 At 50 <sup>th</sup> cycle	 At 34 <sup>th</sup> cycle
D4		 At 9 <sup>th</sup> cycle	 At 36 <sup>th</sup> cycle	 At 40 <sup>th</sup> cycle
D5		 At 50 <sup>th</sup> cycle	 At 50 <sup>th</sup> cycle	 At 40 <sup>th</sup> cycle
D6		 At 50 <sup>th</sup> cycle	 At 50 <sup>th</sup> cycle	 At 40 <sup>th</sup> cycle

Fig. 7 The stone specimens before and after the salt aging test

28 v/v% effective porosities. D3 and D5 were more durable and resistant than D2 and D4, because they had a high percentage of closed porosity (around 4v/v%).

In our investigation and by observing the specimen behavior under the hydrated salt solutions (sodium sulphate and magnesium sulphate), the stone material ruptured during the immersion period. That observation corresponded to the investigations of Yu and Oguchi [16]. They confirmed that the highest crystallization pressure occurs during the immersion period, wherein the salt solution has the greatest degree of supersaturation.

## 5 Conclusions

When evaluating durability under the salt aging test, the role of the open and closed porosity has a significant effect. Even though the porous lithotypes with 4 v/v% closed porosity exhibited high durability against sodium sulfate and sodium chloride, those specimens were not durable enough against magnesium sulfate (where D3 was broken and D5 underwent a gradual material loss, especially during the last cycles). Their bimodal pore system can explain this phenomenon. However, their open porosity is

equal to the porosity of other lithotypes, the pore size distribution is not uniform, so the crystallization of magnesium sulfate damages these stones more.

The stones with the highest open porosity (D2, D3, D4, D5) exhibited a distinct weathering form according to the salt solution and showed different durabilities depending on the closed porosity. The specimens in sodium sulfate solution were characterized by granular disintegration. However, pitted surfaces belonged to the specimens under sodium chloride solution. The exfoliation of the stone surface into thin layers was a characteristic weathering form by aging in magnesium sulfate solution.

## Acknowledgement

Authors would like to thank all workers at Crac des Chevaliers castle specially eng. Ahmad Alboush and eng. Hazem Hanna for their help in the field works.

Authors would also like to thank Katalin Kopecskó, Viktória Rónaky, and Ákos Török for their help in the determination of the mineralogical composition of the lithotypes and in the analysis of the thin sections.

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