First International Conference on Bio-based Building Materials

June 22nd - 24th 2015
Clermont-Ferrand, France

THE "NATURALIME" PROJECT: NEW INSIGHTS INTO THE STUDY OF HEMP-BASED MORTARS FOR NEW CONSTRUCTION AND HERITAGE CONSERVATION

A. Arizzi*, H.Viles

School of Geography and the Environment, University of Oxford, Dyson Perrins Building, South Parks Road, Oxford OX1 3QY, UK

*Corresponding author; e-mail: anna.arizzi@ouce.ox.ac.uk

Abstract

NatuRALiMe (acronym standing for: "Naturally durable: Developing and testing the Resilience of innovative natural Additives for Lime-based conservation Mortars") is a two-year project (2013-2015) funded by the European Union's Seventh Framework Programme (FP7/2007-2013) and being developed at the University of Oxford (UK). The overall aim of this research project is to design suitable and durable mortars made with lime and natural additives (i.e. derivatives of the vegetable and animal world). Interest in studying and using these additives is clearly linked to the need to use sustainable building materials and adopt processes that entail reduced pollution, CO_2 emissions and energy consumption. The first part of this project has been especially focused on the study of hemp-based mortars. Hemp is one of the 10 major primary plants grown as a source of bast fibres and hurds, used for various industrial purposes, such as: textiles, cordage, panelling, biofuel, paper, plastic composites and building materials. The use of hemp hurds in mortars involves important modifications to textural, physical-mechanical and hygrothermal properties compared to those of a typical mortar made with a limestone aggregate. This research project aims to investigate all these inter-related aspects to gain an exhaustive knowledge into this peculiar biomaterial. In particular, new durability tests that consider at the same time temperature and humidity changes, rainfall, salt attack and microbiological growth have been designed to study the resilience of hemp-based mortars under different climatic conditions. The research so far demonstrates the importance of bio-receptivity to hemp mortar durability, something that has not to our knowledge been taken adequately into consideration before. This paper introduces the research work undertaken so far on hemp-based mortars under the *NatuRALiMe* project, aiming to contribute to the knowledge and development of sustainable plant-based building materials.

Keywords:

NatuRALiMe; hemp mortars; microstructure; durability; bio-receptivity.

1 INTRODUCTION

Nowadays the term "sustainability" and its derivatives are used in many different fields of natural and social science. Only five years ago, in the Harvard Business Review, Nidumolu and collaborators wrote that "there's no alternative to sustainable development" and "sustainability is the key driver of innovation" [Nidumolu 2009]. This means that supporting and promoting sustainable technologies not only represents a benefit for the environment but can also generate economic and social development.

As highlighted by Bevan and Woolley [2008], despite strong attention being paid to the use of equipment to reduce energy consumption in buildings, relatively little research has been done on the use of sustainable building materials.

These materials, also called "green", "plant-based", "bio-", "eco-friendly", are intended to improve energy efficiency in buildings, at the same time entailing zero net waste production and low or zero carbon and pollutant emissions during their manufacturing process. Fortunately, the awareness of the importance of developing and improving bio-building materials has increased within the scientific community during the last decade. And it is in within this framework that *NatuRALiMe* has been conceived.

NatuRALiMe is a two-year research project (2013-2015) funded by the European Union's Seventh Framework Programme (FP7/2007-2013) and being developed at the School of Geography and the Environment, University of Oxford (UK), in collaboration with other European research centres, Universities and companies (from United Kingdom, Spain, Italy). The main aim of this research project is reflected in the acronym *NatuRALiMe*, which stands for "Naturally durable: developing and testing the resilience of innovative natural additives for lime-based conservation mortars". The project aims to contribute to the research field of sustainable construction and, as an added value, it also commits to finding sustainable and compatible alternatives for the conservation of architectural heritage.

To fulfil these two inter-related objectives, the project is based on two principles as outlined below.

1.1 The use of lime as a sustainable and compatible material

Lime has been the binder of preference in the majority of construction works undertaken over the last ten thousand years. Examples of lime-based mortars in archaeological sites and ancient buildings include the cities of Yiftah (Israel) and Çatalhöyük (Turkey) (7000-6000 BC) and the Egyptian pyramids (4000-2000 BC). In Europe, the use of lime mortars dates back to Greek and Roman times [Cowper 1927; Henry and Stewart 2011]. Thanks to the use of lime and pozzolans, the Romans were able to build aqueducts, bridges, dams, theatres, amphitheatres, public baths and so on. Many other important examples of European cultural heritage are constructed from masonry, with brick or stone blocks bound together with lime mortar. Furthermore, lime mortars have often been applied as finishing or decorative layers across a surface to provide a protective, smooth and attractive finish.

Unfortunately, with the advent of Portland cement in the 19th century, all the technical and aesthetic benefits embodied in the use of lime have been neglected in exchange for fast setting and low production costs and times. This has favoured the widespread use of Portland cement in all types of construction and, regrettably, also restoration works. The inappropriate use of Portland cement as a repair material in historic buildings has caused the acceleration of decay processes, with final, irreversible damages to our architectural and artistic heritage [Veniale et al. 2003]. Thanks to recent advances in the study of lime mortars the scientific world is managing to convince industry that it is necessary to distinguish between materials according to their use, especially when our historic and artistic heritage is at stake. In this way, the production of "traditional" mortars, especially lime-based ones, is being encouraged because they ensure compatibility, suitability and longevity [Groot et al. 1999; Maurenbrecher 2004].

The use of lime in new construction and repair interventions gives many advantages with respect to Portland cement (Table 1). From the environmental point of view, the production of lime involves much lower burning temperatures compared to those reached during the production of Portland cement, with the consequence that the consumption of fuel and gas emissions are reduced. From the technical point of view, lime allows the masonry to "breathe", thanks to its high permeability to water vapour, and it is a flexible material, because it hardens very slowly. This flexibility reduces the strains between different masonry materials. Moreover, because of its chemical composition, lime does not release harmful substances for the masonry (such as soluble salts). As a summary, the main benefits of the use of lime in place of Portland cement in new construction and repair works are: the reduced carbon footprint and improved energy efficiency, which make lime a sustainable material, and better preservation of the historic masonry, which makes lime a compatible material.

Tab. 1: Main characteristics of limes vs. Portland cement.

Binder	Aerial lime	Hydraulic lime	Portland cement
Common names and types	Dry-hydrated lime (CL, DL) Lime putty or slurry	Natural hydraulic lime (NHL)	Grey or white Portland cement
Burning temperatures	600-900 ºC	900-1050 ºC	>1400 ºC
Setting process	Carbonation	Carbonation and hydration	Hydration
Setting speed	Low	Medium	High
Shrinkage	High	Medium	High
Flexibility	High	Medium	Low
Permeability to water vapour	High	Medium	Very low
Porosity	High	Medium	Low
Soluble salts	-	-	Ettringite, thaumasite, etc.

1.2 The use of natural additives in mortars: the hemp hurds

Natural additives are derivatives of the vegetable or animal world. Their use in mortars is an ancient practice in the field of construction, as indicated by Vitruvius in De Archirectura libri decem (15 BC). Evidence exists for the addition to lime of proteinacious substances such as blood, hair, straw, milk, eggs etc. and sugar-based gums and animal glues etc. [Cowper 1927; Sickels 1981; Garate Rojas 1993; RILEM TC 203-RHM 2009] to improve their workability, waterproofing, mechanical resistance and durability. Unfortunately, records of the dosages no longer exist and neither are their effects on ancient mortars fully understood.

Nowadays additives are essential components of building materials and masons greatly appreciate their use [Hendrickx, 2009]. Nevertheless, their composition, function and characteristics have substantially changed over time since they have had to adapt not only to modern construction techniques but also to the new requirements demanded of industrial mortars. Recently, a number of researchers have investigated the advantages and disadvantages of the use of natural additives in lime mortars [Ruiz-Agudo and Rodríguez-Navarro 2008; Izaguirre et al. 2011; Ventolà et al. 2011].

Hemp hurds as lightweight aggregate in mortars

Hemp is an annual non-food crop, i.e. cultivated for applications outside the food sector, such as bio-fuels, packaging, pharmaceutical products, automobiles and building materials, among others. It can be grown in a wide range of soils without the need for pesticides and fertilisers, and it is useful to grow in rotation with food crops, since it improves the soil, releasing nutrients and reducing weeds [Bevan and Woolley 2008]. Today, 99% of the plant is used, mainly the bast fibres (i.e. fibres obtained from the exterior part of the stem used for cordage, textiles, etc.) [Faruk et al. 2012], the seeds (with high nutritional value) and the hurd (i.e. inner woody part of the stem, also called shiv, Fig. 1).

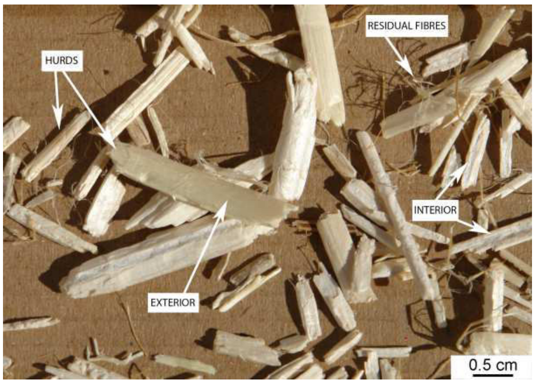

Fig. 1: Hemp hurds used for mortar production (from [Arizzi et al. 2015a]).

In particular, hemp hurds mixed with lime produce a peculiar bio-composite (in which the hemp hurds act as a lightweight aggregate) that is being increasingly used in construction, in a number of applications (such as roof, floor and wall insulation) [Bevan and Woolley 2008]. And, depending on the mix design and the final application, it can be sprayed or even cast into blocks.

In the last few years, new small-scale companies that produce hemp-lime mortars have arisen throughout Europe. Some examples of their commercial products are: Hemcrete® (Hemcore, UK), NaturalBeton® (Equilibrium, Italy), Cannabric® (Cannabric, Spain). The reason for the growing interest in hemp lime mortars is twofold: firstly, hemp is a fast-growing, renewable and carbon sequestrating plant [Pervaiz and Sain 2003] and secondly, hemp-lime mix shows good thermal and acoustic insulating properties [Newman 2008], high breathability and flexibility, zero shrinkage and is effective in regulating relative humidity in the building [Bevan and Woolley 2008].

All the characteristics listed above make hemp-lime mortar a very efficient sustainable building material, worthy of consideration in the *NatuRALiMe* project. Our research on hemp-lime mortars is specially focused at understanding:

- the interactions at the micro-scale between the lime and the hemp hurds during mortar hardening;
- the water transfer processes within the hemp-lime mix;
- the durability and bio-receptivity of hemp-lime mixes under different climatic conditions.

2 PROPOSED TASKS AND METHODOLOGY

As commented above, a number of hemp-lime mixes exist on the market and many more are about to be launched. Our task, as researchers, is to assess these materials, investigating their characteristics and properties, with the final aim to bring light to unanswered questions and provide specific solutions that improve the material performance on site.

It is worth highlighting that hemp-lime mortar properties are far from those of a typical mortar made with lime and a stone aggregate, because they are composed of a natural organic material with very different chemical and physical characteristics to inorganic sand. Hence, the study of hemp-lime mixes can be difficult when using conventional methods. For this reason, the research carried out on hemp-lime mixes within the *NatuRALiMe* project undertakes two different but complementary methodological paths: on the one side, the application of conventional analytical techniques and test methods, so as to enable a comparison between hemp mortars and traditional ones and, on the other side, the development of new tests, specifically conceived for such a novel and peculiar material. The following sections describe the experimental work plan designed for each of the tasks proposed.

2.1 Task 1: The characterisation of mortar components

Objective 1: Studying the chemical, mineralogical and textural characteristics of the limes and the hemp hurds.

Three types of limes (aerial lime in the form of dry powder (CL90S); aerial lime in the form of putty (CL90S PL); natural hydraulic lime (NHL3.5) [BS EN 459-1 2002]) and dry hemp hurds were used for mortar production. These materials were studied by means of the following techniques:

- X-ray diffraction (XRD) and thermal analysis (TGA), to determine the mineral phases of limes;
- Nitrogen Adsorption (NA) and application of the BET method [Brunauer et al. 1938], to determine the specific surface area of limes;
- LASER granulometry, to determine the particle size distribution of limes.
- Field Emission Scanning Electron Microscopy (FESEM), to observe the microstructure of limes;
- Environmental Scanning Electron Microscopy (ESEM), to observe the microstructure of the hemp hurds;
- Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy (ATR FT-IR), to determine the functional groups constituting the carbohydrates of the hemp hurds.

2.2 Task 2: The preparation of hemp-lime mixes and their physical characterisation

Objective 2: Studying the hydric performance of hemp mortars in the hardening state so as to compare them with those of typical lime-aggregate mortars.

Different types of mixes were prepared with a fixed lime:hemp:water dosage by vol. and then cast in standardised moulds (4×4×16 cm^3). Samples were studied during the first month and after three months of curing under average conditions of T = 17 ºC and RH = 75%. The hydric properties of the mixes were characterised following European standards: water absorption [UNI-EN 13755 2008] and drying [NORMAL 29/88 1988]; capillary uptake [UNI-EN 1925 2000]; permeability to water vapour [NORMAL 21-85 1985].

2.3 Task 3: The study of the hardening of lime in the presence of hemp

Objective 3: Studying the hardening processes of lime in hemp-lime mixes, to understand whether and how curing is delayed by the presence of hemp hurds.

This study is still in progress and includes the following investigations:

- A mineralogical and textural study of the limes in the mixes, by means of XRD, TGA, FESEM and ESEM;
- A chemical study of the hemp hurds, by means of solid-state Nuclear Magnetic Resonance (NMR).

2.4 Task 4: The durability study of the hemp-lime mixes

Objective 4: Studying hemp mortar behaviour under real environmental conditions in order to evaluate the advantages and disadvantages of the use of hemp-based building materials in specific areas.

The durability study of the hemp-lime mixes was carried out by means of new accelerated weathering tests specifically designed for this bio-material. Tests were run in a Sanyo-FE 300H/MP/R20 environmental cabinet where temperature and relative humidity variations, rainfall and salt attack can all be simulated. Hemp mortars were subjected to three main climates: Mediterranean, Tropical and Semi-arid. For each climate, both inland and coastal areas were simulated (for the latter the effects of airborne salt were added).

A study of the macroscopic and microscopic modifications of the hemp-lime mixes, caused by exposure to environmental factors, was carried out by comparing control, mid-experiment, and post-experiment samples:

- Mass and volume variations were recorded;
- Colour changes were followed by means of a spectrophotometer, following the European standard [UNE EN 15886 2011];
- XRD analysis were performed to determine changes in the mineral phases and the degree of carbonation;
- ESEM observations were performed to determine changes in mortar texture and mineral phase morphology;
- A microbiological study was performed to determine the growth of micro-organisms.

3 MAIN RESULTS

3.1 The chemical, mineralogical and morphological characteristics of hemp-lime mixes

The results of this study have been recently published [Arizzi et al. 2015a] and are summarised below.

By means of microscopical investigations, we found that the mortar matrix does not fill the large pores and channels constituting the hemp hurds and only superficial deposition can be observed (Fig. 2). Although this is beneficial for the insulating properties of hemp, poor adhesion was found between the matrix and the surface of the hemp hurds. The mineralogical study of the hemp-lime mixes revealed that limes show a delayed hardening in the presence of hemp, with large amounts of portlandite and calcium silicates found in aerial and natural hydraulic lime mixes, respectively, after three months of curing. Moreover, an unusually high amount of vaterite was also found in all mixes after three months, demonstrating a delayed transformation of this metastable polymorph into the stable one (i.e. calcite). The reason is inferred to be an insufficient amount of water available to the matrix of the mixes from the start. This assumption is supported by the ability of hemp hurds to absorb large amounts of water over a long time.

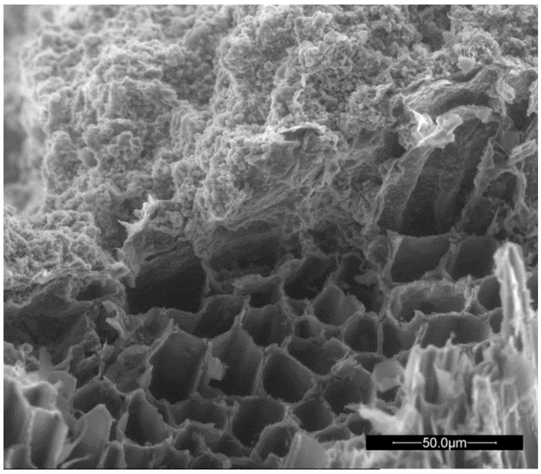

Fig. 2: ESEM image of the contact zone between hemp hurds and matrix in a hemp-lime mix.

3.2 The hydric behaviour of hemp-lime mixes

This part of the research on hemp-lime mixes has also been recently published [Arizzi et al. 2015b] and is summarised below.

Due to the presence of hemp hurds, mortars absorbed a much greater amount of water compared to a generic mortar made with lime and a stone aggregate. However, the hurds did not slow down the drying rate of the mixes. By studying the water absorption and desorption behaviour of a hemp hurd at micro-scale, it has been demonstrated that the hurd remains enlarged after drying and this increases its capacity for absorbing water. This explains why the channels and pores constituting the hurd act as a nearly unlimited water reservoir.

Mortars made with natural hydraulic lime (NHL 3.5) showed the highest transpirability and drying rate and the lowest water absorption by immersion and capillary uptake. Moreover, NHL 3.5 mortars showed less intense bio-deterioration compared to the aerial limes. Although it is believed that lime protects hemp from biological decay [Bevan and Woolley 2008], hemp-lime mortars are subject to microbial growth (especially alkaliphilic microbes and fungi), more or less intensely depending on the environmental conditions (e.g. occurrence of dry and wet zones in the mortars, Fig. 3).

Fig. 3: Example of fungi growth on the surface of hemp-lime mortars subjected to intense wet-dry cycles.

3.3 The durability of hemp-lime mixes

Hemp mortar samples did not show any apparent damage during or at the end of the durability test and salt efflorescence was hindered by washing during simulated rainfall events. Furthermore, the cyclic conditions of temperature, relative humidity and rainfall during the durability tests induced an improvement of mortar hardening, compared to control samples.

Since the growth of microorganisms is strongly influenced by conditions of temperature and relative humidity, bio-receptivity of hemp-lime mortars has been especially examined during the durability tests. The presence of nutrients (organic matter proceeding from hemp) and pre-existent microorganisms (bacteria) in the material substrate, as well as variable environmental conditions and the presence of salt were all determining factors of bio-colonization of the hemp mortars. Natural hydraulic lime (NHL 3.5) was found to be the most suitable for use in combination with hemp hurds, since it slightly reduced the water absorption compared to the aerial lime and also ensured reduced chromatic change and bio-decay.

4 SUMMARY

The ongoing studies on hemp-lime mortars carried out under the *NatuRALiMe* project are providing new insights into the characteristics and performances of this bio-building material.

We have highlighted that the interactions between hemp, an organic component, and lime, an inorganic material, influence the normal processes of mortar hardening. Ongoing investigations on hemp-lime mixes made with fresh hurds and with other elaboration methods and curing conditions are being undertaken to investigate which process is the important control on the delayed hardening of lime in the presence of hemp. This may be a physical process (great absorption of water by the hemp) or a chemical interaction (between organic compounds of the hemp and lime). NMR studies are also being carried out to understand the effect of pH and carbonation on the chemical composition of the hemp hurds.

Tests on the hydric performances of hemp-lime mixes have shown the high water absorption ability of hemp hurds, which has been explained by the irreversible swelling behaviour of hemp hurds at the micro-scale. The hydric tests have also highlighted the potentially high bio-receptivity of hemp-lime mixes, despite the presence of lime that is generally believed to be a strong disinfectant. A realistic durability study has also shown that, using the correct binder (natural hydraulic lime) and taking the necessary preventive measures to reduce microbial growth, hemp-based mortars should be suitable for use in inland and coastal areas under a range of different climatic conditions.

5 ACKNOWLEDGEMENTS

The research leading to these results has received funding from the European Union's Seventh Framework Programme [FP7/2007-2013] under grant agreement n° 326983 [NatuRALiMe]. We are grateful to the people who have collaborated and still collaborate in this project: Prof. Giuseppe Cultrone, Dept. of Mineralogy and Petrology, University of Granada (Spain); Prof. Inés Martín Sánchez, Dept. of Microbiology, University of Granada (Spain); Arch. Monika Brümmer, Cannabric company (Spain); Dr. Donatella Capitani, CNR Rome (Italy). Part of the study carried out on hemp mortars has also been funded by the Spanish research project MAT-2012-34473.

6 REFERENCES

[Arizzi et al. 2015a] Arizzi, A.; Cultrone, G.; Brümmer, M.; Viles H. A chemical, morphological and mineralogical study on the interaction between hemp hurds and aerial and hydraulic lime particles: implications for mortar manufacturing. Construction and Building Materials, 2015, 75, 375-384.

[Arizzi et al. 2015b] Arizzi, A.; Brümmer, M.; Martín-Sanchez, I.; Cultrone, G.; Viles H. Assessing the hydric performances and bio-receptivity of mortars made with lime and hemp for their application in sustainable new construction and repair works. PlosOne, DOI: 10.1371/journal.pone.0125520, 2015.

[Bevan and Woolley 2008] Bevan, R.; Woolley, T. Hemp lime construction. A guide to building with hemp lime composites. HIS BRE Press, Bracknell, UK, 2008. ISBN: 978-1-84806-033-3.

[Brunauer et al. 1938] Brunauer, S.; Emmett, P.H.; Teller, J. Adsorption of gases in multimolecular layers. Journal of American Chemical Society, 1938, 60, 309.

[BS EN 459-1 2002] BS EN 459-1. Description Building lime - Part 1: Definitions, specifications and conformity criteria. British Standards, 2002, London, UK.

[BS EN 1015-11 1999]. BS EN 1015-11. Methods of test for mortar for masonry. Part 11: Determination of flexural and compressive strength of hardened mortar. British Standards, 1999, London, UK.

[Cowper 1927] Cowper, A.D. Lime and Lime mortars. Donhead Ed., 1927. Reprinted by Building Research Establishment Ltd., 1998.

[Faruk et al. 2012] Faruk, O.; Bledzki, A.K.; Fink, H.-P.; Sain, M. Biocomposites reinforced with natural fibres: 2000-2010. Progress in Polymer Science, 2012, 37, 1552-1596.

[Garate Rojas 1993] Garate Rojas, E. Las Artes de la Cal. CBRIC-MEC, Madrid, 1993.

[Groot et al. 1999] Groot, C.; Bartos, P.; Hughes J. Historic mortars: characteristics and tests - Concluding summary and state-of-the-art. International Workshop on Historic Mortars, Paisley, UK, 1999, 443-454.

[Hendrickx 2009] Hendrickx, R. The adequate measurement of the workability of masonry mortar. PhD Thesis, Katholieke Universiteit of Leuven, Belgium, 2009.

[Henry and Stewart 2011] Henry, A.; Stewart, J. Mortars, renders and plasters. English Heritage, Practical Building Conservation. Eds. Martin B, Wood C. Ashgate Publishing Limited, Farnham (UK), 2011.

[Izaguirre et al. 2011] Izaguirre, A.; Lanas, J.; Alvarez, J.I. Effect of a biodegradable natural polymer on the properties of hardened lime-based mortars. Materiales de construccion, 2011, 61, 302, 257-274.

[Maurenbrecher 2004] Maurenbrecher, A.H.P. Mortars for repair of traditional masonry. Practice Periodical on Structural Design and Construction, 2004, 9, 62-65.

[Newman 2008] Newman, G. European decortication and fibre make. Biomaterials-back to the future. Plant fibre technology, 2008. http://www.compositesinnovation.ca/biofibre_workshop/, accessed on January 2014.

[Nidumolu 2009] Nidumolu, R.; Prahalad, C.K.; Rangaswami, M.R. Why sustainability is now the key driver of innovation?. Harvard Business Review, https://hbr.org/2009/09/why-sustainability-is-now-the-key-driver-of-innovation, accessed on 15 January 2015.

[NORMAL 29/88 1988]. NORMAL 29/88. Measurement of the drying index. CNR-ICR, Rome, 1988.

[NORMAL 21-85 1985] NORMAL 21/85. Permeability to water vapour. CNR-ICR, Rome, 1985.

[Pervaiz and Sain 2003] Pervaiz, M.; Sain, M.M. Carbon storage potential in natural fiber composites. Resources, Conservation and Recycling, 2003, 39, 325-340.

[RILEM TC 203-RHM 2009] RILEM TC 203-RHM. Repair mortars for historic masonry. Materials and Structures, 2009, 42, 853-865.

[Ruiz-Agudo and Rodríguez-Navarro 2008] Ruiz-Agudo, E.; Rodríguez-Navarro, C. Effects of additives on lime putty rheology: applications in the design of mortars for conservation purposes. Proceedings of the 1st Historic Mortars conference, Lisbon, 2008.

[Sickels 1981] Sickels, L.B. Organic additives in mortars. Edinburgh Architecture Research, 1981, 8, 7-20.

[UNE EN 15886 2011] UNE-EN 15886. Conservation of cultural property - Test methods - Colour measurement of surfaces. Spanish Association for Standardization and Certification, Madrid, 2011.

[UNI EN 1925 2000]. UNI EN 1925. Natural stone test methods — Determination of water absorption by capillarity. CNR-ICR, Rome, 2000.

[UNI EN 13755 2008]. UNI EN 13755. Natural stone test methods — Determination of water absorption at atmospheric pressure. CNR-ICR, Rome, 2008.

[Veniale et al. 2003] Veniale, F.; Setti, M.; Rodríguez-Navarro, C.; Lodola, S.; Palestra, W.; Busetto, A. Thaumasite as decay product of cement mortar in brick masonry of a church near Venice. Cement and Concrete Composites, 2003, 25, 1123-1129.

[Ventolà et al., 2011] Ventolà, L.; Vendrell, M.; Giraldez, P.; Merino, L. Traditional organic additives improve lime mortars: new old materials for restoration and building natural stone fabrics. Construction and Building Materials, 2011, 25, 8, 3313-3318.