

# Stone properties and weathering induced by salt crystallization of Maltese Globigerina Limestone

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**Abstract:** Most monuments and buildings in the Maltese Islands are constructed of the local Globigerina Limestone. Today, this Globigerina Limestone shows considerable damage in many buildings, particularly through alveolar weathering, which is frequently very intense. Owing to Malta's marine environment, salt crystallization in the stone's pore spaces has been recognized as the main weathering process responsible for the deterioration of the country's monuments. In order to obtain more information on the fabric-dependent weathering processes of Globigerina Limestone, detailed analyses were carried out. Globigerina Limestone samples obtained from stone types with two different known qualities were characterized according to petrographical, geochemical and physical properties. These included porosity, pore radii distribution and tensile strength, as well as water and humidity transport properties. Investigations by means of salt crystallization tests on quarry samples of both stone types reinforced the idea that the extent of salt weathering depends on salt type and concentration and pore-space properties. Visible weathering damage was recorded and evaluated for a representative monument (the Church of Santa Marija Ta' Cwerra in Siggiewi) by means of a monument mapping method, which was carried out twice over a period of 9 years (1995 and 2004). The identified weathering forms were also correlated with a previously developed weathering model for Globigerina Limestone. According to the results of the mapping, salt analyses carried out on samples from the church and salt-loading tests on quarry samples, there exists a significant correlation between visible damage and salt load. The zoning of weathering damage is obviously related to different salt concentrations. The zone with severe weathering damage is characterized by a high concentration of halite. Consequently, salt weathering represents the main damage process for the Globigerina Limestone of Malta.

The Maltese archipelago of Malta, Comino and Gozo lies in the central Mediterranean Sea approximately 90 km south of Sicily. During the geological epochs of the Oligocene and Miocene 30–5 Ma ago, extensive sedimentation took place in this area. This sedimentation led to the laying down of beds of lime- and mudstones (Pedley *et al.* 2002).

The Maltese Globigerina Limestone Formation is one of five main formations, and varies in thickness from 20 to over 200 m. The material used for building is located stratigraphically in the lower part of the Globigerina Limestone Formation, called the Lower Globigerina Limestone. During the deposition of the sediments that eventually formed this stone, far away from the continent and below the action of waves, only minor but variable amounts of clay in suspension were carried from a distant land source. The Globigerina Limestone also contains numerous shells, algae

and planktonic fossils, mainly the planktonic foraminifera Globigerina (Pedley *et al.* 2002).

As can be widely seen in the Maltese Islands, the local limestone has always been used as the predominant building material. The Maltese prehistoric temples, which were constructed approximately 6000 years ago, bear testimony to this (Cassar & Vannucci 2001). Between 1530 and 1798 the Order of the Knights of St John built kilometres of fortifications to protect the island from the expanding Ottoman Empire. Fortifications, impressive churches (Fig. 1) and palaces were built of the local building stone during this period. The capital city of Valletta is included in the UNESCO World Heritage List, as are the prehistoric temples. Even today, the local building stone is still much in demand. Many modern buildings are constructed using the Globigerina Limestone (Fig. 2a) and many local quarries are still active (Fig. 2b).



**Fig. 1.** The Baroque parish church of Siggiewi is one of numerous impressive Maltese buildings constructed from Globigerina Limestone.



(a)



(b)

**Fig. 2.** (a) Modern buildings in Globigerina Limestone show that to this day this limestone is still one of the most important building materials in Malta. (b) This quarry, which is still active today, shows the extraction of Globigerina Limestone. The limestone blocks, here exposed to rain and sun for a short period, were previously left for much longer and led to a hardening of the stone surface.

### Aim of this paper

This paper discusses salt-induced weathering of the Maltese Globigerina Limestone. On the basis of work carried out in the past (Fassina *et al.* 1996; Fitzner *et al.* 1996; Torfs *et al.* 1996) it has been

found to be necessary to obtain more information about the damage processes and to emulate site salt loading and its relation to the stone fabric and different stone types. Thus, this work has had a multifaceted approach. One part consisted of the detailed investigation of the two main qualities of Globigerina Limestone, derived from different layers of a quarry near the village of Siggiewi, which lies in the main quarry area of the Maltese Islands. Here, detailed petrographical and fabric determinations, and numerous petrophysical analyses, have been carried out on these fresh samples to supplement previous work (Vannucci *et al.* 1994; Galan *et al.* 1996; Cassar 1999, 2002) and also to subsequently characterize the weathering resistance of these two limestone types with respect to salt content. In addition, the local occurrence of weathering forms and their distributions were meticulously recorded on two separate occasions for the Church of Santa Marija Ta' Cwerra, also in Siggiewi, where the weathering problem owing to salt load is clearly evident. In the past, detailed scientific analyses of salt load within this building had also been carried out (Fassina *et al.* 1996). The results from laboratory weathering resistance tests with halite, thenardite and epsomite have now been compared with prevailing damage phenomena at the church that have been attributed to salt load. An important part of this work has also been the comparison of the mapping of this church carried out in 1995 with that carried out in 2004, to determine the rate of stone damage on this building. This work has also been useful to help determine materials and methods used for recent conservation work on the four external façades of the church.

### Deterioration of Globigerina Limestone

Today it is often, although not always, the case that the stone in older buildings in Malta is badly deteriorated. Frequently, the main deterioration phenomenon is alveolar weathering. A model developed for Globigerina Limestone deterioration some years ago explains that the weathering process initiates by the dissolution and reprecipitation of the mineral calcite, which at first leads to the formation of a thick and compact superficial crust (Vannucci *et al.* 1994; Fitzner *et al.* 1996). Areas of stone that demonstrate a loss of calcite are observed to become weaker. In addition to this, the main weathering process responsible for the deterioration of the building stone has been recognized as salt crystallization in the pore spaces of this very porous limestone (Cassar 2002). For this reason, parts of the hardened and compact surface fall off. The main source of the

salt is the surrounding marine environment (Torfs *et al.* 1996). The  $\text{SO}_x$  loads that derive from urban pollution and lead to the development of sulphates can here be mostly disregarded, except in areas downwind from power stations (Torfs *et al.* 1996). The weathering intensity varies because of local differences in the salt types and content in buildings, and because of differing quality and weathering resistance of the natural building stone itself (Cassar 2002).

Local stoneworkers distinguish macroscopically between two building stone qualities: 'Franka' and 'Soll'. Whereas 'Soll' represents *bad* quality building material, 'Franka' tends to resist the local environmental conditions well. In the fresh state, the two types cannot be distinguished visually, although a geochemical test and the pore-radii distribution may help to identify the two qualities (Farrugia 1993; Fitzner *et al.* 1996; Cassar 1999; Cassar & Vella 2003). In addition, on abandoned quarry faces 'Franka' and 'Soll' can be seen to differ in their weathering intensities. It can also be seen that in buildings, and occasionally in quarries, both types can sometimes coexist in the same horizontal layer, forming local areas with different geometries and shapes. As the weathering of the good-quality Globigerina Limestone leads to hardening of the stone surface, fresh quarry stones were in the past exposed to rain and sun for a long period before being utilized (Fig. 2b). This practice has been abandoned, primarily for economic reasons.

### Sampling

At the start of the present testing programme, quarry owners were asked to supply two different stone types – what they considered to be 'Franka' (i.e. *good* stone) and 'Soll' (i.e. *bad* stone). These will be identified in this paper as Type I (*good* stone) and Type II (*bad* stone). Four standard stone blocks, measuring 229 × 260 × 610 mm, of each type were supplied.

### Analytical methods

Petrographic analyses (in polarized light) on standard thin sections of both Globigerina Limestone types were performed to obtain a qualitative description of different fabric parameters of the investigated rock samples (e.g. mineralogical composition, properties of detrital and authigenic components). Previous work had concentrated on only one type of Globigerina Limestone (Vannucci *et al.* 1985; Galan *et al.* 1996). To analyse the bulk rock composition, X-ray fluorescence (XRF) was carried out (cf. Cassar 1999). For a quantitative

determination of pore-size distribution, mercury porosimetry was used (cf. Brakel *et al.* 1981).

The total accessible porosity of the two stone types was also characterized – samples were thus measured using buoyancy weighing. The dry weight, the water-saturated weight and the weight immersed in water of cubic samples (65 × 65 × 65 mm) were. To determine the directional dependence of capillary water absorption, the same device as for the buoyancy weighing and cubes with the same dimensions were used, but were only dipped 1 cm into water. To analyse the degree of saturation (S-value) of free moisture absorption, the sample weight after 24 h of water immersion was again measured. The S-value represents the ratio between the free capillary water uptake and the maximum uptake under vacuum.

The water vapour diffusion resistance value,  $\mu$ , of the limestones was studied using the wet-cup method. This value characterizes the diffusion resistance of a porous material compared to an equal inactive air film. Slices of the stones ( $\phi$  40 × 10 mm) were used as covers on the cups. The relative humidity difference caused moisture to flow through the porous material from the side with higher moisture (inside at 100%) to the side with lower relative humidity (outside at 50%). The moisture flow was obtained by weighing the cups at various times until a steady state was reached.

The tensile strength ( $\sigma_z$ ) was determined by means of the 'Brazilian test', which involves disc-shaped specimens. The diameter of the samples was 40 mm and the length was 20 mm. To obtain the average value, a minimum of four samples were used. A constant strain rate of  $0.3 \times 10^{-6} \text{ mm s}^{-1}$  ( $\approx 10^{-5} \text{ s}^{-1}$ ) was applied.

Ultrasonic velocity measurements were carried out on cubic rock samples (65 × 65 × 65 mm). Transient times of ultrasonic pulses (piezoceramic transducers, resonant frequency 1 MHz) were measured in three orthogonal directions using the pulse transmission technique (Birch 1960, 1961).

The thermal expansion behaviour of the investigated samples was measured on cylindrical specimens ( $\phi$  15 × 50 mm). This was determined as a function of temperature. For this dilatation experiment, a heating cycle from 20 to 90 °C was employed. The experimental set-up allows simultaneously investigation of six samples. The thermal expansion coefficient,  $\alpha$ , expresses the volume change of a material as a function of temperature.

Hygic expansion was determined on cylindrical samples ( $\phi$  15 × 50 mm), which were preconditioned at 30% relative humidity and room temperature. Afterwards, the samples were completely immersed in distilled water. The accuracy of the incremental displacement transducer is 1.0  $\mu\text{m}$ .

## Results

### Rock fabric

The investigated samples of both types of Globigerina Limestone can be described as soft and almost pure limestone with a pale cream–yellow colour. They are fine grained and homogenous. In thin sections, it was confirmed that in both types large concentrations of randomly distributed and non-orientated microfossils, mainly round planktonic globigerinae and some elongated forms, occur. They often make up 80 vol.%, whereas the micritic matrix is only about 20 vol.%. Finely dispersed iron oxides and iron hydroxides, mainly limonite, can also be observed. No clear differences were distinguished between the two stone types, as had already been recognized in previous studies (Cassar 1999).

The limestone fabric is grain-supported with a micritic matrix, and can be described as foraminiferal packstone, although wackestones also occur (Cassar 2004). The pore space is formed by intergranular pores, secondary solution pores and often by empty fossil chambers. As can be seen from the geochemical investigations (X-ray fluorescence) the Globigerina Limestone contains minor amounts of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> phases (Table 1). These are a result of sparse quartz and clay minerals (cf. Cassar 1999, 2002; Galan *et al.* 1996).

**Table 1.** Properties of two fresh samples of Globigerina Limestone

Stone properties	Type I	Type II
Bulk composition [wt%]		
CaO	52.2	51.9
SiO <sub>2</sub>	2.70	3.90
Al <sub>2</sub> O <sub>3</sub>	1.20	1.10
MgO	0.85	0.86
Fe <sub>2</sub> O <sub>3</sub>	0.73	0.59
Porosity [vol%]	36.46	34.59
Pore-radii distribution [%]		
0.001–0.01 μm	0.00	0.71
0.01–0.1 μm	5.56	10.04
0.1–1 μm	27.85	47.02
1–10 μm	66.59	41.91
> 10 μm	0.00	0.33
Average pore radii [μm]	1.06	0.56
Capillary water uptake ( <i>w</i> -value) [kg m <sup>2</sup> · h <sup>-0.5</sup> ]	6.74	8.73
Water vapour diffusion resistance, μ	7.78	7.83
Saturation degree	0.69	0.76
Tensile strength [MPa]	2.96	2.83
Ultrasonic velocity, V <sub>p</sub> [km s <sup>-1</sup> ]	2.95	2.84
Thermal dilatation coefficient [K <sup>-1</sup> ]	2.32	4.51
Hygric expansion [mm m <sup>-1</sup> ]	0.20	0.25

### Physical properties

Both types of Globigerina Limestone are very porous with a relatively low tensile strength and high water absorption (Table 1). The effective porosity for both investigated types is comparable at approximately 35 vol.%, whereas distinct differences in the pore-radii distribution occur. Type I has, at 67%, a high proportion of pores in the range above 1 μm. In contrast, Type II has only 42% of its pores in this range. Correspondingly, the pore volume of pores smaller than 1 μm is 33% for Type I, whereas Type II has a very high proportion of these pores, at 58%. Concerning the water absorption coefficient of Globigerina Limestone, both investigated types show a very high *W*-value ranging between 6.74 and 8.73 kg m<sup>2</sup> h<sup>-0.5</sup>. Thus, they can be described as rapidly absorbing stones. Site observations indicate that large amounts of water are absorbed during heavy rain. There is practically no run-off from the walls, even though guttering is absent in most Maltese buildings. The saturation value, i.e. the ratio between capillary water uptake (*w*) and effective porosity of the Globigerina Limestones, is, at 0.69 and 0.76, relatively low.

The physical properties are dependent on the limestone fabric and prove the presence of only slight diagenetic hardening. Otherwise, the two stone types show few differences. For example, the tensile strengths of the selected stone types differ slightly between 2.96 and 2.83 MPa. Even the ultrasonic velocities (in this case the compressional wave velocities, V<sub>p</sub>) are almost comparable: 2.95 and 2.84 km s<sup>-1</sup>. With regards to thermal dilation, however, a notable difference can be detected. The dilation coefficient of Type I is, at 2.32 K<sup>-1</sup>, almost half that of Type II at 4.51 K<sup>-1</sup>. A special phenomenon seen in both samples is a pronounced hygric expansion. This notable property is attributed to the presence of small amounts of swelling clay minerals, in particular the minerals smectite and illite-smectite (Vannucci *et al.* 1994).

### Damage mapping

The monument mapping method is a phenomenological, but meaningful, tool for the non-destructive registration of decay features. The stone surface is examined visually and observable changes are compared to the original condition of the building stone. The applied mapping method was based on a classification scheme proposed by Fitzner & Kownatzki (1990, 1991), Fitzner *et al.* (1992, 1995) and Kownatzki (1997).

The current mapping was carried out on the Church of Santa Marija Ta'Cwerra and focused primarily on a comparison with preceding

investigations, which had also included a detailed mapping of all four external façades of the church in 1995 (Fassina *et al.* 1996; Fitzner *et al.* 1996; Torfs *et al.* 1996). This comparison was aimed at determining and classifying the progress of deterioration with time.

#### *Church of Santa Marija Ta' Cwerra*

The church is located in the centre of Siggiewi, a village in the SW of Malta about 3 km from the coast. A building in this same location dates back to the 16th century, while the present monument was rebuilt in the 18th century. It is a small free-standing church of 10 × 10 m square and is built entirely of the local Globigerina Limestone. Only the lower courses have been covered with plaster, presumably to stop progressive deterioration. The weathering response of Globigerina Limestone to salt loading is a significant phenomenon at this church.

Over the last decade this monument has been extensively investigated. These studies included, besides the mapping of damage forms, several types of analyses aimed at understanding and quantifying the salt load (Fassina *et al.* 1996; Fitzner *et al.* 1996; Torfs *et al.* 1996). The damage recorded in a detailed monument mapping exercise 9 years previously (Fitzner *et al.* 1995) has now been compared with recent mapping (2004) to illustrate the changes in the damage forms and intensities after these years.

The work by Fitzner *et al.* (1995) determined the back-weathering rates. As the back-weathering provides insufficient information about the local damage distribution on heterogeneous back-weathered façades, a detailed mapping of damage forms was additionally performed. The main advantage of this approach is that the damage observed can be attributed to a damage phase of the weathering model by Vannucci *et al.* (1994) and Fitzner *et al.* (1996). Thus, an index of each individual stone conforming to the state of weathering is

possible and a better correlation for future works can be derived.

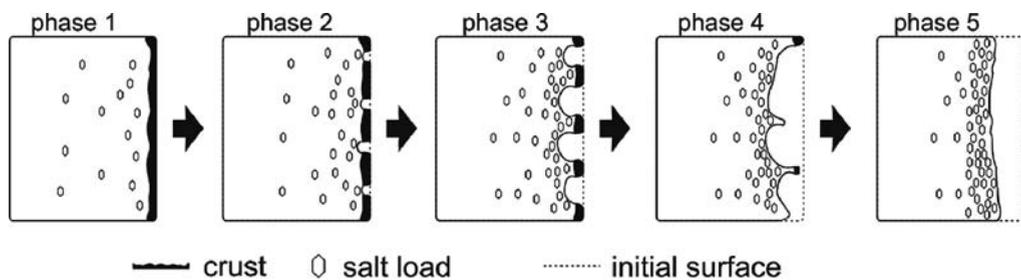
#### *Mapping results and correlations to weathering model*

The predominant damage phenomena occurring at the church were seen to be 'relief' and 'back-weathering'. In the model for the damage processes suggested by Vannucci *et al.* (1994) and Fitzner *et al.* (1996), these authors distinguished five phases of the damage development (Fig. 3). The classification of the current mapping was harmonized with the damage phases of this weathering model.

The original, non-weathered stone surface was one of the mapping forms identified. Following the damage model, at this stage of preservation, the formation of a superficial crust by re-precipitation of dissolved calcite can be seen to be taking place (phase 1, Fig. 3).

Slight-medium 'back-weathering' in the form of alveolar weathering was also observable. This is the mapped form called 'initial relief'. In this case, the stone surface is back-weathered through the formation of neighbouring cavities. Following the damage model by Vannucci *et al.* (1994) and Fitzner *et al.* (1996), this state represents damage phases 2 and 3 (both phases could not be distinguished on site). Local back-weathering with the formation of cavities can be traced back to cracking and/or partial loss of the crust owing to mechanical stress provoked by salt crystallization. Further accumulation of salt behind the crust leads to a detachment of the stone material in the form of granular disintegration and flaking within the cavities. The cause of the preferred back-weathering of the cavities is probably the increase of evaporation in areas where the crust has been lost.

The mapped form 'advanced relief' describes a weathering state where material loss and the formation of alveoli is very pronounced (phase 4 of the damage model). Also, connection of the



**Fig. 3.** Schematic representation of damage development, divided into five distinct phases (modified from Fitzner *et al.* 1996; explanation in text).

alveoli occurs. The septa of the honeycomb structure here are severely back-weathered, but are still recognizable.

The mapped form of ‘back-weathering’ represents the final deterioration state (phase 5 of the damage model) and follows after the ‘advanced relief’ weathering. The septa of the honeycombs are here totally back-weathered so that a more or less plain surface has developed and the edges of the blocks become rounded. Although back-weathering represents the final state of decay in the damage model, progressive material loss in form of flaking and granular disintegration is still subsequently observable on site.

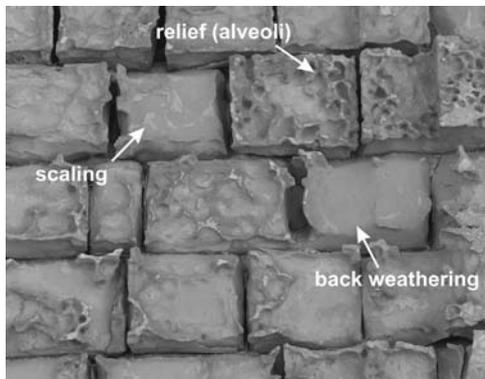
In Figure 4, the distribution of damage forms is shown for the south façade of the church. The uppermost part of this façade is characterized by

original and undamaged stone surfaces. However, in the lower parts of the wall the predominant damage phenomena are ‘relief’ and ‘back-weathering’ (Fig. 5). The middle part of the mapped façade is dominated by ‘initial’ and ‘advanced relief’ forms. Furthermore, a zoning of the relief forms can be observed. ‘Advanced relief’ is characterized by irregular back-weathered stone surfaces, mainly located in the lower middle part, while ‘initial relief’ development is observed in the upper middle part. Material loss in the areas where both relief forms occur is characterized by granular disintegration and crumbling in the cavities.

Very severe damage can be noticed in the lower parts of the walls, directly above the plastered lower courses (Fig. 4). In this part of the masonry



**Fig. 4.** Mapping of damage forms on the Church of Santa Marija Ta' Cwerra (south façade; 2004) reveals a clear distribution of weathering phenomena. Three zones in vertical order can be distinguished. Severe damage in the form of back-weathering occurs in the area immediately above the plastered lower courses. Further up, the middle zone is seen to have severe to moderate damage. Here alveolar weathering additionally affects numerous building stones. With the exception of a few building stones, the uppermost zone has remained in relatively good condition. These stones have developed a red–brown patina typical of good local building stone. There are, however, areas with organic deposition.



**Fig. 5.** Detachment and loss of stone material characteristic of the south façade. Relief in the form of alveoli and back-weathering by scaling are the prevalent weathering forms.

‘back-weathering’ dominates, and stone decay occurs by flaking and contour scaling. Flakes and larger detached parts of the stone surface are characterized by a bright brownish colour.

The intensity of stone loss evaluated during the mapping has been subdivided according to the estimated depth of the back-weathered surface, namely, slight (<3 cm), moderate (3–5 cm) and severe (>5 cm). Back-weathering of the whole stone surface can be observed on the lower part of the church, above the plastered lower courses. However, the comparison between the mappings from 1995 (Fitzner *et al.* 1995) and 2004 (Rothert 2004) showed that the weathering intensity had changed only slightly over the years.

#### Damage mechanisms

The distribution of decay features on the south façade indicates a significant influence of moisture by capillary water uptake. The most severe deterioration is in fact observed on this façade, whereas the north and east façades are less affected. This is most probably explained by the sun’s radiation being a critical instigator of damage. The sun influences the water evaporation rate and, consequently, the capillary suction. However, characteristic damage phenomena were observed on all four façades of the church.

The state of decay of the church can be correlated with salt-loading data from Fassina *et al.* (1996). The reported anion content of the wall masonry shows a typical distribution (Table 2) with sulphates mainly concentrated in the lower parts of the walls, while chlorides and nitrates occur in the upper parts. This salt distribution also indicates vertical capillary rising damp. Thus, the content and

**Table 2.** Water-soluble anion content of drilled core samples at different heights and depths (Fassina *et al.* 1996)

Height [m]	Depth [cm]	Cl <sup>-</sup> [%]	NO <sub>3</sub> <sup>-</sup> [%]	SO <sub>4</sub> <sup>2-</sup> [%]
0.5	0–5	0.12	0.22	0.70
0.5	5–15	0.10	–	0.22
1.5	0–5	0.67	0.44	0.30
1.5	5–15	0.76	0.27	0.18
2.5	0–5	2.27	0.72	0.20
2.5	5–15	1.12	0.62	0.18
3.5	0–5	0.49	0.56	0.19
3.5	5–15	0.47	0.55	0.18

distribution of the salts in the wall confirm the results of our mapping. X-ray diffraction analyses by Fassina *et al.* (1996) demonstrated the predominance of halite. In addition, both crystalline phases (thenardite and mirabilite) for sodium sulphate were detected. The origin of the sulphates is probably from the mortar.

#### Salt resistance tests

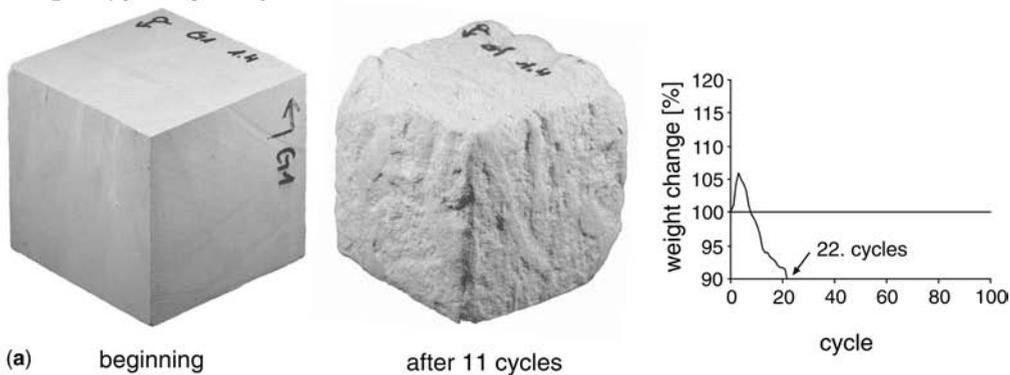
Salt-loading tests in the laboratory were carried out to verify the weathering susceptibility of the two types of Globigerina Limestone. For this purpose crystallization tests by means of halite, thenardite and epsomite were carried out on a number of stone cubes (65 × 65 × 65 mm). The samples were submitted to wetting and drying cycles as follows: loading with 10% salt solution for 4 h, followed by a drying cycle with a duration of 16 h at 60 °C. After cooling to room temperature, the weight change was determined.

The weathering effect of sodium sulphate is considered to be a result of the transformation of the water-free thenardite (Na<sub>2</sub>SO<sub>4</sub>) to the hydrated phase mirabilite (Na<sub>2</sub>SO<sub>4</sub>·10H<sub>2</sub>O). The salt hydration is coupled with a volume increase of about 300% (Price & Brimblecombe 1994). The transformation of the water-free kieserite (MgSO<sub>4</sub>·H<sub>2</sub>O) to the hydrated phase epsomite (MgSO<sub>4</sub>·7H<sub>2</sub>O) is also associated with a volume increase of about 173%. These tests were carried out to investigate how hydration pressure may affect the durability of Globigerina Limestone. For sodium chloride (NaCl) crystallization pressure is held to be the main damage factor. Both the expected hydration pressure and the crystallization pressure are greater than the tensile strength of porous natural stone. In addition, the rock fabric may also affect the durability of natural stones, whereas the pore-radii distribution should be of additional importance (Sneath 1984).

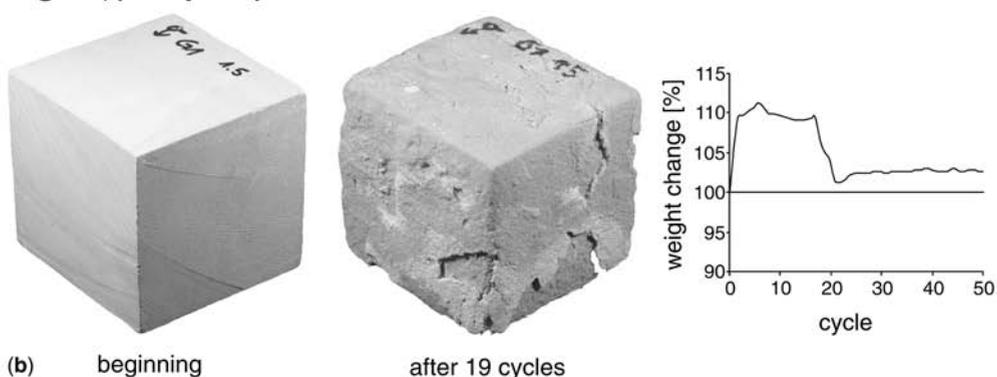
The results of the salt action show that the limestone samples for both stone types submitted to the sodium sulphate test exhibited a slight granular disintegration at the surfaces already after the

second cycle. For the samples seen in Figure 6a, back-weathering is most pronounced parallel to pre-existing sedimentary structures, evident after the sixth cycle. The tests were discontinued after a

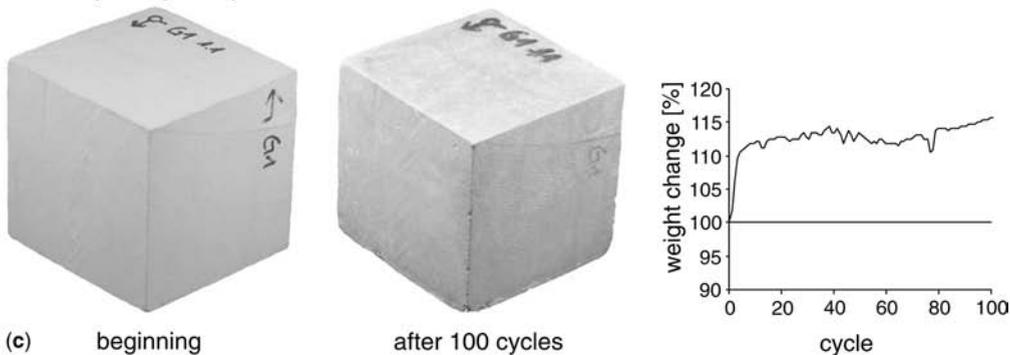
### $\text{Na}_2\text{SO}_4$ (sample II)



### $\text{MgSO}_4$ (sample II)



### $\text{NaCl}$ (sample II)



**Fig. 6.** Results of the salt-loading tests. (a) Samples that have undergone the sodium sulphate test, at the beginning, and showing definite deterioration after 11 cycles of artificial weathering. (c) Largely unchanged sample that has undergone the sodium chloride test, at the beginning, and after 100 cycles. On the right-hand side, the respective change in weight is plotted against the number of loading cycles.

loss in weight of 10%. The overall result was that the stone cubes of Type II resisted 22 or 24 such cycles, whereas the Type I cubes behaved differently. Here, for one sample, the critical weight loss was achieved after 17 cycles, while for another sample this happened after 51 cycles. This indicates that the latter samples were probably more heterogeneous.

The salt-loading tests with magnesium sulphate demonstrated that the Globigerina Limestone also showed a slight granular disintegration at the edges and the surfaces after the second weathering cycle. After the ninth cycle and again after the 19th cycle, an obvious scaling effect occurred (Fig. 6b). A total of 50 salt-weathering cycles were performed, although after the 25th cycle no further macroscopic changes occurred. The changes in weight were at a maximum after the sixth cycle (where an 11% increase in weight was registered), which means that the samples retained a large amount of the  $MgSO_4$ . After the breakaway of the scales, a decisive weight loss was ascertained.

In contrast, the samples loaded with sodium chloride were only slightly affected, even after 100 test cycles (Fig. 6c). For both investigated stone types an increase in weight was observable, which is again the consequence of salt enrichment. Only slight granular disintegration occurred at the edges of these specimens. Thus, the damage caused by sodium chloride was only slight when compared to the effects of sodium sulphate.

## Conclusion

The damage and the salt content distribution in the walls of the Church of Santa Marija Ta' Cwerra show a distinct correlation. This suggests that salt weathering is the main damage process for Globigerina Limestone on the island of Malta. The salt-loading tests in the laboratory demonstrate damage primarily for sodium sulphate and, to a lesser extent, for magnesium sulphate. On the other hand, the lack of damage by loading with sodium chloride indicates that the high halite content in the building is not the only cause of the observed damage. To obtain more information on the damage processes, further research is necessary to emulate the site loading and its relation to the stone fabric. This is of particular importance because the conservation approach in Malta has changed in recent years. In the past, the Maltese carried out stone replacement because the required natural stone was to be found in great quantities and the stone-working skills still existed. Today, preservation by means of modern conservation methods is accepted and largely carried out. However, to attain an acceptable degree of preservation by this approach, knowledge of the weathering processes

and the contamination paths is of fundamental importance.

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## References

- ARNOLD, A. & ZEHNDER, K. 1990. Salt weathering on monuments. In: *Advanced Workshop Analytical Methodologies for the Investigation of Damaged Stones*, 14–21 September 1990, La Goliardica Pavese srl, Italy.
- BIRCH, F. 1960. The velocity of compressional waves in rocks up to 10 kilobars. Part I. *Journal of Geophysical Research*, **65**, 1083–1102.
- BIRCH, F. 1961. The velocity of compressional waves in rocks up to 10 kilobars. Part 2. *Journal of Geophysical Research*, **66**, 2199–2224.
- BRAKEL, J. VAN, MODRY, S. & SVATA, M. 1981. Mercury porosimetry: State of the art. *Powder Technology*, **29**, 1–12.
- CASSAR, J. 1999. *Geochemical and mineralogical characterisation of the Lower Globigerina Limestone of the Maltese Islands with special reference to the 'soll' facies*. Ph.D. thesis, University of Malta.
- CASSAR, J. 2002. Deterioration of the Globigerina limestone of the Maltese Islands. In: SIEGSMUND, S., WEISS, T. & VOLLBRECHT, A. (eds) *Natural Stone, Weathering Phenomena, Conservation Strategies and Case Studies*. Geological Society, London, Special Publications, **205**, 25–41.
- CASSAR, J. 2004. Composition and property data of Malta's building stone for the construction of a database. In: PŘIKRYL, R. & SIEGL, P. (eds) *Architectural and Sculptural Stone in Cultural Landscape*. Karolinum Press, Prague, 11–28.
- CASSAR, J. & VANNUCCI, S. 2001. Petrographical and chemical research on the stone of the megalithic temples. *Malta Archaeological Review*, **5**, 40–45.
- CASSAR, J. & VELLA, A. J. 2003. Methodology to identify badly weathering limestone using geochemistry: case study on the Lower Globigerina Limestone of the Maltese Islands. *Quarterly Journal of Engineering Geology and Hydrogeology*, **36**, 85–96.
- FARRUGIA, P. 1993. *Porosity and related properties of local building stone*. B.E. & A. dissertation, University of Malta.
- FASSINA, V., MIGNUCCI, A., NACCARI, A., STEVAN, A., CASSAR, J. & TORPIANO, A. 1996. Investigation on the moisture and salt migration in the wall masonry and on the presence of salt efflorescences on stone surface in the church of Sta. Marija Ta' Cwerra at Siggiewi, Malta. In: ZEZZA, F. (ed.) *Origin, Mechanisms and Effects of Salt on Degradation of Monuments in Marine and Continental Environments*. Proceedings, European Commission Research Workshop on Protection

- and Conservation of the European Cultural Heritage, Bari, Italy. Research Report, **4**, 89–100.
- FITZNER, B. & KOWNATZKI, R. 1990. Bauwerkskartierung—Schadensaufnahme an Naturwerksteinen. *Der Freiberufliche Restaurator*, **4**, 25–40.
- FITZNER, B. & KOWNATZKI, R. 1991. *Klassifizierung der Verwitterungsformen und Kartierung von Natursteinbauwerken*. Jahresberichte aus dem BMFT-Forschungsprogramm Steinerfall – Steinkonservierung, **1**, 1–13.
- FITZNER, B., HEINRICHS, K. & KOWNATZKI, R. 1992. Classification and mapping of weathering forms. In: DELGADO RODRIGUES, J., HENRIQUES, F. & TELMO JEREMIAS, F. (eds) *Proceedings of the 7th International Congress on Deterioration and Conservation of Stone*, 15–18 June, Lisbon, Laboratório Nacional de Engenharia Civil, Lisbon, Portugal, 957–968.
- FITZNER, B., HEINRICHS, K. & KOWNATZKI, R. 1995. Weathering forms-classification and mapping. In: SNETHLAGE, R. (ed.) *Denkmalpflege und Naturwissenschaft, Natursteinkonservierung I*. Ernst and Sohn, Berlin, 41–88.
- FITZNER, B., HEINRICHS, K. & VOLKER, M. 1996. Model for salt weathering at Maltese Globigerina Limestones. In: ZEZZA, F. (ed.) *Origin, Mechanisms and Effects of Salt on Degradation of Monuments in Marine and Continental Environments*. Proceedings, European Commission Research Workshop on Protection and Conservation of the European Cultural Heritage, Bari, Italy. Research Report, **4**, 333–344.
- GALAN, E., AIRES-BARROS, L., CHRISTARAS, B., KASSOLI-FOURNARAKI, A., FITZNER, B. & ZEZZA, F. 1996. Representative stones from the Sanctuary of Demeter in Eleusis (Greece), Sta. Marija Ta' Cwerra of Siggiewi (Malta) and Bari (Italy) and Cadiz (Spain) Cathedrals: petrographic characteristics, physical properties and alteration products. In: ZEZZA, F. (ed.) *Origin, Mechanisms and Effects of Salt on Degradation of Monuments in Marine and Continental Environments*. Proceedings, European Commission Research Workshop on Protection and Conservation of the European Cultural Heritage, Bari, Italy. Research Report, **4**, 77–85.
- KOWNATZKI, R. 1997. *Verwitterungszustandserfassung von Natursteinbauten unter besonderer Berücksichtigung phänomenologischer Verfahren*. Aachener Geowissenschaftliche Beiträge, **22**.
- PEDLEY, M., CLARKE, M. H. & GALEA, P., 2002. *Limestone Isles in a Crystal Sea. The Geology of the Maltese Islands*. P.E.G., Malta.
- PRICE, C. & BRIMBLECOMBE, P. 1994. Preventing salt damage in porous materials. In: ROY, A. & SMITH, P. (eds) *Preprints of the Contributions to the Ottawa Congress, Preventive Conservation – Practice, Theory and Research*. IIC, London, 90–93.
- ROTHERT, E., 2004. *Gefügeabhängige Verwitterung von Kalksteinen durch Salzkristallisation im Porenraum*. MSc. thesis, Universität Göttingen.
- SNETHLAGE, R. 1984. *Steinkonservierung*. Bayerisches Landesamt für Denkmalpflege, Arbeitshefte, **22**.
- TORFS, K., VAN GRIEKEN, R. & CASSAR, J. 1996. Environmental effects on deterioration of monuments: case study of the Church of Sta. Marija Ta' Cwerra, Malta. In: ZEZZA, F. (ed.) *Origin, Mechanisms and Effects of Salt on Degradation of Monuments in Marine and Continental Environments*. Proceedings, European Commission Research Workshop on Protection and Conservation of the European Cultural Heritage, Bari, Italy. Research Report, **4**, 441–451.
- VANNUCCI, S., ALESSANDRINI, G., CASSAR, J., TAMPONE, G. & VANNUCCI, M. L. 1994. The prehistoric megalithic temples of the Maltese Islands: causes and processes of deterioration of Globigerina Limestone. In: FASSINA, V., OTT, H. & ZEZZA, F. (eds) *Conservation of Monuments in the Mediterranean Basin. Proceedings of the 3rd International Symposium, Venice, Italy, Soprintendenza di Beni Artistici e Storici di Venezia, Italy*, 555–565.
- VANNUCCI, S., CASSAR, J. & TAMPONE, G. 1985. The treatment of a typical soft limestone with different consolidants: a comparative study. In: BOLLETTINO INGEGNERI, *Florence, Italy, Collegio degli Ingegneri della Toscana, Florence, Italy*, 3–11.