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Evaluation of salt weathering in stone using a new test procedure: a comparative laboratory study

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Abstract. Exposure to natural weathering and deleterious agents poses a significant risk to traditional porous building materials, like stone and mortar, with salt crystallization featuring as a prominent degradation mechanism. Despite the existence of standardized laboratory tests to assess the salt crystallization resistance of stone, the international scientific community still seeks more reliable and reproducible alternatives. In this framework, RILEM TC ASC-271 recently recommended a novel accelerated test procedure for assessing the resistance of natural stone and fired-clay brick units against salt crystallization. This study assesses RILEM's recommendation by comparing results from laboratories in Cyprus and the UK on a particular lithotype. The interlaboratory results show similar mass losses, with higher values reported with an increase in the salt concentration used. Micro-destructive cutting tests adopted to locate the salt crystallization front at the end of the procedure revealed layers of increased resistance behind the evaporation surface of the test specimens because of pore clogging. The reproducibility of the results reported and the efficiency of the micro-destructive cutting techniques employed in mapping the salt front suggest that the new test recommendation may be used effectively in the lab to predict damage due to salt crystallization in natural stone.

Keywords: Salt crystallization, Natural stone, Micro-destructive tests, RILEM.

1 Introduction

Salt crystallization is among the most damaging weathering phenomena affecting building materials [1, 2], leading to profound and often detrimental effects on the structural integrity and aesthetic value of historic monuments. Throughout history, civilizations have grappled with the challenges posed by salt-induced weathering, as evidenced by the weathered facades of ancient structures and the gradual erosion of monumental sculptures.

The sources of soluble salts are diverse and ubiquitous, ranging from natural, such as saline soils, sea spray, and airborne pollutants, to anthropogenic activities and biological agents [3, 4]. Salts infiltrate building materials during construction or are deposited in them over time, remaining dormant until the right environmental conditions (temperature and relative humidity) trigger their activation. Upon activation, soluble salts may crystallize within the porous matrix of building materials, exerting significant pressure on pore walls [5] and leading to cracking, spalling, and overall deterioration [6, 7]. This process is exacerbated by cyclic wetting and drying, as well as fluctuations in temperature and humidity. The manifestation of salt crystallization, whether as visible efflorescence on material surfaces or concealed sub-florescence within pores, presents unique challenges for conservators and heritage professionals. Efflorescence, while often regarded as visually unsightly, is generally considered less harmful compared to sub-florescence, which can lead to pore clogging and irreversible damage. Early detection of sub-florescence is crucial for implementing timely conservation interventions and mitigating the long-term effects of salt crystallization.

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Despite the existence of standardized laboratory tests for the assessment of salt crystallization resistance of natural stones, researchers worldwide are continuously seeking reliable and reproducible alternatives. In this framework, RILEM TC ASC-271 recently recommended a novel accelerated test procedure for the assessment of the resistance of natural stone and fired-clay brick units against salt crystallization [8].

In this paper, the authors follow the aforementioned salt crystallization test protocol and carry out an inter-laboratory comparison using a specific lithotype, aiming to assess the efficacy of the recommended test procedure. Additionally, this study delves into innovative methodologies of micro-destructive testing to spatially map the progression of salt crystallization, thereby facilitating a more comprehensive examination of salt distribution within stone substrates. Through the advancement of knowledge regarding the dynamics of salt crystallization, the authors aim to contribute to the development of effective strategies for safeguarding heritage materials for future generations.

2 Materials and Methods

2.1 Materials and Sample Preparation

Lympia stone, extracted from the broader Lympia village region within the district of Larnaca in south-eastern Cyprus, was used in this study. XRD analysis confirms that the composition of this stone is almost exclusively calcitic, with minor traces of quartz. The stone exhibits a notable open porosity > 40%, a unimodal size distribution and a capillary absorption coefficient ca. 140.5 $g/m^2s^{1/2}$ [9].

The RILEM TC 271-ASC protocol adopted for sodium sulfate (Na_2SO_4) salt crystallization testing involved six core specimens (50±0.5 mm diameter and 50±0.5 mm height) for each laboratory; these were meticulously prepared in Cyprus with precision drilling techniques to minimize substrate damage. After drying and sealing to maintain their moisture content, the specimens in each lab underwent an accumulation phase, during which they absorbed Na_2SO_4 solution at different concentrations through capillary action, as shown in Table 1. This was followed by a propagation phase consisting of four cycles, each lasting three weeks. During each cycle, the specimens were subjected to specific environmental conditions, including temperature and humidity control. Damage assessment after each cycle involved detailed photographic documentation and categorization based on established atlases. Total material loss was quantified post-test, providing valuable insights into the impact of salt crystallization cycles.

Table 1. Summary of the specimens tested and the concentration of solution used.

Sample ID	Salt concentration solution	Laboratory setting
L1CY	5% Na_2SO_4	University of Cyprus, Cyprus
L2CY		
L3CY		
L4CY	10% Na_2SO_4	
L5CY		
L6CY		
L1UK	5% Na_2SO_4	Newcastle University, United Kingdom
L2UK		
L3UK		
L4UK	10% Na_2SO_4	
L5UK		
L6UK		

2.2 Micro-destructive assessment techniques

The scratch tool and the Drilling Resistance Measurement System (DRMS) were used to locate the salt crystallization front at the end of the test. The two aforementioned micro-destructive techniques follow the same principle of operation, determining the resistance of a material to either scratching or drilling;

the result in both cases is directly related to the mechanical properties of the test specimen. Both techniques had previously been used for the evaluation of a range of stone properties [10], and to provide key information on the assessment of stones in situ [11]. They have also been utilized for the evaluation of the mechanical properties of mortars [12, 13, 14], whilst they further seem to offer new paths for understanding salt distribution patterns and assessing the extent of salt-induced damage within porous materials [15, 16, 17].

The scratch tool operates by creating shallow grooves with incremental depth (d = 0.01-2 mm) on a specimen's surface, using a 10 mm wide polycrystalline diamond compact (PDC) cutter. During the test, both the normal (F_n) and tangential (F_t) components of the force acting on the cutter are recorded (see Equations (1) and (2)); ε signifies the intrinsic specific energy, and ζ denotes the ratio of vertical to horizontal force on the cutting face [18].

$$F_t = \varepsilon w d \tag{1}$$

$$F_n = \zeta \varepsilon w d \tag{2}$$

In this study, all specimens underwent an initial rectification through successive shallow grooves made by the machine’s PDC cutter to level their surfaces. Subsequently, five consecutive scratches of constant depth (d = 0.05 mm) were made on each specimen (Fig. 1a). The forces (F_n and F_t) recorded during these scratches were utilized to estimate the average total cutting force (F), which was then plotted against the PDC cutter’s position.

The Drilling Resistance Measurement System (DRMS) comprises a drilling machine and a computer for synchronous processing, visualization, and data storage. The system is equipped with a diamond drill bit and measures the drill position, the penetration force, and the rotational and penetration speeds.

In this study, measurements were conducted using a 5 mm diameter twist diamond drill bit with a rotational speed of 600 rpm, a penetration rate of 10 mm min^{-1}, and a depth of 10 mm. Three holes were drilled on each sample (Fig. 1b) to determine the average drilling resistance, reflecting the force reacting to the drill bit’s weight or thrust.

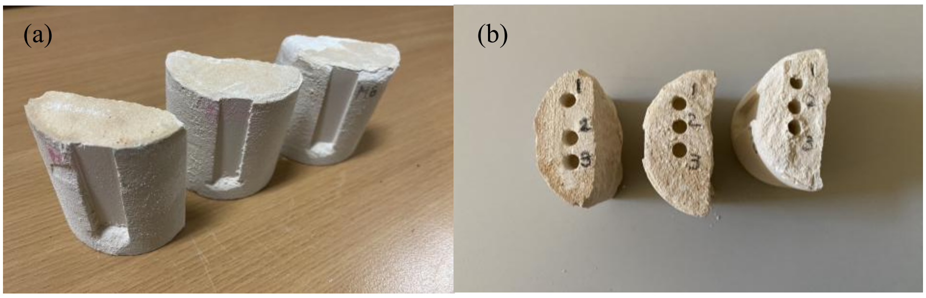

Fig. 1. Samples subjected to micro-destructive tests: (a) scratching groove; (b) drilling holes.

3 Results and Discussion

A summary of the macroscopic (visual) observations made during the test is provided in Table 2. Table 3 lists the damage type, severity and mass losses reported during this interlaboratory study.

The results indicate significant similarities in terms of visual inspection, damage type, severity, and mass losses. Moderate damage was reported for all specimens contaminated with the 5% salt solution; this was mainly evidenced through a powdery surface due to efflorescence. In contrast, severe damage occurred in the specimens contaminated with 10% Na_2SO_4; this appeared as loss of surface cohesion and bursting due to sub-florescence.

The comparable average mass losses reported in the two laboratories for samples treated with either 5% or 10% Na_2SO_4 salt solutions underscore the consistency and reliability of the experimental procedure adopted. The lab in Cyprus obtained an average mass loss of 0.72 g for samples treated with 5% w/w Na_2SO_4 salt, while the one in the UK obtained an average mass loss of 0.65 g for the same samples. Higher mass losses were reported for the samples treated with 10% w/w Na_2SO_4 salt, as expected: 3.70 g in the laboratory in Cyprus, compared to 3.81 g in the laboratory in the UK.

Table 2. Macroscopic (visual) observations on specimens contaminated with Na_2SO_4.

Sample ID	Reference	Accumulation	Propagation				End of test
			Phase 1	Phase 2	Phase 3	Phase 4	
L1CY							
L1UK							
L2CY							
L2UK							
L3CY							
L3UK							
L4CY							
L4UK							
L5CY							
L5UK							
L6CY							
L6UK							

Table 3. Damage type, severity of damage and material loss.

Sample ID	Damage type	Severity	Material loss (g)
L1CY	Efflorescence, Crust, Powdery	Medium	0.90
L1UK	Efflorescence, Crust, Powdery	Medium	0.69
L2CY	Efflorescence, Crust, Powdery	Medium	0.66
L2UK	Efflorescence, Crust, Powdery	Medium	0.65
L3CY	Efflorescence, Crust, Powdery	Medium	0.60
L3UK	Efflorescence, Crust, Powdery	Medium	0.62
L4CY	Bursting with sub-florescence	High	3.43
L4UK	Loss of surface cohesion with sub-florescence	High	4.01
L5CY	Bursting with sub-florescence	High	4.22
L5UK	Loss of surface cohesion with sub-florescence	High	3.68
L6CY	Bursting with sub-florescence	High	3.45
L6UK	Loss of surface cohesion with sub-florescence	High	3.73

The cutting resistance patterns depicted in Fig. 2 and Fig. 3, arising from the scratch tool and the DRMS, were utilized to identify alterations, such as pore clogging, in the microstructure of the salt-contaminated samples compared to the reference ones. Following salt contamination, a prominent peak was observed with the scratch tool ca. 0-2 mm behind the evaporation surface of the specimen contaminated with 10% salt solution. Albeit such a peak was not evident in the case of the sample contaminated with the 5% salt solution, the intensity of the overall scratching resistance was increased in both groups of contaminated samples (compared to the reference).

The cutting resistance peaks observed in the DRMS tests demonstrated peaks ca. 0-1 mm behind the evaporation surface of both groups of salt-contaminated samples, irrespective of the salt solution concentration used. These were more pronounced in the samples contaminated with the 10% salt solution. Since such peaks were absent in the reference samples, one may safely assume that they are attributed to pore clogging caused by the accumulation of salt crystals behind the evaporation surface of the salt-contaminated samples.

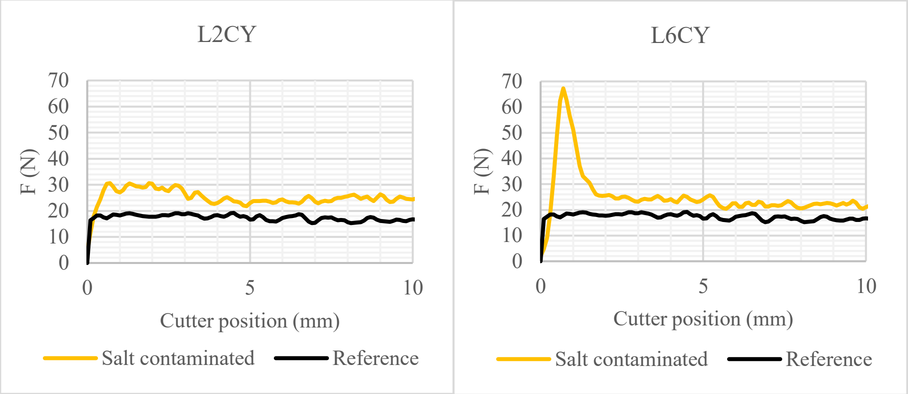

Fig. 2. Average scratch tool patterns for indicative salt-contaminated and reference samples (refer to Table 1 for sample code and description).

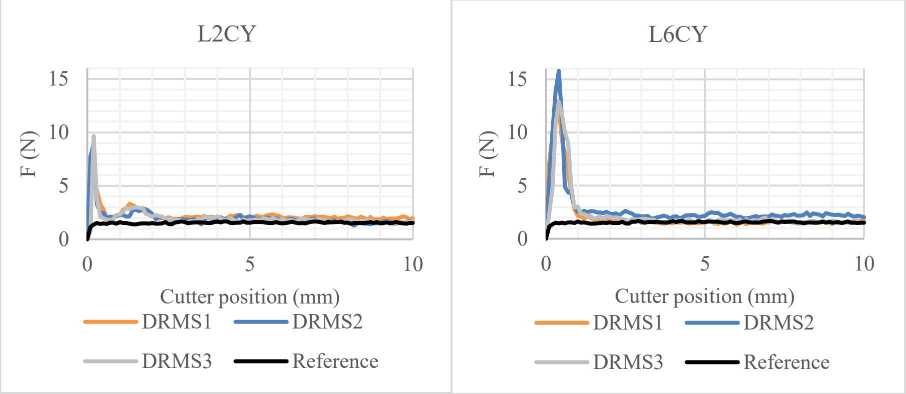

Fig. 3. DRMS patterns for indicative salt-contaminated and reference samples (refer to Table 1 for sample code and description).

4 Conclusions

This research focused on evaluating the salt crystallization test protocol recommended by RILEM TC-ASC-271, through interlaboratory comparison of results on a specific lithotype. The findings suggest notable similarities in the visual inspection and the mass losses reported by both laboratories after the test, thus confirming the reliability and reproducibility of the new test method.

Furthermore, this research concentrated on the use of two micro-destructive cutting techniques for the analysis of salt distribution in stone. These revealed the presence of significant peaks in cutting resistance ca. 0–2 mm behind the evaporation surface of the salt-contaminated specimens. Such peaks were absent in the reference samples and may thus be attributed to the tendency of Na_2SO_4 to crystallize near the evaporation surface of limestones, essentially leading to pore clogging. The versatility and efficacy of both micro-destructive techniques hereby used in assessing salt distribution in limestones render them valuable tools for preventive conservation. Especially, the DRMS could be employed on-site in monuments and historic structures to identify pore clogging before it escalates, thus serving as an early warning system for conservators to implement remedial actions and prevent damage from sub-florescence in masonry materials.

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