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**Publication date**

2021

**Document Version**

Final published version

**Published in**

Proceedings of the SWBSS

**Citation (APA)**

Nunes, C., Godts, S., Maria Aguilar Sanchez, A., Slížková, Z., & Lubelli, B. (2021). Towards a new salt crystallisation test: Comparison of salt contamination procedure. In B. Lubelli, A. Kamat, & W. Quist (Eds.), *Proceedings of the SWBSS: Fifth International Conference on Salt Weathering of Buildings and Stone Sculptures* (pp. 69-78). TU Delft OPEN Publishing.

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# TOWARDS A NEW SALT CRYSTALLISATION TEST: COMPARISON OF SALT CONTAMINATION PROCEDURES

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## KEYWORDS

Crystallisation test, porous materials, salt accumulation, evaporative surface

## ABSTRACT

The RILEM TC 271-ASC is currently developing a new ageing test to assess the resistance of porous building materials to salt crystallisation. The new test consists of two phases: salt accumulation and damage propagation. This paper focuses on the salt accumulation phase; this phase should promote salt crystallisation close to the evaporative surface of the substrates (common situation onsite) without leading to salt efflorescence or damage. Damage should take place in the propagation phase, which is not addressed in this paper. This work compares the effectiveness of three contamination procedures for salt accumulation: P1) salt contamination by capillary absorption of salt solution, followed by drying; P2) continuous capillary absorption of salt solution; P3) placement of salt crystals on the surface of the materials followed by the conditioning of the specimens at high relative humidity until complete dissolution of the salts. The results of P1 and P2 procedures presented in this paper are detailed in a separate publication, while this paper focuses on P3 procedure and compares the results. The effectiveness of each procedure has been evaluated by assessing the salt distribution in the specimen using ion chromatography and scanning electron microscopy. The results show that P3 is technically the least complicated to set up and does not entail the risk of development of salt efflorescence or damage. However, it can lead to salt migration within the specimens to a greater depth, hence future research is proposed to counteract this effect.

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# 1 INTRODUCTION

Salt crystallisation is one of the most critical deterioration processes observed in porous materials due to its ubiquitous presence worldwide and the complexity of the caused damage phenomena. These aspects hinder the development of effective approaches for counteracting its deleterious action and for repairing existing objects. The lack of a practical and realistic test to evaluate the resistance of porous materials to salt crystallization has motivated the launch of the RILEM Technical Committee 271-ASC (TC-ASC): Accelerated laboratory test for the assessment of the durability of materials with respect to salt crystallization [1].

A recent literature review on the subject [2] has identified a large variety of tests performed by researchers, mainly due to the limitations of existing standardized tests. A crucial issue in salt crystallization tests relates to targeting accumulating salts close to the evaporative surface of the materials [3]. In fact, salt accumulation in a thin layer close to the material's surface accelerates the occurrence of damage because it leads to a high degree of pore filling, thus leading to high crystallization pressure that onsets damage [4]. Besides, salts that have crystallized close to the evaporative surface respond faster to relative humidity and temperature changes, so they may undergo more frequent dissolution-crystallization cycles, accelerating the damage process and reproducing better the situation onsite.

Hence, the TC-ASC salt crystallisation test under development encompasses two phases: (1) salt accumulation at the evaporative surface of the materials and (2) salt propagation phase entailing cycles of dissolution and crystallisation. This work solely addresses the (1) salt contamination phase by investigating the capability of different salt contamination procedures in accumulating salts in a thin surface layer of the specimens' evaporative surface. The propagation of damage by cycles of dissolution-crystallization is not encompassed in this study.

Three contamination procedures aiming at accumulating salts close to the evaporative surface of porous building materials are compared: P1) salt contamination by capillary absorption of a salt solution followed by drying; P2) continuous capillary absorption of a salt solution; P3) placement of salt crystals on the surface of the materials followed by the conditioning of the specimens at high relative humidity until complete dissolution of the salts. This paper focuses on the analysis of the results of P3 procedure and compares it with those obtained with P1 and P2; the latter two procedures are comprehensively reported in [5].

## 2 EXPERIMENTAL PART

### 2.1 Materials

Two natural stones (Migné and Maastricht limestones) with similar composition (ca. 97% calcite), high homogeneity, but very different moisture transport properties were selected for this study to facilitate the comparison of the results. Migné stone has an open porosity of ca. 28% and a unimodal pore size distribution around 0.5-2  $\mu\text{m}$ . Maastricht stone has 51% porosity and a unimodal pore size distribution centred around 45 $\mu\text{m}$  [6]. A comprehensive characterization of these stones can be found in D'Altri et al. [6]. Core specimens of 50 mm diameter and 50 mm height

were used. All the specimens were sealed on the lateral sides using parafilm. The tests were carried out with three replicates per stone type and procedure.

## 2.2. Salt contamination procedures

Three contamination procedures were designed: P1) salt contamination by capillary absorption of the salt solution followed by drying; P2) continuous capillary absorption of salt solution; P3) placement of salt crystals on the surface of the materials followed by the conditioning of the specimens at high relative humidity (RH) until complete dissolution of the salts (Figure 1).

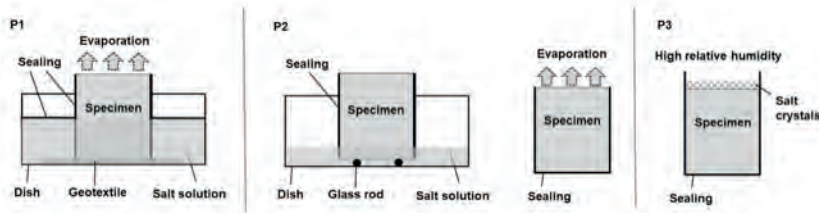


Figure 1: Schematic representation of the procedures used in the study: P1) Salt contamination followed by drying; P2) Continuous capillary absorption; P3) Salt crystals on the top of the specimen and dissolution with high RH.

In the three procedures, the specimens were contaminated with either sodium chloride or sodium sulphate; both NaCl and Na<sub>2</sub>SO<sub>4</sub> were ACS reagents  $\geq 99.0\%$  (Fluka) with similar grain size (NaCl: 71%-0.25mm, 13%-0.5mm, 12%-0.125mm; Na<sub>2</sub>SO<sub>4</sub>: 83%-0.25mm, 13%-0.5mm, 4%-0.125mm). Salt content of 1wt.% with respect to the dry specimens' mass was chosen as it is considered a realistic amount of salt in the outer layer (first centimetres) of salt damaged building materials onsite [7]. Moreover, using the same salt content enables an easier comparison of the procedures. Considering the stone density and the cylindrical specimens' volume, 1wt.% corresponds to 1.91g of salt in Migné and 1.37g in Maastricht. Following is a detailed description of the procedures.

### *P1) Salt contamination by capillary absorption followed by drying*

Capillary absorption of salt solution through the specimen's bottom surface followed by drying at 20°C and 10% RH. The salt solution concentration is calculated to contaminate the specimen with 1wt.% of salt with respect to the specimen dry weight. After contamination, the specimen's bottom is sealed with parafilm, and the specimens are dried until 80% of the water evaporates.

### *P2) Continuous capillary absorption*

Continuous capillary absorption by immersion of the bottom of the specimen in salt solution and simultaneous drying through the top surface at 20°C and 10% RH forms this procedure. The amount of water used in the preparation of the solution is twice higher than that used in procedure P1. Therefore, the salt solution concentration is lower than in P1, but the final amount of salt in the specimen is still equal to 1wt.% of the dry specimens' mass. Evaporation of the solution from the contamination vessel during the test is prevented by covering the vessel's surface with

parafilm. After contamination, the specimen's bottom is sealed with parafilm and dried until 80wt.% of the water evaporates.

### *P3) Salt crystals placed on the top of the specimen and dissolution with high RH*

The defined amount of salt needed to contaminate the two stones is placed and evenly distributed on the specimens' top (contamination surface). Before placing the salt crystals on the specimens, the specimens are conditioned at 95% RH and 35°C (dissolution conditions) until their weight is stabilized. Afterwards, the specimens with salt crystals on their top surface are again conditioned at 95% RH and 35°C, and the dissolution of the salts is monitored daily with the naked eye until complete dissolution.

## **2.3. Assessment of salt distribution**

After salt contamination, as described previously, the specimens were weighed and dried at 60°C to avoid salt migration to the specimens' depth before analysing their distribution. Salt efflorescence, when present, was collected with a soft brush, and its mass was determined. The specimens were subsequently split into two halves with a hammer and chisel, and one half analysed with ion chromatography (IC) and the other half with scanning electron microscopy (SEM).

For the analysis of the NaCl or Na<sub>2</sub>SO<sub>4</sub> content with IC (Metrohm), measurements were carried out on powder samples drilled at two locations of each sample up to the following depths (same hole): 0-5, 5-10, 10-15, 15-20, 20-30, 30-40, and 40-50 mm. The salt amount present in each layer was calculated to the percentage of the total salt content in the dry sample. The average amount of each ion (in wt.%) of three specimens per contamination procedure was calculated to the average relative percentage of the total salt content.

SEM observations were carried out on thin sections produced from 2x2 cm pieces of the stones. The samples were embedded under vacuum in low viscosity epoxy resin and, after polymerization, ground with silicon carbide grinding paper. The final polishing was carried out with water-free diamond polishing suspensions. The polished surfaces were coated with a 15 nm carbon layer, then observed under SEM (FEI QUANTA 200 3D), high vacuum, and 20 kV voltage.

## **3 RESULTS AND DISCUSSION**

In procedure P3, the salt crystals were evenly distributed on the specimens' surface (Figure 2.a). However, after almost all salt dissolved, a few patches of salt crystals remained on the surface of both stones, as shown in Figures 2.b) and c), probably due to an uneven distribution of the salt crystals or irregularities in the stone. In general, it took one more day for the complete dissolution of the remnant salt patches.

Figure 3 shows the specimens at the end of the test with procedures P1 and P2. In general, salt efflorescence developed in both stones with both procedures and both salts. In the case of contamination with Na<sub>2</sub>SO<sub>4</sub>, slight damage occurred in Migné with procedure P1 and in Maastricht with procedure P2

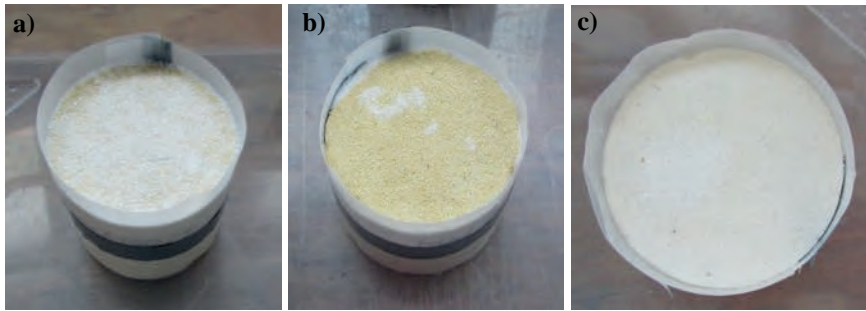


Figure 2: Specimens contaminated with procedure P3: a)  $\text{Na}_2\text{SO}_4$  crystals on the top of Maastricht surface before starting dissolution at high RH; b) Surface of Maastricht with  $\text{Na}_2\text{SO}_4$  crystal patches on the top one day before complete dissolution; c) Surface of Migné with  $\text{NaCl}$  crystals on the top crystal patches on the top one day before complete dissolution.

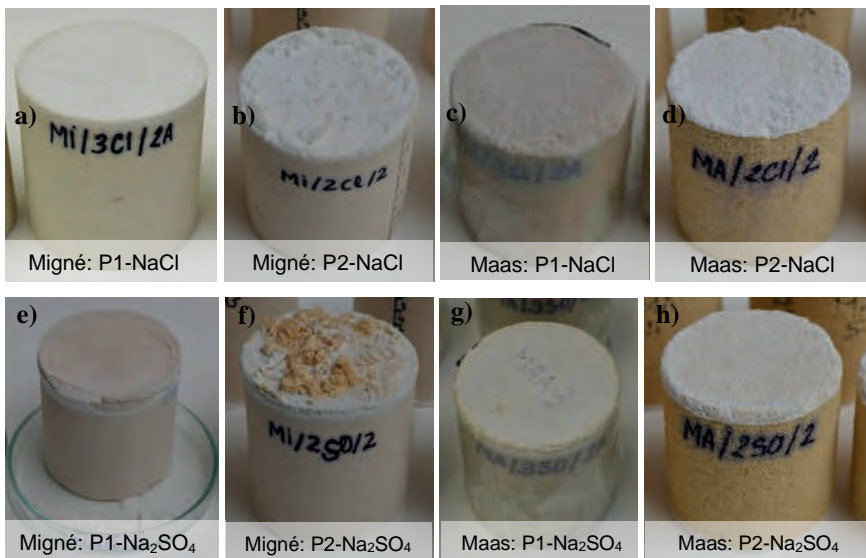


Figure 3: Specimens after salt contamination with procedures P1 and P2.

The time for the complete dissolution of the crystals in each substrate and type of salt with procedure P3 is shown in Table 1. The complete dissolution of the salt crystals placed on the less porous stone's surface (Migné) took one day longer compared to the more porous stone (Maastricht). This is linked to the fact that Migné is contaminated with a higher amount of salt to achieve 1 wt.% of salt in the specimens. Besides, Maastricht stone is slightly more hygroscopic than Migné: during weight stabilization of the stones at the thermohygro-metric dissolution conditions of the test ( $35^\circ\text{C}$ , 95%) before commencing the test, Maastricht absorbs twice the amount of water compared to the Migné (0.2 wt.%-Maastricht and 0.1 wt.%-Migné), which, among others, can facilitate a faster dissolution of the salt.

Sample	Salt	$t_{\text{diss}}$ (days)
Migné	NaCl	4
Maastricht	NaCl	3
Migné	Na <sub>2</sub> SO <sub>4</sub>	7
Maastricht	Na <sub>2</sub> SO <sub>4</sub>	6

Table 1: Time for the complete dissolution ( $t_{\text{diss}}$ ) of the salt crystals placed on the top of the stone specimens according to procedure P3 ( $\pm$  standard deviation): Migné: 1.91g of salt, Maastricht: 1.37g of salt.

After the complete dissolution of the salt with procedure P3, the stone surface was observed under a microscope; Figure 4 shows the stones' surface after the complete macroscopic dissolution of the salt crystals. Even though salt crystal patches were observed on the surface of both stones (Figure 2), only in Migné stone contaminated with NaCl, salt patches of melted salt were observed on the top of the specimens (Figure 4.a) corresponding with the areas where crystal patches remained before complete dissolution (Figure 2.c).

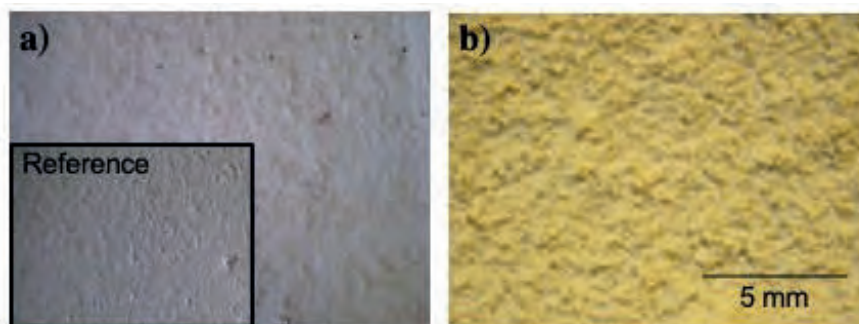


Figure 4: The surface of specimens under the binocular microscope after salt dissolution with procedure P3: a) Migné contaminated with NaCl showing a salt layer corresponding to a patch of ca. 1cm<sup>2</sup>; b) Maastricht contaminated with NaCl with no detectable salt on the top surface.

The salt distribution assessed with IC is given in Figure 5. In general, procedure P3 leads to salt spreading within both stones' matrix with both salts. In the case of NaCl, a high salt content was registered at the opposite surface of contamination, which in the case of Migné is even higher (ca. 10% ) than at the contamination surface. The NaCl melted layer observed on the top of Migné stone under the binocular microscope (Figure 4.a) is not evident in the IC results (Figure 5.a), probably because the salt is concentrated in the first millimetre of the substrate. In procedure P3, there is no risk of the development of salt efflorescence or damage.

The procedure P1 was the most effective in accumulating NaCl in a thin layer beneath the stones' evaporative surface without causing too much salt efflorescence or damage. Procedure P2 with Na<sub>2</sub>SO<sub>4</sub> was slightly more effective than P1 in accumulating the salt close to the evaporative surface of Migné but also caused a

substantial amount of salt efflorescence (ca. 60%). In the case of Maastricht, P2 procedure was more effective in accumulating salts at the surface, but damage was observed after brushing the efflorescence.

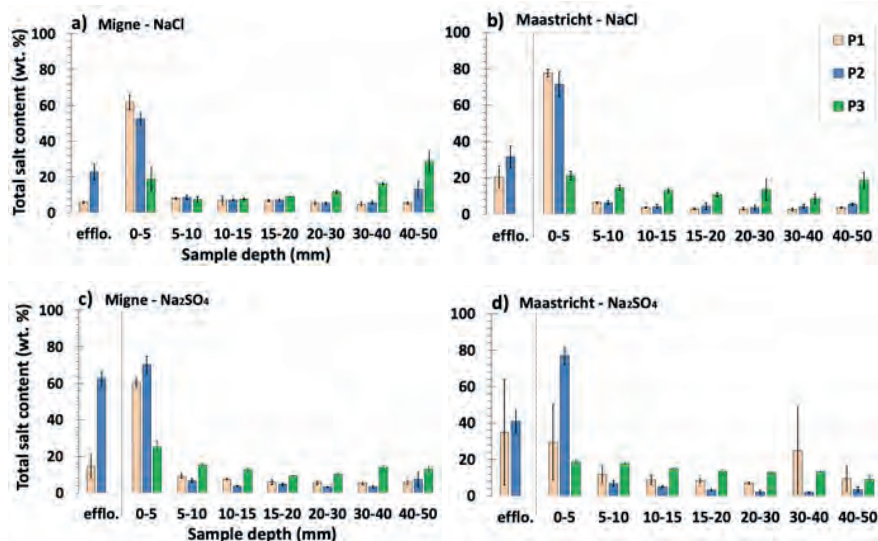


Figure 5: Salt content in the specimens' depth determined with IC: a) Migné contaminated with NaCl; b) Maastricht contaminated with NaCl; c) Migné contaminated with Na<sub>2</sub>SO<sub>4</sub>; d) Maastricht contaminated with Na<sub>2</sub>SO<sub>4</sub>. Note: *efflo.* corresponds to the weight of salt efflorescence collected in respect to the amount of salt introduced in the specimens.

SEM observations of the thin sections of the stones subjected to procedure P3 allowed detecting a continuous layer of NaCl on the top of Migné stone's surface (Figure 6.a), which is in line with the observations performed under the binocular microscope (Figure 4.a). SEM microphotographs also showed NaCl scattered within the matrix of Migné (Figure 6.b), in accordance with IC results. In the case of contamination with Na<sub>2</sub>SO<sub>4</sub>, no salt layer was observed on the top of Migné but somewhat scattered within the matrix. Similar results were obtained in Maastricht stone with Na<sub>2</sub>SO<sub>4</sub>, i.e., no salt layer was observed at the top surface but rather dispersed in the matrix (Figure 6.c and d). Due to a technical problem, it was impossible to analyse Maastricht stone contaminated with NaCl under SEM.



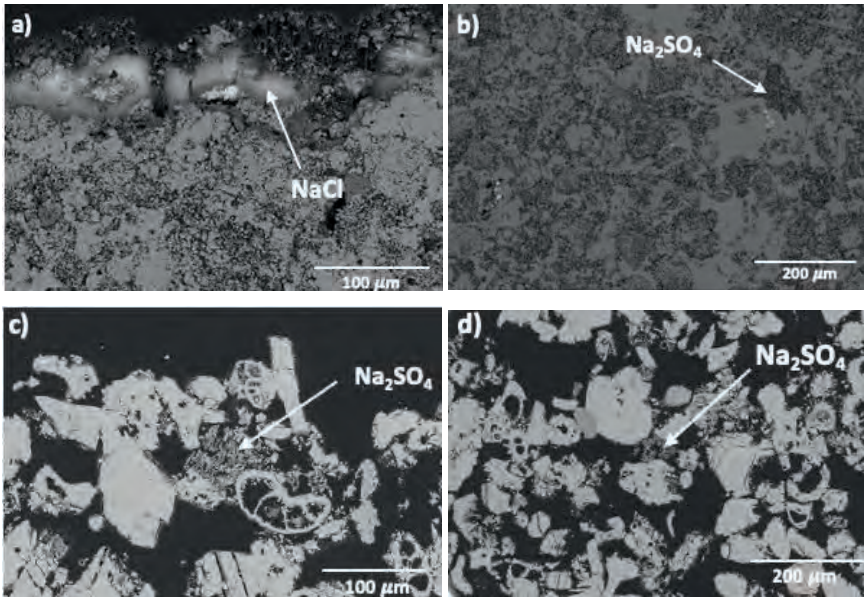


Figure 6: SEM microphotographs of specimens contaminated with procedure P3: a) layer of NaCl on the top of Migné; b) patches of  $\text{Na}_2\text{SO}_4$  detected within the matrix of Migné; c) patches of  $\text{Na}_2\text{SO}_4$  on the top of Maastricht; d) patches of  $\text{Na}_2\text{SO}_4$  scattered in the matrix of Maastricht.

## 4 CONCLUSIONS

Procedure P3 is fast and easy to perform and does not entail the risk of the development of salt efflorescence or damage. However, it can lead to the migration of a substantial amount of salt in the specimens' depth. The migration of salt deep into the matrix of the substrates is to be assigned to a non-perfect salt distribution of the salt crystals on their top surface, thus leading to salt patches subsequently extending the time for complete dissolution.

An even distribution of salt crystals on the top of the substrates may avoid remnant patches during the dissolution phase, limiting the time for complete dissolution, hence the migration of salts in the depth of the specimens. Therefore, further tests should be performed in the future, focusing on ensuring a perfect distribution of the salt crystals on the specimens' surface. The effect of the size of salt crystals should be studied to facilitate the homogeneous distribution of the salt on the top and speed up the dissolution.

The results indicate that the procedures using a salt solution to contaminate the specimens (procedures P1 and P2) are far more effective in accumulating salts close to the evaporative surface of the specimens than placing salt crystals on the top followed by dissolution at high relative humidity (P3). However, the procedures using a salt solution have the risk of developing salt efflorescence and damage.

## ACKNOWLEDGEMENTS

All authors are thankful to all the other RILEM TC ASC-271 members for their contribution to discussions. Z. Slížková acknowledges support from the Czech Academy of Sciences under program "Strategy AV21 – 23. City as a Laboratory of Change; Construction, Historical Heritage and Place for Safe and Quality Life".

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