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# Sodium chloride crystallization in a “salt transporting” restoration plaster

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## Abstract

In recent years several types of restoration plasters, especially developed for salt loaded substrates, have been introduced on the market. The behaviour of these plasters in the field is, however, not always satisfactory. The reasons for failures may be found both in the moisture and salt transport behaviour of the substrate/plaster combination and in the severity of the situation (salt load, moisture supply and environmental conditions).

In the present paper a restoration plaster for salt loaded substrates is examined. Its properties and composition are investigated in laboratory by means of multiple techniques (MIP, PFM, ESEM, XRD and FTIR). Particular attention is given to the study of the salt (NaCl) crystallization in the plaster. Field situations are considered in which this plaster shows damage after a few years from its application. The knowledge of the specific situation, together with the information gained in the laboratory research on the plaster, explains the fast development of the decay.

*Keywords:* Characterization; SEM; Durability; Physical properties; Restoration plaster

## 1. Introduction

Salt crystallization in porous materials constitutes one of the most frequent causes of decay of buildings, in a wide range of environments [1,2]. Among salts commonly found in walls, sodium chloride (NaCl) is one of the most abundant and ubiquitous. In spite of the several theories developed to explain salt crystallization damage, no unanimous opinion exists yet on the mechanism(s) causing the damage [3,4]. The most accepted theories explain the damage as the results of the pressure produced on the pore walls of the material by the crystallization of salt filling the pores [among others 5–7]. Damage occurs when the pressure produced by salt crystallization exceeds the mechanical strength of the material. The pressure that can be developed depends on the pore size in which crystallization occurs and on the supersaturation ratio, being these two factors strictly interrelated [7]. Since at equilibrium high crystallization pressures can be developed only in very small pores [8,9], a non-equilibrium situation has been supposed in order to explain the damage in building materials in which only large pores are present [10–12].

Salt can be transported in a porous material only if dissolved in water. During drying of a wall salt is transported to the evaporation front, where it crystallizes and accumulates. Generally the evaporation front is located near the surface of the wall, with as a consequence that the wall finish, i.e. the plaster, runs the highest risk of decay. Even though the plaster applied in restoration is usually considered as a sacrificial layer, a long service life of the plaster is pursued. In the last decades, apart from traditional plasters, ready-to-use restoration plasters, especially developed for salt loaded substrates, are available on the market. These plasters work either by accumulating the salts in the inner layer, (“salt accumulating” plasters, usually containing a water repellent in the outer layer) where enough space should be provided for their crystallization, or by transporting them to the surface, where they should crystallize in the form of harmless efflorescence (“salt transporting” plaster). Plasters that completely block salt (“salt blocking”) or water and water vapour (“moisture sealing”) from penetrating in the plaster layer are less common and usually not recommended in restoration [13].

The behaviour of restoration plasters in the field is not always satisfactory. A better knowledge of the properties of the

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plaster (composition, physical properties and moisture transport behaviour) together with a sound awareness of the field situation (plaster/substrate combination, moisture and salt load, environmental conditions) may help in avoiding unsuccessful applications in practice.

In this paper a ready-to-use restoration plaster for salt loaded substrates, M60 by Jahn, is studied in detail. According to the producer's description, this plaster does not limit moisture and salt solution transport (i.e. can be classified as "salt transporting" plaster). The composition, the physical properties and the moisture and salt solution transport behaviour of this restoration plaster have been investigated in laboratory. The location of salt (NaCl) crystallization in the plaster has been studied by means of Mercury Intrusion Porosimetry (MIP) and Environmental Scanning Electron Microscope (ESEM). The results obtained from the laboratory research have been related to the salt crystallization damage observed in the field.

## 2. Laboratory investigation on the plaster

### 2.1. Composition and physical properties

The composition and the physical properties (porosity and pore size distribution) of the plaster have been determined on specimens prepared in laboratory. Since the physical properties of the mortar are strongly influenced by the suction of the substrate on which it is applied [14], the plaster slabs were not prepared in a mould but on top of a brick substrate and then detached after one day. A filter paper was used between substrate and plaster to facilitate the detachment.

The composition of the plaster has been investigated by means of several techniques. Thin sections have been prepared to be studied by means of Polarized and Fluorescent Microscopy (PFM). This technique allows to investigate plaster composition, coarse porosity, voids and also decay in the material. Fig. 1 is a photomicrograph showing the presence of large voids between the cement binder and the aggregate. Two types of aggregate can be distinguished: a siliceous aggregate (in minor percentage) and a round hollow aggregate. This aggregate, as shown also by means of an Environmental Scanning Electron Microscope (ESEM, Philips XL-FEG SEM equipped with Secondary Electron and Backscatter Electron detectors) (Figs. 2, 3) seems not connected to the porous network, and represents the closed porosity of the plaster. The hollow aggregate, as shown by XRD analyses, is constituted by mullite ($Al_4Si_2O_{10}$).

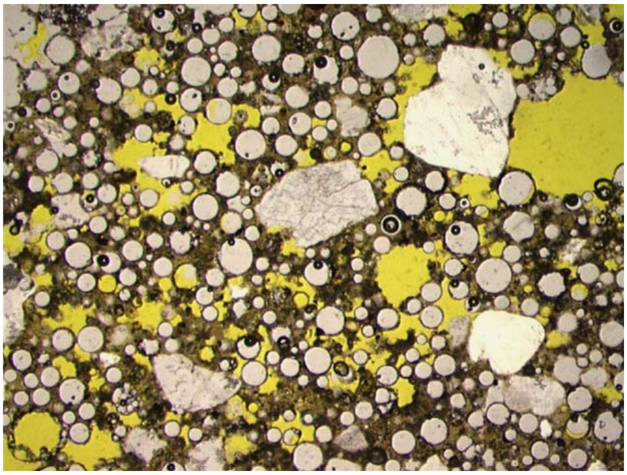

Fig. 1. PFM photomicrograph of a thin section of the restoration plaster (25×, picture size 5.4×3.5 cm).

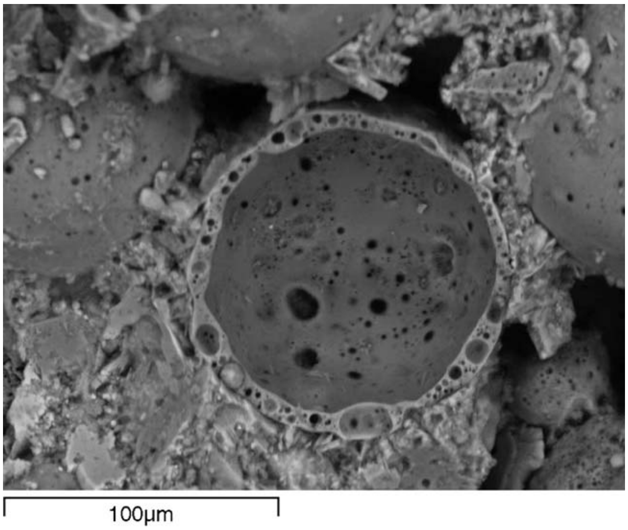

Fig. 2. ESEM photomicrograph of the hollow aggregate.

The presence of an additive, most probably modified cellulose, has been identified by Fourier Transformed Infrared Spectroscopy (FTIR). This additive has been probably mixed in the plaster to improve the curing process by retaining water and limiting shrinkage and crack formation during drying.

The porosity and pore size distribution have been determined. The following measurements have been performed:

- open porosity and pore size distribution by Mercury Intrusion Porosimetry (MIP) (Fig. 4)
- open porosity and density measured by immersion on 5×5×2 cm specimens
- closed and open porosity by point counting

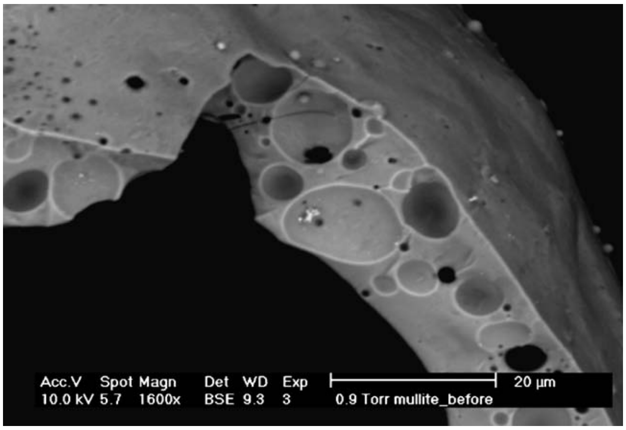

Fig. 3. ESEM photomicrograph of the walls of the hollow aggregate.

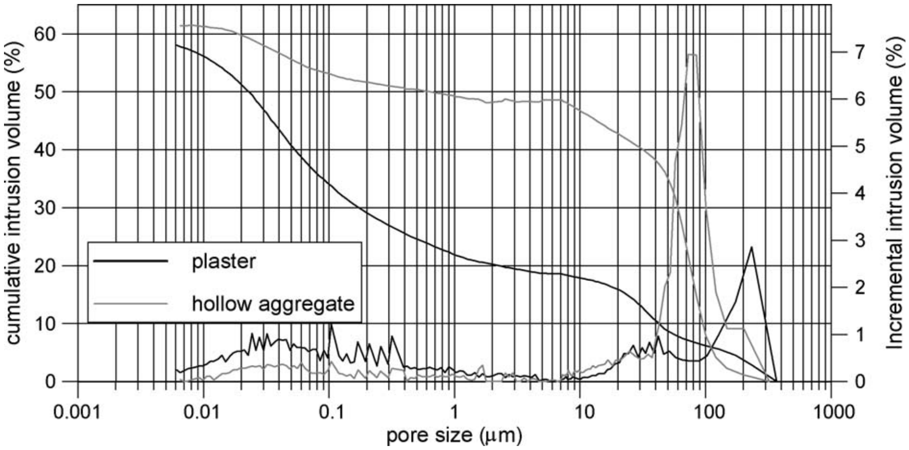

Fig. 4. Pore size distribution of the restoration plaster and of the hollow aggregate as measured by MIP.

The open porosity of the restoration plaster measured by MIP turned out to be 58% V/V. The porosity is mainly constituted by large (> 10 μm) and by very small pores in the range of capillary condensation (< 0.1 μm). Some questions arise when considering the very high value of porosity: either damage (the hollow spheres may be broken due to the pressure developed in the MIP) or deformation of the structure can be supposed. Both explanations are likely to show up in the cumulative intrusion curve as a steep increase in the range of the smaller pores. To further check these results, MIP measurements were performed on the hollow aggregate, after separating it from the binder. Again a very high porosity was registered (Fig. 4), probably due to breaking and filling of the hollow spheres. The fine pores present on the mullite walls (Fig. 3) may also contribute to the amount of fine porosity. From the MIP measurements it can be concluded that only part of the fine porosity measured in the plaster is due to the binder; the rest is most probably due to the mullite spheres.

The porosity of the plaster measured by immersion under vacuum according to the RILEM Recommendation CPC 11.3 [15] gave similar results (59% V/V). Also in this case the fact that part of the mullite spheres becomes accessible to water is supposed to explain the high values registered.

Subsequently, point counting measurements have been performed on the thin section. These measurements have allowed defining the closed porosity (constituted by the hollow spheres) and the amount of voids and pores (> 5 μm). The closed porosity represents 28% of the area; voids and pores larger than 1 μm amount to about 19% of the surface. This last value is comparable to the porosity obtained by MIP when considering only pores larger than 5 μm.

### 2.2. Moisture and salt solution transport

The absorption and drying behaviour have been determined on 5 × 5 × 2 cm specimens sealed on 4 lateral sides with epoxy resin. The absorption behaviour and the Water Absorption Coefficient (WAC) have been determined according to the European Standard prEN 1015-18 [16]. The drying has been measured at 20 °C 50% RH after saturating the specimens at atmospheric pressure and sealing them on 5 sides.

The pore size distribution of the material strongly influences its absorbing and drying behaviour: because of the presence of small pores, the plaster shows a slow absorption and drying (Fig. 5). Besides, the presence of cellulose, that can retain water even in secondary moistening, further slows the drying. It is

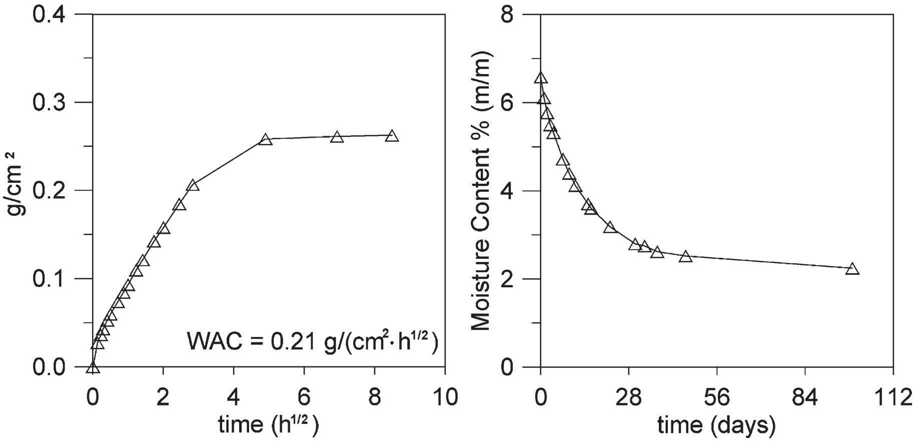

Fig. 5. Absorption (left) and drying (right) behaviour of the restoration plaster.

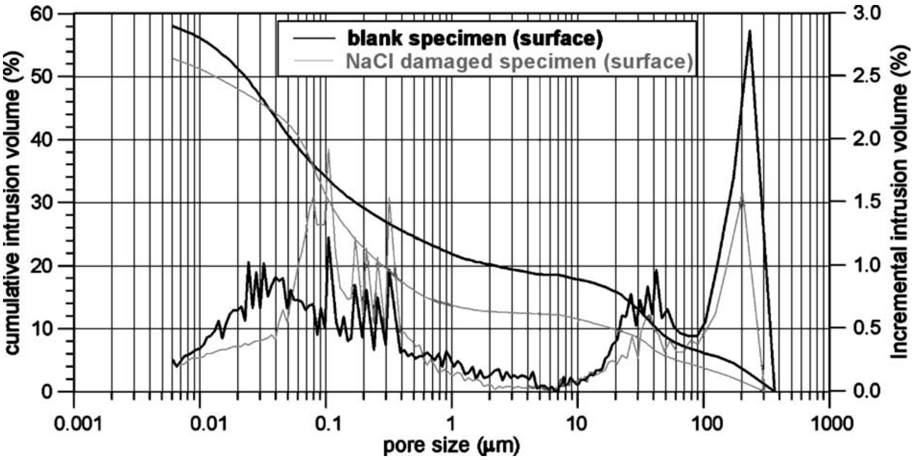

Fig. 6. Pore size distribution of the restoration plaster, before and after contamination and crystallization, as determined by mercury intrusion porosimetry.

interesting to notice that the results obtained were not expected for this plaster: being proposed as a plaster with open and porous structure, a more effective moisture transport was foreseen.

The moisture and salt solution transport were studied by wetting the plaster specimens with a defined amount of water and NaCl solution (22% by weight) and let them dry at different environmental conditions. At the end of the drying the salt contaminated specimens were dry sliced and the salt distribution was measured. The detailed results of this study are reported elsewhere [17]. Here it is worth to mention, in order to stress the extremely slow drying, that:

- as expected, the already slow drying of the plaster is further slowed down by the presence the hygroscopic NaCl
- the moisture content in the plaster after 4 months of drying at 20 °C 50% RH is 2.3% by weight in case of the reference sample and more than double in the NaCl contaminated sample.

### 2.3. Location of salt crystallization

Salt (NaCl) crystallization tests were performed in laboratory at different environmental conditions. The detailed results are reported elsewhere [17]. In this paper particular attention is given to the MIP and ESEM investigations. These measurements aimed at the assessment of the preferential location of the salt in the pores of the plaster and at the study of the effect of the crystallization on the structural integrity of the material.

In order to study the location of salt crystallization in pores, the pore size distribution before and after crystallization was measured by MIP on specimens dried at 60 °C 0% RH. The measurements were performed on 2 mm layers sampled from the surface of the 2 cm thick plaster specimens, before and after NaCl crystallization. The presence of salts in the sampled layer was first checked by ESEM.

The pore size distributions obtained are shown in Fig. 6: a decrease in the percentage of pores smaller than 0.05 µm and larger than 0.5 µm as well as an increase in the percentage of pores in the 0.05–0.1 µm range is measured after crystallization. How this reduction of porosity can be explained is not clear: it may be due to an actual filling of the pores, to salt crystallizing at the pore entry or to a reduced deformability of the plaster to the pressure exerted by the mercury due to the presence of the salt.

The location of salt crystallization in pores, i.e. the pore size in which the salt had preferentially crystallized, was investigated also by means of ESEM. Broken cross sections as well as the evaporation surface of plaster specimens contaminated with NaCl were observed by means of a Back-Scattered Electron (BSE) detector. The chemical composition of the crystals was checked by means of an Energy Dispersive Spectroscopy X-ray Microanalysis. From the several observations it can be concluded that the salts crystallize in the binder structure, accumulating mainly at the interface with the aggregate (Fig. 7). No salt has been detected inside the hollow aggregate confirming that this does not take part in the moisture transport. The mullite spheres are therefore not meant for salt accumulation, as it often happens for lightweight aggregate used in restoration plasters. It was not possible to detect salt in small pores, also because of the limited resolution due to the use of broken, not polished, sections; most of the visible pores (larger than 0.5–1 µm) appear to be empty (Fig. 8).

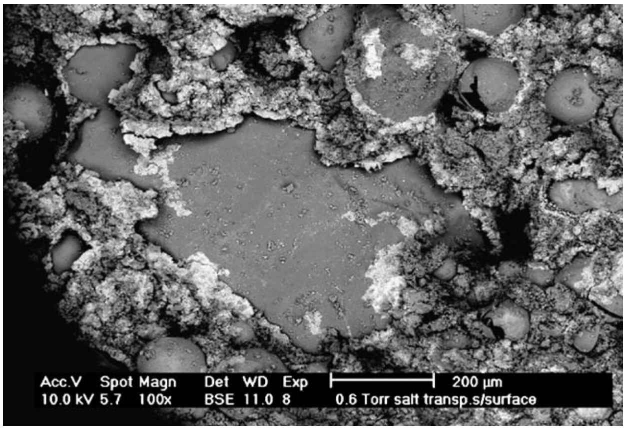

Fig. 7. ESEM photomicrograph showing NaCl crystallization at the binder/aggregate interface (surface of the plaster).

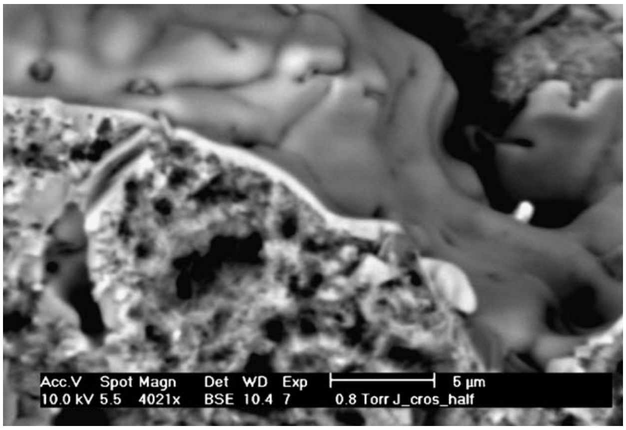

Fig. 8. ESEM photomicrograph showing NaCl crystallizing as a layer over the binder (cross section of the plaster).

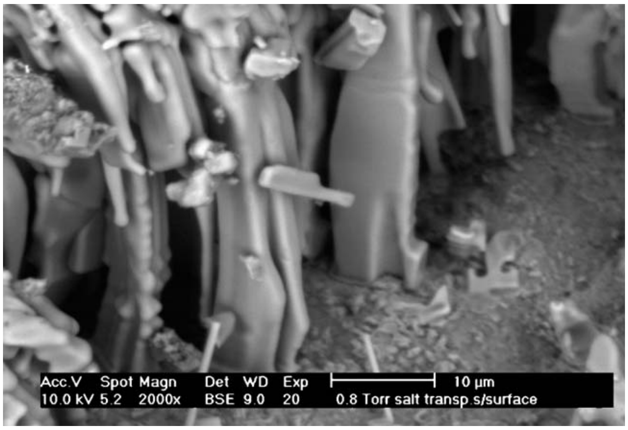

Fig. 10. ESEM photomicrograph showing needle-like NaCl crystals growing from the binder (surface of the plaster).

In specimens subjected to several salt crystallization and dissolution cycles damage occurred at the surface in the form of sanding: the loss of cohesion observed may be due to salt growing at the interface between the binder and the aggregate and pushing them apart (Fig. 9).

The crystal habits of the sodium chloride have been also studied by means of a BSE detector. The crystal shape may vary from a smooth layer (usually observed in the bulk of the plaster) (Fig. 8) to an agglomeration of more or less regularly shaped crystals. Elongated needle like crystals are often observed on the evaporation surface, growing from the binder (Fig. 10).

## 3. Performance of the plaster in the field

The above described “salt transporting” plaster has been often applied in restoration of buildings with a high salt load. From a survey performed on several monuments restored in the past 20 years, it appears that the performance of this restoration plaster may vary. The actual performance depends on the moisture and salt load of the wall, on the substrate/plaster combination and on the environmental conditions [18]. A high moisture and salt load of the substrate may threaten the good performance of this plaster. Besides, applying this fine porous plaster over a coarse substrate with a high salt and moisture load will enhance moisture and salt solution transport from the coarse substrate to the fine porous plaster layer where it will accumulate. The effect of the pore size distribution of the substrate/plaster combination on the moisture and salt solution transport has been measured in laboratory [19], and verified in practice on case studies [20,21].

The location of salt accumulation depends, apart from the moisture transport properties of the material or combination of materials, on the moisture supply and the evaporation rate [22]. Although this plaster is proposed by the producer as a material favouring salt transport from the masonry to the plaster surface, the moisture transport in this plaster is quite slow. From this it derives that, depending on the moisture supply and on the evaporation rate, this “salt transporting” plaster can turn into a “salt accumulating” plaster, i.e it leads to crystallization of the salt in the inner layer of the plaster.

The influence of the environmental conditions on the salt accumulation and the damage have been verified by comparing two field situations in which this plaster was applied: the exterior of a building in Curaçao, where high temperature and

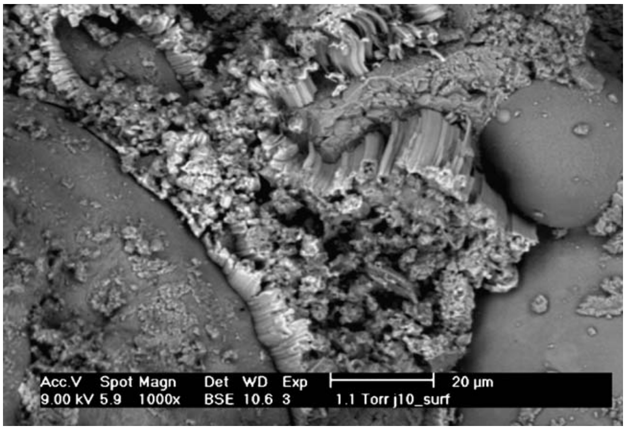

Fig. 9. ESEM photomicrograph showing NaCl crystallizing at the binder/aggregate interface (surface of the plaster).

Fig. 11. Location of salt accumulation in damaged (A) and undamaged (B100) cores (building in Curacao, NLA).

Fig. 12. Evolution of the damage in the years 2000 (left) and 2004 (right) (St. Nicholas Church, Brouwershaven, The Netherlands).

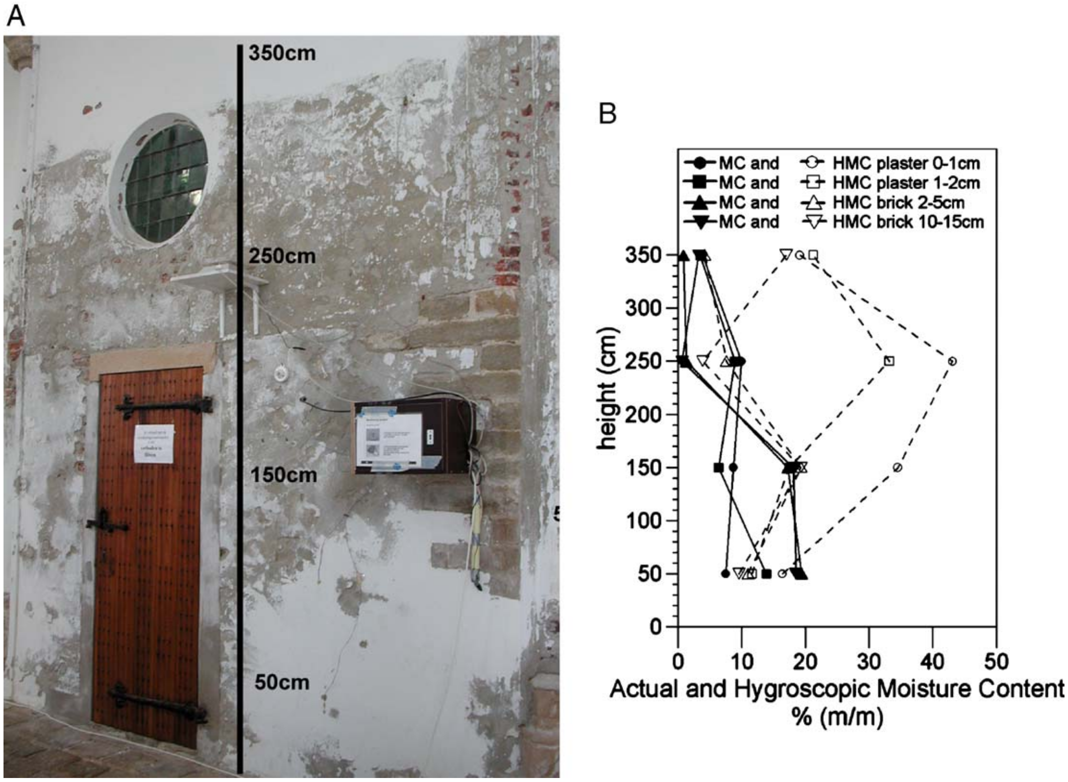

Fig. 13. Left: Sampling area; right: Actual Moisture Content (MC) and Hygroscopic Moisture Content (HMC) in the wall, showing that in the upper part of the damaged area no rising damp is present (St. Nicholas Church, Brouwershaven, The Netherlands).

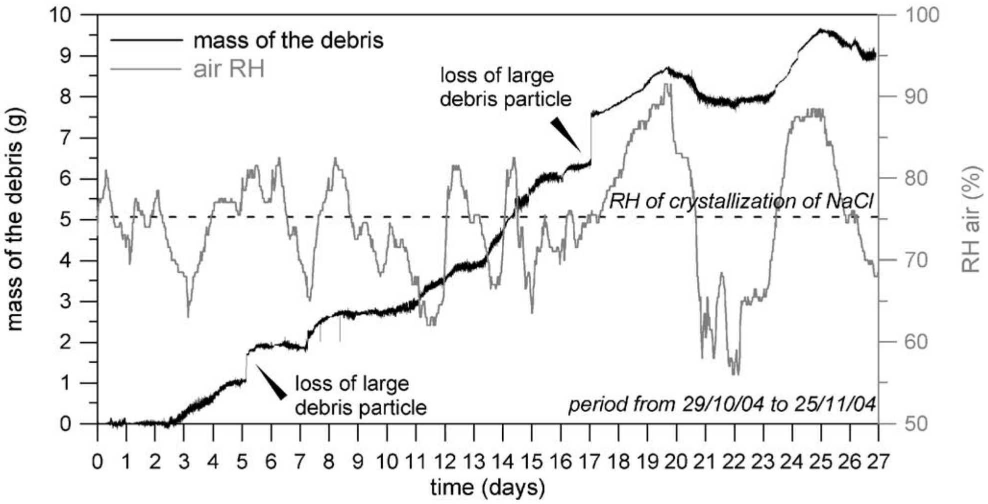

Fig. 14. Combined monitoring of the debris falling from the plastered wall and of the air RH: the graph shows that most of the debris are falling during the periods in which the RH increases (St. Nicholas Church, Brouwershaven, The Netherlands).

strong wind are present, and the interior of a church in the Netherlands, where moderate temperature and high RH exist.

In Curacao, after 7 years from the application of the plaster, cores were sampled from damaged and sound areas of the wall. It resulted that in the undamaged areas, where the plaster layer was thicker (4–5 cm), the salt had accumulated inside the plaster resulting in no visible damage. In the damaged area, where the plaster was thinner (2 cm), the salt had reached the surface giving place, instead of harmless efflorescences as hoped for a “salt transporting” plaster, to crystallization in the pores of the material and therefore to damage (Fig. 11). The damage appeared as peeling of the paint and sanding of the plaster layer after only 3 years from the application.

In the church of Brouwershaven, where the evaporation rate is slow and the moisture supply sufficient, salt accumulates near the outer surface of this plaster where it crystallizes, not only as harmless efflorescences, but also in the pores of the material, causing damage. Moreover, as soon as the salt has accumulated near the surface, it becomes very sensitive to the RH changes. If the salts present in the plaster are hygroscopic, (as in this case), they may undergo repeated dissolution/crystallization cycles whenever the RH of the air varies through the RH of crystallization of the salt(s). These dissolution/crystallization cycles enhance the decay. This process has been clearly demonstrated to be the cause of the damage occurring in the interior of the above mentioned church [9]. Due to the RH changes the hygroscopic salts present just beneath the surface of the plaster, dissolved and crystallized several times in a week, enhancing the decay. Clear increase of the damage has been observed in few years time (Fig. 12) in areas in which the RH of the air was the only moisture source (Fig. 13).

The damage, measured as debris falling from the wall, has been monitored together with the RH of the air: it resulted that most of the debris were falling when the RH increased and the salt, cementing the de-cohesioned particles of the plaster, dissolved (Fig. 14). This process has been reproduced and studied in ESEM chamber. Plaster scales sampled from a damaged area of the wall have been placed in the ESEM over a Peltier stage and the RH increased until dissolution of the salt occurred. When the salts dissolved the particles of the plaster, not anymore cemented by the salt crystals, fell apart.

## 4. Discussion and conclusions

A ready-to-use restoration plaster for salt loaded substrates has been studied in detail. Its composition and pore size distribution have been measured and related to the moisture and salt solution transport in the plaster. The very fine porosity (<0.1 μm) and the presence of an additive result in a slow absorption and drying behaviour. The slow moisture transport implies a slow transport of salt to the surface. For this reason the salt reaches the outer surface of the plaster only on the long term; if the evaporation is enhanced (as for example under high temperature or airflow) the salt will tend to accumulate inside the plaster. Therefore this plaster, proposed as “salt transporting” plaster, may behave, depending on the boundary conditions, as a “salt accumulating” plaster.

Sodium chloride crystallization in the plaster has been studied in order to define the preferential location of salt crystallization and the effect of it on the material structural integrity. The ESEM investigations showed that the salt precipitates mainly at the aggregate-binder interface: salt crystals growing at the interface may be the cause of the sanding often observed in this plaster. It was not possible to assess unambiguously, neither by MIP nor by ESEM, the pore size in which the NaCl preferentially crystallizes.

The results found in laboratory help in explaining the behaviour of this plaster in the field. The fine pore size distribution results in the practice of an effective extraction of salt solution from coarse porous substrates. Therefore the presence of a coarse porous substrate with a high moisture and salt load may constitute a risk factor for a successful application of fine porous plasters like this.

The slow moisture and salt solution transport observed in this plaster may lead to salt accumulation inside the plaster layer. From the investigated case studies it resulted that, if the evaporation is enhanced (for example by hot climate and wind) and the plaster is thick enough, the salt does not reach the surface and no visible damage is observed. In this case the plaster behaves according to the "salt accumulating" working principle. When the evaporation is slower or/and the plaster layer thin, the salt accumulates near the surface where it crystallizes not only in the form of harmless efflorescences, as expected for a "salt transporting" plaster, but also inside the pores of the material, causing damage.

A sufficient thickness of the plaster layer is therefore necessary to avoid surface damage.

Moreover, in a "salt transporting" plaster, hygroscopic salts tend to accumulate near the surface and are therefore very sensitive to the RH changes of the air. They may undergo dissolution/crystallization cycles whenever the RH of the air crosses the RH of crystallization of the salt. This has been verified to be a damage mechanism affecting this plaster in practice. This plaster, as well as other "salt transporting" plasters, is therefore less suitable for situations in which frequent RH changes, leading to dissolution/crystallization cycles of the salts, are present.

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