Historic mortars and new repair materials for the Architectural Heritage: Selected Papers from the 5th Historic Mortars Conference (HMC 2019)

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The 5th Historic Mortars Conference was held in Pamplona, at the University of Navarra, Spain, during June 19-21, 2019. HMC 2019 hosted more than 150 scientific communications from the five continents. The underlying theme was the interest in the Historic Mortars from multiple points of view:

- Analysis of ancient materials, either in archaeological sites or in structures of the Architectural Heritage.
- Studies on a wide variety of materials (such as renders and plasters, grouts, masonry materials and coatings) and compositions (earth-based mortars, gypsum plasters, hydraulic mortars, air lime mortars, mortars with pozzolans, natural and Roman cement mortars).
- Research works on historic production, processing and application, including lime technologies.
- Archaeometry studies.
- Conservation issues: diagnosis, decay and damage mechanisms, case studies.
- Preservation and repair materials: consolidation products, new mortars with improved properties, additives and admixtures, use of nanotechnology to obtain highly efficient repair products, preventive conservation.
- Requirements for repair works and materials and adequacy of the testing procedures.

The scientific community has devoted increasing efforts to the mortars of the Built Heritage, as key elements of the architectural structures. The following can be mentioned as the most relevant topics of the current research lines in this field:

- The understanding of the performance of their main components (binding materials, aggregates and, if present, additives and admixtures).
- The elucidation of the old recipes and the methods of application to broaden the historical background on these previous cultures and to support the decision making for preservation.
- The influence of the different factors in the preparation of the mortars/renders/grouts (among them, the ratios and quality of the different raw materials, the ratio of mixing water, the application procedures, the characteristics of the substrates, the specific climatic conditions), for ancient materials as well as for the repair ones.
- The durability of the materials under different surroundings (environmental factors, pollution, weather conditions).
- The methods of testing of the different properties, their reliability and practical implementation (in situ and in lab conditions).
- The design and choice of the most suitable repair materials and the monitoring of their effectiveness.

The present Special Issue of the International Journal of Architectural Heritage includes a set of 12 selected articles, authored by international experts, presented as scientific communications during the conference. The authors have substantially extended and revised the articles that have been peer-reviewed. Altogether the contributions provide an interesting general view of the recent research lines in the field of historic and repair mortars of the Architectural Heritage, the methods of testing and evaluation, the new materials -including nano-structures- to be applied in restoration works and the challenges that should be faced in the near future.

The analysis of ancient materials, mortars and renders, of the Architectural Heritage is a classical issue in this field. The knowledge about ancient techniques, materials and their performance, provides a solid basis for decisions about conservation. In this line, the study of Portuguese stucco marble in buildings from 18th to 20th centuries is the objective of the paper by Freire et al. (2020). 15 stucco samples from different historic buildings, most located in Lisbon and Oporto, such as Paulistas church, Clérigos church and Bijou Manor House, are analysed. The results show that the samples are multiphase gypsum plasters. It is concluded that high temperatures of calcination (above 350 ºC) were used, due to the significant amounts of anhydrite. The use of different animal glues is also confirmed, in good agreement with the ancient recipes of stucco marble. The aesthetical appearance like stone of these materials is also studied, confirming the processes of grinding and polishing, in which the use of beeswax and oils provides glossy characteristics. The high quality and good conservation state of these stucco marbles is ascribed to the use of high purity raw materials, animal glues, high calcination temperatures and a good application technique. All these results provide a solid support to the decision-making of conservation and restoration actions.

Válek et al. (2020) present another study about stucco decorations of the 17th century at Červená Lhota Castle in Southern Bohemia. X-ray Computed Tomography is applied to study the different ornamental pieces of stucco and reveals key information about the inner structure and the method of attachment of the different stucco elements. Matrix-assisted laser desorption/ionization time-of-flight mass (MALDI-TOF) spectrometry and FTIR-ATR inform about the presence and nature of organic compounds. The research work shows that the pieces are made of four elements: a rather heterogeneous core mortar, a surface made of a stucco mortar (thickness 5-25 mm). a fine adhesive mortar layer to attach some small decorative elements and a coarse adhesive mortar to attach the whole piece to the main structure. The core and the adhesive mortars are made of lime-gypsum binder, keeping a very similar proportion of ca. 1:0.7‑0.9:3.6‑4.7 (hydrated lime powder : gypsum : sand by mass. The stucco mortar is made only of lime (hydrated lime or lime putty) and sand. The use of animal glues, probably in percentages below 5% in mass, is proven by the identification of collagen due to its use as a setting retarder for the gypsum-lime binding system. The animal glue is also identified as a part of the coating for the final finishing of the surfaces. In the same vein as that of the work by Freire et al. (2020), the authors underline the critical role played by the high quality of the raw materials for the achievement of durable stucco mortars (in this case, the air lime: absence of under-burnt particles, controlled particle size with small grains and possibly maturation of slaked lime).

Thacker (2020) presents a comprehensive work dealing with medieval cultural landscape. The paper focuses on the construction of Castle Roy and studies the materials of the surviving building structures by petrographic, archaeobotanical and radiocarbon analyses of the mortars. Potential surrounding sources of the materials are also used for comparison purposes. Bayesian techniques are employed to estimate the constructional chronology, which result in dating from late 12th to early 13th century period. The paper provides an accurate landscape analysis, documentary evidences and gives insight in the masonry of the structure by analysing the mortars. A remarkably interesting approach of the interaction between the historic building and the environment and with other historic buildings of the medieval period in Scotland is also described.

Moving from the Medieval Age to the 20th century, the work by Štukovnik et al. (2020) presents an interesting study of historic cement-based mortars used for the Rupnik Military Fortification line in Slovenia. The Rupnik Line was part of the fortified defence system in the border between the Kingdom of Italy and the Kingdom of Yugoslavia, dating from 1920. The bunkers which are the objective of this work were built between 1938 and 1941. Reinforced concrete and special renders are the two main components of these machine-gun nests. The renders were designed mainly to camouflage the military structures. The Portland cement is the main binder and the aggregate is crushed limestone or dolomite rock (locally available). The occurrence of a dedolomitization process, as part of a progressive alkali-dolomite reaction in a real environment, is visible in some samples: the secondary calcite formation (carbonate halo) is identified by microscopic observations. Slag addition (ground granulated blast furnace slag, GGBFS) is responsible for the greenish colour of the rendering mortar, which enables the effective camouflage of the structures. The alternating application of different renders allows the mimicking of the surrounding natural environment.

The techniques and materials for consolidation of the mortars is another topic that has been considered. Michalopoulou et al. (2020) pursue in their article the achievement of this objective by using nanotechnology. The study focuses on four different nanolime dispersions to be used for consolidation of stones and mortars. The approach of the study is to increase the amount of water content above 10% v/v while preserving the colloidal stability of the dispersions and their carbonation potential. The addition of 2-propanol and the use of distilled water enriched with O_2 nanobubbles are the strategies for the achievement of the objectives. The penetration of the dispersions is tested in porous sandstone and in a historic lime mortar. The increase of the ratio of 2-propanol results in the formation of spherical $Ca(OH)_2$ agglomerates, due to the development of Ca-alkoxide phases. An enhancement of the colloidal stability of the dispersions and of their penetration ability is observed. The use of oxygen nanobubbles in dispersions reduces the size of the nanoparticles agglomerates and promotes the development of hexagonal $Ca(OH)_2$ nanoparticles. Despite the limited colloidal stability of O_2 nanobubbles dispersions, they exhibit high penetration ability.

Normand et al. (2020) give an insight in the performance of nanolime, ethyl silicate and acrylic resin for the consolidation of a 14th century wall painting of the Chartres Cathedral, in France. one of these. The work focuses on the consolidation of wet and salt contaminated wall paintings in-situ in a temperate climate. Furthermore, this contribution presents a comparative mid-term assessment of the different products, which display contrasting behaviours. Because the large amount of water in the porous structure of the wall painting, the solvent carriers are non-aqueous (isopropanol and ethanol). The results show that neither the nanolime nor the ethyl silicate improve the cohesion of the paint layer. On the contrary, the acrylic resin increases notably the superficial cohesion according to the cotton-swab test. Concerning the colour or gloss changes, nanolime and ethyl silicate lead to the appearance of a quite persistent white haze. In the case of nanolime, this is ascribed to the superficial carbonation, whereas the possibility of a chemical reaction with pigments or salts of the wall painting is postulated for the ethyl silicate. The acrylic resin gives quite acceptable results, showing no changes over the three months after application. Slight modifications are observed nine months after consolidation, likely due to drier microclimate conditions. Ethyl silicate induces strong hydrophobicity on the surface, which is deemed to be detrimental at long term owing to the fostering of salt crystallization underneath the consolidated layer and the subsequent detachment risk. Despite some opposite results published in the literature, in the specific microclimatic conditions of the Chapter Hall of the Chartres Cathedral, the consolidation with acrylic resin shows the best results at mid-term.

The development of new and improved materials for repair works is other significant research line. The paper by Kyriakou, Rigopoulos and Ioannou (2019) studies the enhancement of the carbonation of lime-based renders by the addition of nano-sized olivine basalt and dolerite quarry waste. The mafic ophiolitic rocks have been proved to be efficient in CO_2 immobilization. The rock materials containing Ca and/or Mg-their silicate minerals react with CO_2 to form carbonate minerals (stable calcite and/or dolomite). Interestingly, the research work investigates the effect after replacement of the binder fraction up to 15%. Carbonation depth profiles, microstructure and mechanical properties of the renders are assessed. In comparison with the plain lime renders, the nano-modified mixtures show an outstanding increase of the carbonation depth, being the ultrafine basalt more effective. The higher specific surface area and the abundance of forsteritic olivine are identified as key factors to enhance the carbonation. In contrast, the compressive strength of the renders is improved only with ultrafine dolerite. Besides the common availability of these rocks, the study will have favourable environmental impact coming from a lime production/consumption decrease.

The article by Porter et al. (2020) presents formulations of microgrouts based on diethyl oxalate with the aim of stabilizing painted lime plasters detached from limestone substrates. Requirements for consolidation grouts for wall paintings are reviewed. Diethyl oxalate reacts with calcium carbonate to form stable and low soluble calcium oxalates. Its great penetration potential and its clear, colourless characteristics increase the interest of this compound. The paper studies the use of a different calcium carbonate aggregate in the formulation, and the effect of the increasing concentration of diethyl oxalate on the binding ability of the microgrout. The results show that the formation of calcium oxalate products is responsible for the microgrout cohesion, although water availability is seen to be imperative for the hydrolysis reaction. Grouts prepared with pure ethanol carrier are not effective. The increase in the diethyl oxalate concentration does not lead to a cohesion improvement because the limited solubility of the diethyl oxalate. Water from the substrate plays an interesting role and contributes to the reaction, allowing different possibilities for the formulation of the grouts.

Speziale et al. (2020) present new multifunctional coatings for protecting stones and lime mortars of the Architectural Heritage. The coatings include a 3D superhydrophobic structure, based on inorganic ceramic oxides, in which photocatalytic nanoparticles are incorporated. The nano-heterostructures are of TiO_2-ZnO in two mass compositions, 10-90 and 50-50, and they are active under solar-light radiation. Different superplasticizers are also added to prevent the nanoparticles from agglomeration, resulting in more effective coatings. Optimized coatings are applied onto limestone, sandstone, granite and air lime mortar. They show good self-cleaning ability (as measured by dye degradation) and a high level of hydrophobicity. Polycarboxylate-based and melamine sulfonate superplasticizers are observed to be the most effective for the coating effectiveness. Accelerated climatic ageing of the treated samples shows that some of the optimized coatings keep a good level of photocatalytic activity and almost no changes in the water repellency, confirming their durability.

The relevance of the nanosized materials is also highlighted by Tsardaka and Stefanidou (2019). In their contribution, they give an insight in the effect of different types of nanomaterials, namely nano-silica, nano-alumina and nano-calcium oxide on the properties of air lime pastes. Specimens with the sole addition of one of these types are prepared (at a maximum of 3 wt. % of binder) and studied after 28 and 90 curing days. The addition of nano-silica promotes the C-S-H formation, increasing the compressive strength of the pastes and giving rise to a denser structure. The main role of nano-alumina is to act as a filler, creating a continuous crystal net at a microstructural level. Remarkably positive results are observed for the combination of nano-alumina and nano-calcium oxide, with improvement of the compressive strength, open porosity, water absorption results and microstructure. This synergistic effect appears as one of the most relevant conclusions of the work. For real application of these combinations, the authors indicate that long-term properties and durability should be further evaluated.

Earth-based plasters and mortars have been used for centuries, from vernacular architecture to monuments. The obtaining of repair materials and the understanding of the role of the clay mineralogy in these mortars is of the utmost importance. These points have been tackled in the contribution by Lima, Faria and Santos Silva (2020). The article describes the preparation of three earth-based mortars, with clayish earth of different mineralogy: illite, kaolinite and montmorillonite. Mortars are prepared with 1:3 volumetric ratio of clayish earth and siliceous sand and a comprehensive characterization of their properties at fresh and hardened state as well is carried out. Two characteristics are extremely important for the practical application of plasters: the drying shrinkage and the adherence. Concerning the former, the appearance of cracks is observed on montmorillonitic mortar, is rare on illitic mortar and absent on kaolinitic mortar. The adherence of the illitic mortar achieves the minimum required values, whereas is significantly low for the other clayish earths. One more advantageous characteristic of the illitic mortar is its better resistance to dry abrasion, which is important for the lifecycle of the plaster. The authors point to the need of further studies to fully understand the factors driving the water vapour adsorption and desorption due to the impact of these cycles to the balance of the relative humidity of the indoor environment of the buildings with these earth-based plasters.

An interesting topic is the reliability of the test methods and their adequacy for binding materials, such as lime, substantially diverse from the cement. In line with the activities of the RILEM Committee 277-LHS: Specifications for testing and evaluation of lime-based repair materials for historic structures, the manuscript by Pachta, Papayianni and Spyriliotis (2020) deals with the correlation between laboratory and field test methods to assess lime-based grouts. Three types of grout mixtures are tested, prepared with lime-pozzolan, lime-pozzolan-white cement and natural hydraulic lime.

The research work evidences that the field results are in good agreement with the equivalent laboratory ones for fresh state properties such as fluidity, penetrability and volume stability. Test methods are defined by relevant standards and literature based on practice. The authors conclude that field tests can be reliably applied on site to adjust the performance of lime-based grouts.

I would like this Special Issue to be interesting for scientists, architects, conservators, restorers, heritage consultants and stakeholders involved in the study and application of mortars of the Architectural Heritage. The readership of the journal will surely take advantage of the vast new knowledge about historic mortars and new materials for repairing.

I would like to highlight the effort and kindness of the two editors of the journal, Professors Pere Roca and Paulo B. Lourenço, for accepting and promoting this special issue. I am very grateful to them. I would like to thank all the authors of the articles and the reviewers, for accepting the invitation to contribute to this Special Issue of the International Journal of Architectural Heritage.

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