SELF-HEALING LIME MORTARS: OVERVIEW OF AN EXPERIMENTAL INVESTIGATION FROM MATERIAL TO SUBASSEMBLY SCALE

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Abstract
Natural hydraulic lime-based mortars are recommended for retrofitting operations in historical buildings, primarily because of their high chemical, physical and mechanical compatibility with the existing mortars; moreover, their autogenous and engineered self-healing capacities make them a suitable material for the aforementioned interventions.

The authors’ group has undertaken a comprehensive investigation on the topic, developing tailored methodologies for the assessment and characterization of the healing capacity of lime based mortars at the material and structure subassembly level, and have addressed a constitutive modelling approach which incorporates the effects of healing scenario on the response of lime based mortars. At the material level the methodology was based on pre-damaging specimens, at different ages and levels of damage (70% of the compression strength in pre-peak regime; 90% of the compression strength in post-peak regime), curing them under different exposure conditions for the healing to occur, and retesting them after scheduled “healing period” to evaluate the recovery of physical and mechanical properties. At the sub-assembly level the same concept was applied to brick-mortar specimens, to evaluated the effects of healing on the brick-mortar bond, meant a crucial to the structural behaviour of the masonry render.

The paper summarizes the main results obtained with a focus on the scalability of the healing results from the material to the subassembly to the structural level, as required for engineering applications.

Keywords: self-healing, lime-mortars, brick-mortar subassemblies, architectural heritage.
1. INTRODUCTION

Lime-based mortars—consisting of lime as a binder, fine aggregates (generally river or quarry sand) and water—have been used since ancient times in stone and brick masonry buildings, dating back to as early as 12,000 BC in Palestine and Turkey [1] and, through ancient Greek and the Roman Empire [2], have been continuously used almost without interruption so far. Several reasons can be called to justify this long-time use of lime as binder, including availability, relatively easy production, good workability and adhesion to different subgrades, as well as the ability of accommodating movements, such as settlements, and avoiding the resulting stress concentrations that can cause failure in brick masonry wall. In recent years, the use of lime-based mortars in restoration of cultural heritage has increased, also because of their chemical-physical and mechanical compatibility with old renders. Several studies can be found in the literature investigating the composition and properties of mortars for repairs, with an increasing interest in the use of pure lime and hydraulic lime mortars [3,4]. As a matter of fact, cement based mortars are not compatible with ancient masonry, not only because of their higher mechanical strength and stiffness and because they may introduce soluble salts, [5], whereas lime mortars, due to their high chemical purity, hardly can act as efflorescence promotors.

As a matter of fact, due to dissolution, transport and re-precipitation of calcium compounds, it can also have autogenous self-healing properties, as firstly recognized by Anderegg [6], in 1942. He attributed this capacity, in mortars with high lime content, to the deposition of calcite in cracks and also noticed that mortar made from hydrated dolomitic lime had a better performance than one high-calcium quicklime. Lubelli et al. [7], through microscopy analysis, highlighted that the presence of water, even in form of air moisture, is the necessary condition for the occurrence of self-healing. They observed that, in case of a relevant amount of free lime, water could be responsible of its dissolution and transport to the damaged area. Therefore, autogenous-healing is possible if atoms or molecules can move from their initial position to the micro-crack surfaces, and there can re-crystallize to form calcium compounds. These processes of transport and re-precipitation observed for both CaCO3 and Ca(OH)2 into voids and micro-cracks, are responsible for the reconstruction of the matrix trough-crack continuity and build-up of an enhanced load bearing capacity. The same authors also developed a test procedure to reproduce the self-healing of lime-based mortars (both pure calcium and magnesium-calcium) in laboratory conditions. The main conclusion of these studies is an interesting relationship between the healing capacity and the precipitation of magnesium phases. So far, several studies have regarded to self-healing capacity of lime-based mortars as a sort of bonus, mainly in old masonry constructions [3], but these properties have seldom been sought through an appropriate material design. The possibility of engineering this process with the addition of innovative constituents in traditional lime mortars, is therefore of the utmost interest. The aim is to obtain advanced “smart” lime mortars, which are able to enhance the durability of the whole building, while maintaining the authenticity of structural brick elements.

In this paper the results of a comprehensive investigation are presented which focused on the development and calibration of an experimental methodology to ascertain and quantify the self-healing capacity of lime mortars through the recovery of physical (ultrasonic pulse velocity UPV) and mechanical (compressive strength) properties. Autogenous self-healing was investigated together with stimulated/engineered one, via both commercial crystalline additives and tailor-made capsules containing different “active” compounds. The effects on brick masonry subassemblies have been also preliminary assessed.
2. EXPERIMENTAL CAMPAIGN: MATERIALS AND TEST METHODS

With the aim of reproducing the composition and performance of mortars found in existing historical building, a reference lime mortar was produced, consisting of natural hydraulic lime (NHL5) with the addition of calcium hydroxide (Ca(OH)_2), in equal mass proportion, and with dolomitic sand (lime to sand weight ratio equal to 1/3). A water to lime mass ratio equal to 0.22 was employed. The composition of the lime is listed in Table 1. 50 mm side cubes were cast with the mortar, which were demoulded 96 hours after casting (during which time they stayed in a lab environment) and then stored in a room at (23 ± 2)°C and (50 ± 4%) RH until testing.

The mortar compressive strength was first determined at 14, 28, 56 and 84 days. Before testing, UPV tests were performed on each specimen, according to three directions. At the same ages, other specimens were pre-damaged by loading them respectively at 70% of the compressive strength in the pre-peak regime and at 90% of the same strength in the post-peak regime. Three specimens per each level of pre-loading and age were pre-damaged. Before and after pre-damaging, UPV measurements were taken for these specimens as well. Six damaged pre-loaded specimens per level, together with six further undamaged specimens per each pre-cracking age, were then cured in water for 14 and 28 days respectively.

At the end of the curing periods as above, all specimens were re-analysed by means of UPV test and re-tested up to the maximum compressive strength and then re-cured in water for further 14 days. At the end of this second curing period, specimens were finally tested to failure (UPV measurements were taken once again before this first test). Tests were performed on a Galdabini Sun 20 testing machine under displacement control, at a displacement rate equal to 0.2 mm/min.

The recovery, in case, of compressive strength along the cracking/healing cycles and the comparison with the strength evolution of undamaged specimens will allow the effects of healing to be quantified and connected to the recovery of the damage as through UPV tests.

For stimulated/engineered healing a simpler version of the programme was implemented, consisting of the following phases (per each type of mix/healing stimulating agent):
- after 28 days of curing, one specimen was tested for evaluating compressive strength;
- four specimens were pre-damaged at 70% of the compressive strength as above;
- two out of these four specimens were immediately retested after pre-damaging;
- the remaining two pre-damaged specimens were tested to failure after 14 days in water.

A commercial crystalline admixture (Penetron Admix®,) dosed at 3% by weight of lime, was used as healing stimulator, already widely employed in concrete [8-11]. The admixture consists of proprietary active chemicals, highly hydrophylic, able to react with the calcium hydroxide.

As for the encapsulated agents, four types of coated granules were produced and tested:
- hydraulic lime-based, with an inorganic shell (NHL5_NaF – sodium fluoride), with organic shell (NHL5_AM_AF maleic/phtalic anhydride) and with shellac shell;
- cement based, with two different types of organic shell (CEM_AM; CEM_AF) or shellac;
- calcium hydroxide Ca(OH)_2 with maleic/phtalic(AM/AF) anhydride or shellac shell.
Capsules were dosed from 3 to 6% by mass of the lime. Further details can be found in [12]. The whole experimental campaign is summarized in the Gantt chart in Figure 1.

Table 1. Chemical analysis of the main constituents of lime.

<table>
<thead>
<tr>
<th>CaO</th>
<th>MgO</th>
<th>Al₂O₃</th>
<th>K₂O</th>
<th>Na₂O</th>
<th>Fe₂O₃</th>
<th>SiO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>62%</td>
<td>1%</td>
<td>5%</td>
<td>0,7%</td>
<td>0,3%</td>
<td>3,5%</td>
<td>21%</td>
</tr>
</tbody>
</table>
In order to evaluate the influence of the self-healing capacity of the mortar on brick-mortar masonry subassemblies, shear tests were performed on masonry triplets consisting of three clay bricks and two 10 mm thick mortar layers, tested as per EN 1052-3. Bricks 250x120x60 mm³ were used, with free water absorption equal to 10% by mass, dry density 1660 kg/m³, compressive strength 18 MPa and splitting strength ± 2.2 MPa. Bricks were conditioned by submerging them in water for 200 seconds. A first series of triplets, cured at 23°C ± 2° and 50% ± 4% RH for 28 days, was tested with and without transverse normal stress equal to 0.6 MPa, to determine the shear bond strength. Then, a second series of samples was pre-loaded up to 70% of the shear bond strength, determined as above, immersed in water for 3 months and re-tested to failure. Reference undamaged samples undergoing the same curing history as the damaged ones were tested as well. All tests were performed with and without a transverse normal stress equal to 0.6 MPa and using a Galdabini Sun 20 testing machine under displacement control at a displacement rate equal to 0.2 mm/min. The transverse pre-compression force, was applied through four transversal threaded rods by a torque wrench, held constant during the whole test, and measured before and after the tests.

3. EXPERIMENTAL AND NUMERICAL RESULTS

3.1 Autogenous healing: material level

Figure 2 shows the results of compressive strength tests performed on undamaged and damaged specimens, for both levels of damage, at different ages and for the different scheduled post-damage curing periods. It can be first of all observed that the compressive strength of the mortar decreases over time, a phenomenon attributable to the dry lab environment in which the specimens were kept, which may have promoted the formation of microcracks. The strength increase after curing in water, even for virgin specimens, is likely to confirm this assumption, since water, penetrating through crevices, may reactivate hydration reactions.

The effects of healing are also evident. Specimens cured in water continuously undergo a recovery of the strength, up to level comparable to virgin ones. Moreover, even if tested up to
the maximum stress and recured in water, specimens did exhibit the capacity of gaining back the original load bearing capacity, if not higher. This phenomenon progressively decreases with the age of the first damage, being due to the reactions which involve potentially reactive material, which is progressively consumed along the aging of the specimens.

From the results of the UPV tests, shown in Figure 3 in terms of damage (grey bars) and healing ratios (blueish bars), a damage and healing ratios were defined as follows:

Damage ratio \( D = 1 - \frac{v_d}{v_0} \) \hspace{1cm} (1a)

Healing ratio \( H = \frac{v_h}{v_0} - 1 \) \hspace{1cm} (1b)

where \( v \) is the UPV, subscripts \( 0, d, h \) refer respectively to the virgin, damaged and healed state.

Figure 2: effect of loading level/age and of curing regime on mortar compressive strength
The healing capacity decreases with the age of predamage, most significantly for longer ages, whereas it increases with the duration of the curing. The level of damage also affects the healing capacity, the latter being far less significant for specimens damaged in the post-peak regime. Significantly, a prolonged curing in water (28 days) can promote a healing sufficient to gain back the pristine level of performance even for heavily damaged specimens, preloaded up to 90% of the compressive strength in the post-peak regime.

![Figure 3](image)

Figure 3: damage and healing degree as function of pre-loading level, age and curing regime.

### 3.2 Stimulated/engineered healing: material level

Figure 4 shows the healing ratio for the different types of encapsulated agents. As expectable, capsules containing cement provide the highest healing, whereas for other encapsulated materials the results are quite comparable, if not sometimes worse, with those of the reference mortar. It is worth remarking that these results may have also been affected by the actual compressive strength of the mortars with encapsulated additions, which in some case was significantly lower that for the reference mortar, since capsules may act as defects.

In order to quantify the stimulating effect of the crystalline admixture on the healing capacity an index of healing \( \text{ILR} \) was calculated as follows:

\[
\text{ILR} = \frac{f_{m,\text{after healing}} - 0.7 f_{m,28d}}{f_{m,28f}}
\]

where \( f_m \) denotes the compressive strength of the mortar, pedices specifying at which stage it was measured. The beneficial effects of the crystalline admixture are clearly evident (Figure5), for both specimens immersed in water and cured in air. It can be hypothesized that the admixture reacts with water even in the form or air moisture, sealing the cracks and fostering the recovery.

As time goes by, the sealing proceeds and water can pass through the matrix to a lesser and lesser extent, slowing the rate of performance recovery, which stabilizes after six months.

### 3.3 Autogenous and stimulated healing: subassembly level

All masonry triplet specimens failed in sliding shear, which allowed to process the results in terms of Mohr-Coulomb criterion:

\[
f_v = f_{v0} + \mu \sigma
\]

where \( f_v \) is the shear strength; \( f_{v0} \) is the shear strength with zero normal stress applied (which can be regarded as a “cohesion” parameter); \( \mu \), the friction coefficient. Results are summarized in Table 2 (in brackets the scattering). The effect of healing on pre-damaged subassemblies is likely to bring a significant recovery of the performance, up to about 85% of the capacity of
virgin specimens in terms of cohesion, and even better in terms of friction. The effect of the admixture as well appears to be significant in terms of friction and quite negligible in terms of cohesion. This may be due to an increase in the roughness of the interface surfaces, which, while counteracting with the shear strength under no confinement, can significantly benefit from the presence of normal stress, as occurs in real masonry assemblies.

Table 2. Effects of pre-damage and healing on subassembly behaviour parameter

<table>
<thead>
<tr>
<th></th>
<th>Reference mortar</th>
<th>With crystalline adm.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( f_{0} [\text{MPa}] )</td>
<td>( \mu )</td>
</tr>
<tr>
<td>Undamaged - instantaneous test</td>
<td>0.19 (0.07)</td>
<td>0.29 (0.14)</td>
</tr>
<tr>
<td>Undamaged - 3 months in water</td>
<td>0.41 (0.08)</td>
<td>0.20 (0.13)</td>
</tr>
<tr>
<td>Pre-damaged - 3 months in water</td>
<td>0.35 (0.12)</td>
<td>0.57 (0.16)</td>
</tr>
<tr>
<td>Pre-damaged - 6 months in water</td>
<td>0.20 (0.09)</td>
<td>0.47</td>
</tr>
</tbody>
</table>

### 4. CONCLUSIONS

The results of the experimental investigation summarized in this paper have shown that lime mortars possess an inherently autogenous healing capacity, depending on the level on age of damage. Being the self-healing dependent of dissolution, transport and re-precipitation of
calcium compounds, its effectiveness depends on the available amount of the latter, the degree of carbonation of the mortar and the presence of phases affecting the solubility of calcium bearing compounds, besides the curing environment conditions. The presence of crystalline admixtures enhances the aforementioned self-healing capacity and speeds up the sealing of the cracks and the recovery of the mechanical properties. Coated granules, in case containing the same mortar as an active principle, also may interesting enhance the healing, the scalability of their production to an industrially feasible level being the major hindrance to their use in real scale applications. Effects of the healing on the mechanical performance of subassemblies are likely to suggest that further healing of the mortar addition may not increase the cohesion value, though positively affecting the friction coefficient.

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REFERENCES