Development of a new accelerated salt crystallization test

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TNO Built Environment and Geosciences has developed, in cooperation with other institutes in the European project Compass[1], a new accelerated salt crystallization test. In this paper the design process leading to the definition of this test is described. Preliminary tests studying the influence of the environmental conditions on the damage development showed that the use of a high, but still realistic, temperature can enhance salt damage. The use of wet-dry cycles was found to be more effective than a continuous immersion of the specimens in salt solution. Experiments on different substrate/plaster combinations pointed out the relevance of the pore size of the system for the occurrence and type of decay: a coarse porous substrate may lead to faster damage to the plaster than a fine porous substrate. On the basis of the results obtained from this preliminary research a new accelerated test has been developed. The developed test procedure has been used in the crystallization experiments performed in the COMPASS project and has been proven to be effective even for the so-called "salt resistant" plasters.

Key words: Salt crystallization, accelerated crystallization test

1   Introduction

The durability of plasters, when applied on salt loaded substrates, may vary a lot. It is therefore very useful, both for plaster producers and people involved in restoration, to evaluate the durability of plasters to salt weathering by means of accelerated tests performed in laboratory. An effective crystallization test should satisfy the following requirements:

- obtain results in a relatively short period of time (few months) even in salt resistant materials as restoration plasters;
- be able to discriminate the durability of different materials;
- obtain a realistic type of damage, similar to the decay observed in practice

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1   *EU Project COMPASS 'Compatibility of Plasters And renders with Salt loaded Substrates in historic buildings' (EVK-CT 2001-00047).*

Unfortunately, the existing salt crystallization tests [WTA, 1992; RILEM, 1998] are usually not effective enough when applied on salt resistant plasters. Previous experiments showed that, in crystallization tests, damage appears only after several months of testing in laboratory and only in case of specimens contaminated with the most aggressive salts like sodium sulphate [Wijffels and Van Hees, 2001; Wijffels et al ,1997]. However, the same plasters, when applied in practice, may show considerable damage after a few years from the application [van Hees, 2003].

One of the most common limits of the existing crystallization tests consists in the relatively slow drying, usually at 20°C 50%RH [RILEM, 1998]: at these conditions, the plaster takes several months to dry, and this reduces the possible number of rewetting, and consequent dissolution-crystallization cycles, in a defined period of time. Besides, most of the test procedures prescribe to dry the sample in a climatic room at constant temperature and RH conditions, whereas temperature and RH cycles may contribute to the damage. Moreover, the existing procedures specifically developed for testing the durability of plasters [WTA, 1992] do not prescribe to test a plaster on a substrate, whereas the substrate/plaster combination plays a crucial role in the development and in the type of salt damage. For example, damage as the loss of bond of the plaster layer from the substrate, often observed in buildings, can not be simulated by tests performed on the plaster only.

In this paper, the process which has lead to the definition of an effective crystallization test is described. This has consisted of two phases studying the influence of several parameters on the effectiveness of the test. In a first run of experiments the effect of the drying conditions and the salt contamination procedure is studied (section 2). In a second series the consequences of the substrate/plaster combination on the plaster decay are considered (section 3). The results obtained in these preliminary phases have allowed defining a new accelerated crystallization test (annex).

2 Study of the effect of drying conditions and salt contamination procedure on the decay

The drying rate of the specimen is one of the most important parameters when designing a new accelerated crystallization test. Therefore, a preliminary investigation was carried out to determine the drying rate in six different climatic conditions and the consequences on the damage.

Another important variable to be checked is the way of contaminating the specimens with salt solution: this may occur by continuous immersion or wet-dry cycles. Both contamination procedures have been tested in this first run of experiments.

2.1 Materials

One combination of materials was selected on which all the different variables (drying conditions and contamination procedures) have been evaluated. The specimens used in this first series of experiment are 5×5×4.5 cm cubes constituted by a plaster layer (thickness 2 cm) applied on a brick substrate (thickness 2.5 cm).

The brick is a fired-clay brick. The plaster is a ready-to-use restoration plaster for salt loaded substrates: it is a cement-based accumulating system constituted by two layers with mixed-in water repellent. The physical properties of the substrate and of the plaster are reported in table 1.

Table 1: Table 1 Properties of the ground and top layer of the plaster

	Dry density (Kg/m³)	Total porosity* (% by volume)	WAC** Kg/m² √s
Fired-clay brick	1691	36.2	0.303
Plaster -ground layer	837	37.5	0.022
Plaster - top layer	803	43.5	0.004

* determined according to [Klugt van der and Koek, 1994]
** determined according to [EN 1015-18:1996]

2.2 Procedure

After preparation and curing of the brick/plaster specimens (details of the specimen preparation can be found in [Lubelli et al., 2003], all the specimens were sealed with epoxy resin on the lateral sides. The specimens were contaminated with NaCl solution. Two different procedures have been used for contaminating the specimens:

- continuous immersion of the bottom of the specimen in NaCl solution of 10% by weight
- initial immersion of the bottom of the specimen in an amount of NaCl solution sufficient to reach a 1.5% of salt in the specimen, expressed as percentage of the dry weight. Once 80% of the water was evaporated, the specimens were re-wetted, by capillary rise from the bottom, with a quantity of demi-water equal to the quantity of solution used in the first wet-dry cycle.

In both cases, the drying of the specimens was allowed only through the upper surface.

Six different drying conditions were selected:

- 20°C 50%RH - constant: reference condition, prescribed in the RILEM test [RILEM, 1998], and with which the others are compared.
- High temperature (50°C, 50%RH) – constant: it aims, by the use of high temperature, remaining however at a realistic level, to enhance the drying and allow more wet-dry cycles.
- Low RH (35%) at 20°C – constant: as the previous, aims to increase the drying rate.
- Temperature cycles: one week at 20°C 50%RH, followed by one week at 50°C 50%RH. In this case, the effect of differential thermal expansion may play a role in the damage.
- RH cycles: one week at 20°C 50%RH followed by one week at 20°C 96%RH. These conditions can produce crystallization-dissolution cycles more frequently than what is obtained by rewetting and drying the specimen.
- Combined temperature and RH cycles: these cycles, between 20°C 65% RH and 35°C 20% RH, are obtained by means of IR lamps that are kept 8 hours on and 16 hours off.

*2.3 **Results***

The aim of these preliminary tests was to define the boundary conditions (drying conditions, contamination procedure) most effective for the development of salt damage. Even if at the end of the testing period no damage was visible yet, the effectiveness of the different parameters could be evaluated on the basis of the salt distribution within the plaster, a high salt content at a certain level in the plaster indicating a higher risk of damage.

2.3.1 Effect of the different environmental conditions on the drying speed
The use of high temperature, either obtained in a climatic box or by means of IR lamps, has the most important effect on the drying rate. Moreover, the use of high temperature, apart from causing a faster crystallization leading to more harmful non-equilibrium crystals [Rodriguez Navarro and Doehne, 1999], is the only way to dry this plaster (and cause crystallization of the salt) in a reasonable period of time. Specimens conditioned at 20°C/50% and at 20°C/35% do not dry completely after a few months. Specimens subjected to RH cycles between 50 and 96%RH even show an increase in weight in case of plaster slabs: the hygroscopic moisture taken up during the high RH period does not evaporate completely during the low RH period, leading to an average gain of weight. It is clear that no crystallization can occur in these conditions and, therefore, no damage.

2.3.2 Effect of the different environmental conditions on the salt distribution
All the studied drying conditions lead to a similar salt distribution, consisting in salt accumulation in the inner layer, according with the salt accumulating principle of the tested

plaster (Figure 1). Therefore, no choice for one drying condition could be made on the basis of the salt distribution.

2.3.3 Effect of the different contamination procedure on the salt distribution
The specimens undergoing wet-dry cycles show the highest salt accumulation indicating that this test procedure is more harmful than a continuous immersion in salt solution (compare figures 1 and 2).
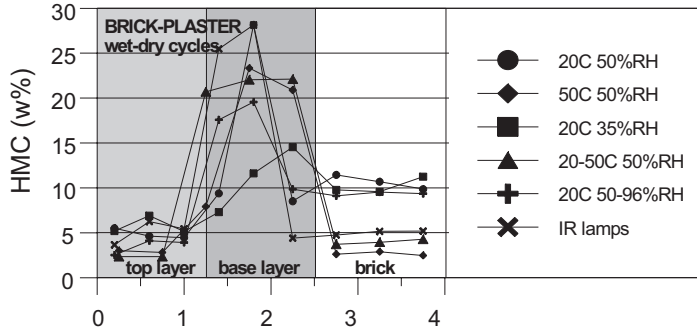

Figure 1: Hygroscopic moisture distribution at 96%RH of brick-plaster specimens undergoing wet-dry cycles
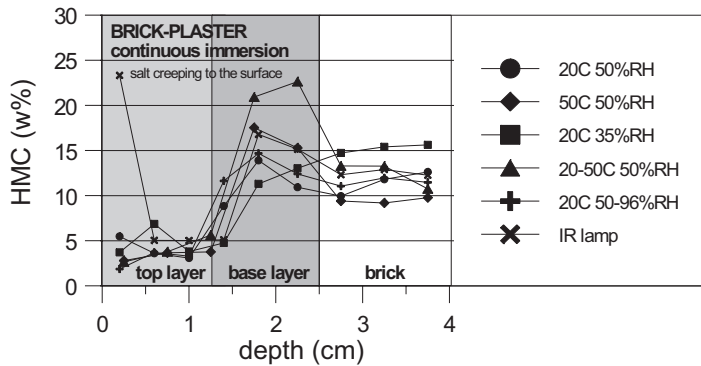

Figure 2: Hygroscopic moisture distribution at 96%RH of brick-plaster specimens continuously immersed in salt solution

Considering the transport of salts during subsequent cycles, it is visible that repeated wet-dry cycles transport the salts towards the surface (Figure 3), where they could even cause the development of surface damage. A procedure allowing as many as possible repeated wet-dry cycles should therefore be preferred.
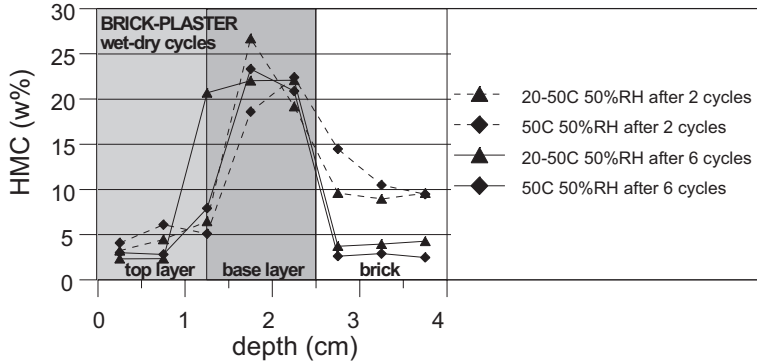
Figure 3: Hygroscopic moisture distribution at 96%RH of brick-plaster specimens measured after 2 and 6 wet-dry cycles

3   Effect of substrate/plaster combination on the decay

Main goal of this second phase of the research was to evaluate the influence of the substrate properties (mainly pore size distribution) on the salt solution transport in the substrate/plaster system and on the damage development. This research on the influence of the substrate had also the aim to validate theoretical concepts on salt transport between porous medias having different pore systems as developed by [Petkovic et al., 2004].

3.1 Materials

The accelerated test was performed on 4 combinations of two plasters and two substrates. Half of the specimens were painted with silicon emulsion paint. The materials were chosen on the base of their pore size distribution. The following substrates were used:

• Bentheim sandstone, coarse porous
• Calcium silicate brick, fine porous

Figure 4 shows the pore size distribution of both substrates. In table 2, the main properties of the substrates are given.

From figure 4, it can be concluded that the calcium silicate brick is more porous than the Bentheim sandstone, 27 to 24%. The pores of the calcium silicate brick are much finer and the pore sizes are more spread. The pore dimensions vary from 0,01 µm to 50 µm. The pore size distribution of the Bentheim sandstone is completely different. Most of the pores have a diameter between 10 and 50 µm.
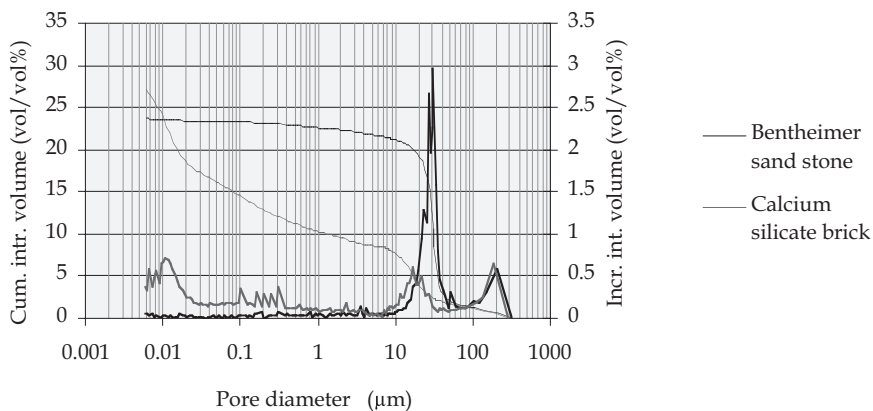
Figure 4: Pore size distribution of the substrates

Table 2: Characteristics of the substrates

	Bentheim sandstone	Calcium silicate brick
average pore size [μm]	25	0,15
free water absorption [% by volume]	15,6	22
bulk density [kg/m³]	2036	1898
porosity [% by volume]	24	27
Water Absorption Coefficient (WAC) [kg/(m²√s)]	0,73	0,09

The pore size distribution influences the water absorption coefficient (WAC). The Bentheim stone absorbs water much faster than the calcium silicate brick. On these substrates, two types of plaster were applied, made of the same components but mixed using different ratios.

The plaster used in the accelerated test is composed of sand (S), blast furnace slag cement (BFS) and lime (L) in the following proportions:

- 1 BFS : 4 L : 20 S, binder/aggregate ratio of 1 to 4
- 1 BFS : 4 L : 10 S, binder/aggregate ratio of 1 to 2

Figure 5 shows the pore size distribution of plaster 1:2 applied on both stones. Figure 6 shows the 1:4 plaster.
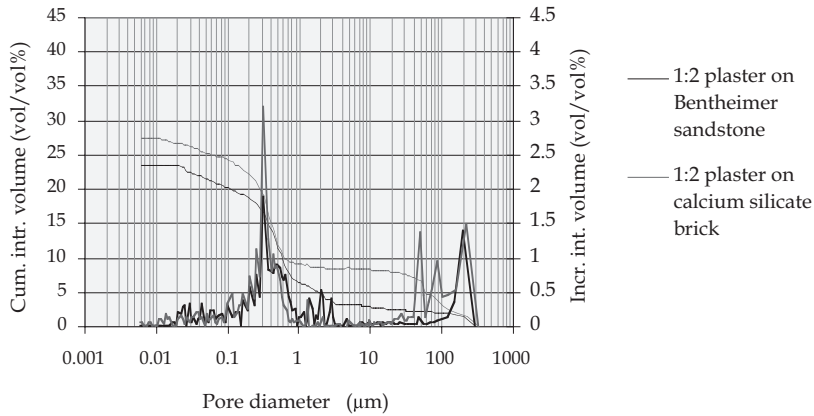
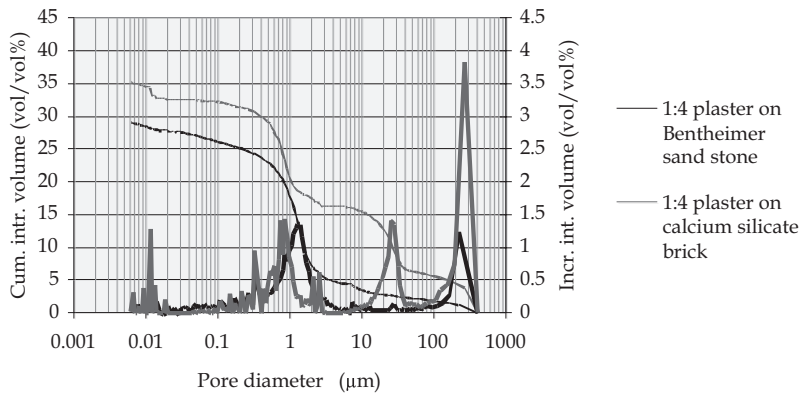*Figure 5: Pore size distribution of the 1:2 plaster on both substrates*

Figure 6: Pore size distribution of the 1:4 plaster on both substrates

*3.2 **Procedure***

Based upon the results of the preliminary test described in section 3, the procedure for the accelerated crystallization test was designed as follows:

1. The specimens are contaminated with a salt solution (concentration 10% by weight)
2. The specimens are sealed on the lateral sides with epoxy resin
3. The drying takes place in cycles of temperature and relative humidity: 8 hours 20°C-65% RH followed by 16 hours 35°C-20% RH

4.   After evaporation of at least 80% of the salt solution, the specimens are contaminated
      again. Wet-dry cycles are used according to the procedure described in 2.2 but in this case
      instead of demi-water, a salt solution (10% by weight) is used so that the salt amount in
      the specimen is increased with 1.5% by weight of the specimen at each contamination
      cycle.

3.3 *Results*

3.3.1 Drying

Drying of the specimens in the prescribed temperature and RH conditions occurs quite fast. The
moisture content at the start of the test was approximately 6% by weight. After 5 weeks more
than 80% of the salt solution was evaporated and the specimens were contaminated again. This
is a relatively short period of time which allows to repeat more wet-dry cycles during a test
period of few months.

3.3.2 Damage

Damage to the surface of the specimens was assessed every week by visual inspection and
photographical monitoring. The amount and type of damage were recorded.

Amount and development of the damage

Table 3 shows the amount of damage occurred to the different specimens. The number indicates
the severity of the damage in a range from 0 to 5: 0 corresponds to no damage, 5 indicates
maximum damage. The last column gives the average over the testing period. The higher the
number, the more and earlier the damage occurred.

Table 3 shows a clear difference in the seriousness of damage as a result in the difference in
substrate. The damage to the plaster on the coarse porous Bentheim sandstone was much more
severe than the damage to the plaster on the fine porous calcium silicate brick. This is explained
by the fact that the plaster will absorb more effectively the salt solution from a coarse porous
substrate than from a fine porous one. Salts will be transported in the plaster where they will
crystallize causing damage. From this it emerges that an accelerated crystallization test to
distinguish difference in durability of plaster can be better performed using coarse
substrate/plaster combinations.

Table 3: Damage surveyed in the specimens

nr	substrate	plaster	paint	Time (weeks)										Average
				1	2	3	4	6	8	10	12	14	16	
1	Bentheim sandstone	1:2	Paint	2	3	4	4	4	5	5	5	5	5	4,2
2			No paint	3	3	5	5	5	5	5	5	5	5	4,6
3		1:4	Paint	2	2	3	3	4	4	4	5	5	5	3,7
4			No paint	5	5	5	5	5	5	5	5	5	5	5
5	Calcium silicate brick	1:2	Paint	1	1	1	2	2	2	2	2	3	3	1,9
6			No paint				1	4	4	4	4	5	5	2,6
7		1:4	Paint											0
8			No paint					3	3	4	4	4	4	2,2

Differences in surface damage can be observed between painted and not-painted plasters. In general, the damage to the not-painted plasters is more severe than the damage to the painted plaster. Compare the average damage of nr. 2, 4, 6 and 8 to the average of nr. 1, 3, 5 and 7. From the above reported discussion, it may be concluded that the test is effective since damage to the selected plasters occurred within a relatively short period of time (some weeks). Besides, the test is able to point out differences in the durability of the selected substrate/plaster combinations.

Type of damage

Different types of damage were observed during the accelerated crystallization test, which are listed in table 4 and shown in figure 7.

Table 4: Type of damage

nr	substrate	plaster	paint	Types of damage
1	Bentheim sandstone	1:2	Paint	Cracks, peeling
2			No paint	Bulging, efflorescence
3		1:4	Paint	peeling
4			No paint	Bulging, efflorescence
5	Calcium silicate brick	1:2	Paint	Needle like efflorescence
6			No paint	Cracks, efflorescence
7		1:4	Paint	
8			No paint	Small cracks efflorescence

The new test has been proven to be a realistic simulation of the damage in practice. The types of damage obtained are similar to the ones observed on historic buildings. Peeling, blistering of paint and bulging are damage types common on plasters applied on salt loaded substrates.

Figure 7: Peeling of paint (left), bulging, cracks, efflorescence (middle), cracks (right)

3.3.3 Moisture and salt distribution

After 4 drying cycles, the specimens were cut into slices and the distribution of both salt and moisture was determined. The moisture content was measured gravimetrically. The salt distribution was determined by measuring the hygroscopic moisture content of each slice at 95% RH. Figure 8 gives both the moisture and salt distribution profile of the specimen 1:2 plaster on the calcium silicate brick. Figure 9 gives these data for the same plaster on the Bentheim sandstone. The left part of the graph is the plaster, the right part the substrate. The drying surface is on the left.

In both figure 7 and 8, two lines are given. The continuous line indicates the moisture content after 4 drying cycles. The dotted line indicates the hygroscopic moisture content, which gives the salt distribution profile. The moisture distribution shows a difference between the two substrate-plaster combinations: in calcium silicate brick specimens the moisture content in the substrate is higher than in the plaster; in the sandstone specimen the moisture content in the substrate is much lower than in the plaster. Again, this is related to the different pore systems of the substrate/plaster combinations.
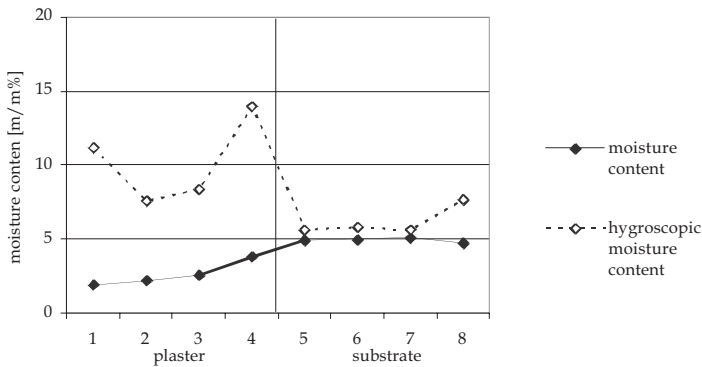

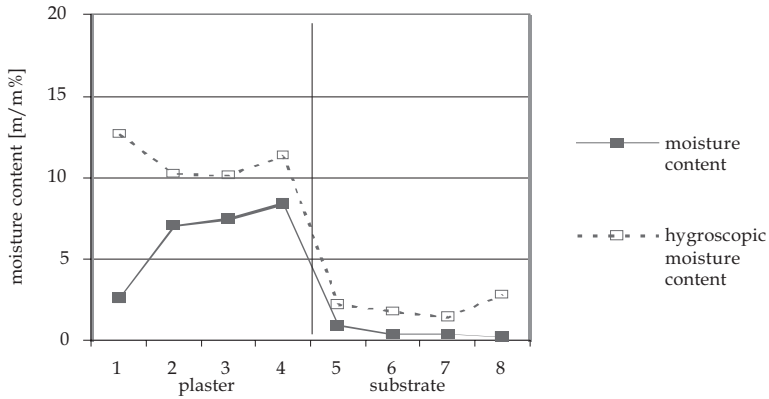*Figure 8: Moisture and salt distribution profile 1:2 plaster on calcium silicate brick*

Figure 9: Moisture and salt distribution profile 1:2 plaster on Bentheim sandstone

4   Conclusions

The new test proved to be effective, since:

- it is able to obtain results in a relatively short period of time (1 to 6 weeks, depending on the type of substrate).
- it is able to discriminate between the durability of different plaster/substrate combinations
- the types of damage observed on the specimen at the end of the test are similar to the ones that occur in plasters in historic buildings.

The new developed has been further used during the European Project COMPASS to evaluate the durability of different plasters (ready-to use products as well as traditional plasters). The reproducibility of the test procedure has been evaluated by performing the same test in different laboratories. The results obtained have been positive and encourage the use of this procedure in salt crystallization tests [Verges-Belmin et al., 2005]

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Annex - Accelerated crystallisation test procedure

Specimens

Specimens are constituted by a plaster layer applied on a substrate.

Number of specimen

The test is carried out in threefold.

Size of specimen

The specimen size is approximately 5×5×5cm. The thickness of the substrate is about 2cm.

Substrate

The salt resistance of different plasters has to be compared using one type of substrate. This substrate should be homogeneous and able to transport the salt to the plaster rapidly. A fired clay brick may be used.

Preparation of the specimens

The specimens are prepared by applying the plaster layer on a substrate. If a brick substrate is used the plaster layer will be applied on each brick and then from each brick 3 specimens will be cut (figure A1). The specimens will be dried and their lateral sides sealed using epoxy resin.
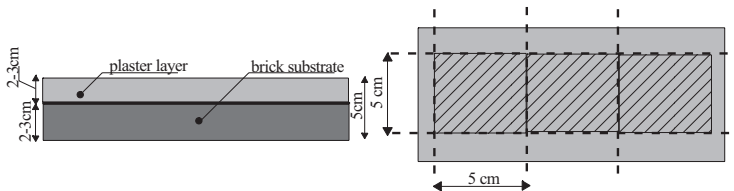

Figure A1: Specimen preparation

The plaster may be applied on the substrate with or without the use of an adhesion layer. The adhesion layer is used when:

- a poor bond between plaster and substrate is expected
- the application procedure of the supplier demands an adhesion layer

The adhesion layer is generally composed of binder and water.

Curing

The curing regime depends on the type of plaster.

Hydrated lime:

When hydrated lime is used, the curing will include an accelerated carbonation.

A suitable carbonation of lime mortar is obtained using the following procedure:

1. pre-curing for 14 days in 20 ℃ / 65% RH

2. drying at 40 ℃ for 24 hours

3. accelerated carbonation in 3% CO_2 with 50% RH

Using this procedure the carbonation takes place in 3 to 7 days.

Combination of hydrated lime and cement

When the binder consists of both cement and lime, the following procedure is proposed:

the plaster is covered with plastic for 7 days

pre-curing for 21 days in 20 ℃ / 65% RH

drying at 40℃ for 24 hours

accelerated carbonation in 3% CO_2 with 50% RH

Cement and other hydraulic binders

Curing consists of the following procedure:

The plaster is covered with plastic for 7 days

Curing for 21 days in 20℃ / 95%RH

Test procedure

1. Seal the lateral sides of the specimen in such a way no evaporation takes places from these sides. An epoxy resin or paraffin will do.
2. Weigh the dry samples and record the mass.
3. Fed the salt solution (10% weight of the anhydrous salt/weight of the solution) into a container.
4. Immerse the bottom of the specimen in the salt solution (concentration 10% by weight) for a period long enough to wet the upper surface of the substrate. Preliminary tests should be done on the substrate in order to define the length of the immersion period. In the case of the fired-clay used in the COMPASS, a period of 5 minutes was necessary. The solution level has to be always enough to allow absorption.
5. Remove the specimen from the container. After removal of free liquid from the lateral sides and base of the specimen, record the mass.
6. Wrap the bottom of the specimen with water vapour tight tape. Record the mass of the specimen again.
7. Place the specimens in the prescribed T and RH conditions. The following temperature and RH cycle is used: 16 hours 40 0C / 20% RH and 8 hours 20 0C / 65% RH.
8. Weight the sample every week and record the mass. Record any appearance of damage (type and seriousness) and efflorescences. Do not remove efflorescence or debris from the surface of the specimens
9. When 80% of the solution absorbed is evaporated, remove the tape from the bottom of the sample and record the mass of the sample. If efflorescences or debris are present on the surface of the specimen, remove them with a soft brush and record the weigh after brushing.
10. Repeat the procedure from point 2

Evaluation of the damage

The following aspects have to be monitored for the evaluation of the damage:

- occurrence of visible damage to the surface or in the bulk of the specimen
- change in mass (drying speed)
- loss of material

When no damage to the plaster occurs, the salt distribution profile at the end of the test is used as indication of the salt resistance of the plaster. The salt distribution also serves as a method to classify the plaster (transporting, accumulating, blocking, etc.)

Determination of the salt distribution profile

In order to determine the salt distribution, the specimens are cut (without the use of water) in 8 slices (4 sliced in the plaster and 4 in the substrate) (Figure A2).
The slices are dried at 60ºC until constant weight and then set at 20 ºC / 96% RH until constant weight is reached The Hygroscopic Moisture Content (HMC) of the slices is calculated as follow:

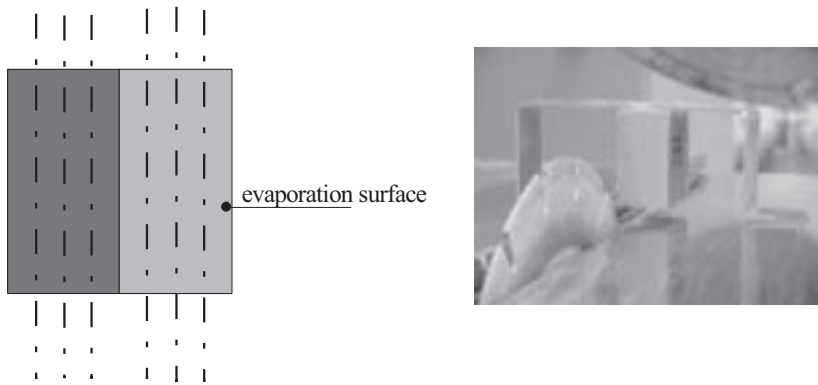$$HMC = 100* \text{(weight at 96\% RH - dry weight)/dry weight}$$

The HMC gives an indication of the salt content.

Figure A2: Slicing of the specimen