Materials and Structures (2023) 56:106
https://doi.org/10.1617/s11527-023-02175-z

RILEM TC REPORT

RILEM TC 277-LHS report: additives and admixtures for modern lime-based mortars

Pagona-Noni Maravelaki · Kali Kapetanaki · Ioanna Papayianni · Ioannis Ioannou · Paulina Faria · Jose Alvarez · Maria Stefanidou · Cristiana Nunes · Magdalini Theodoridou · Liberato Ferrara · Lucia Toniolo

Received: 7 November 2022 / Accepted: 15 April 2023 / Published online: 5 June 2023

Abstract The scope of this collective paper produced in the frame of RILEM TC 277-LHS is to provide sound knowledge on the use of additives/admixtures in lime-based mortars, based on literature and practice. The most widely known additives/admixtures are systematically presented. Their main

---

This review was prepared by the authors within RILEM TC 277-LHS ''Specifications for testing and evaluation of lime-based repair materials for historic Structures'' and further reviewed and approved by all members of the RILEM TC 277-LHS.

This review was prepared by Pagona-Noni Maravelaki, Kali Kapetanaki, Ioanna Papayianni, Ioannis Ioannou, Paulina Faria, Jose Alvarez, Maria Stefanidou, Cristiana Nunes, Magdalini Theodoridou, Liberato Ferrara, Lucia Toniolo within RILEM TC 277-LHS ''Specifications for testing and evaluation of lime-based repair materials for historic Structures'' and further reviewed and approved by all members of the RILEM TC 277-LHS.

Chair: Emer. Prof. Ioanna Papayianni.

Deputy Chair: Dr. Jan Válek.

TC Members: Jose I. Alvarez, Anna Arizzi, Beril Bicer-Simsir, Violeta Bokan-Bosiljkov, Arnaldo M. Carneiro, Claudia Brito De Carvalho, Paulina Faria, Liberato Ferrara, Caspar Groot, Davide Gulotta, John J. Hughes, Ioannis Ioannou, Kali Kapetanaki, Heejeong Kim, Loucas Kyriakou, Cristiana Lara Nunes, Pagona-Noni Maravelaki, Sagrario Martinez-Ramirez, Didem Oktay, Vasiliki Pachta, Andreja Padovnik, Ioanna Papayianni, Chiara Pasian, Sara Pavia, Giovanni Pesce, Ulrike Peter, Meera Ramesh, Divya Rani, Sriram Pradeep Saridhe, Michele Secco, Maria Stefanidou, Petra Stukovnik, Cristina Tedeschi, Magdalini Theodoridou, Lucia Toniolo, Jan Valek, Rob van Hees, Maria Rosario Veiga.

P.-N. Maravelaki (✉) · K. Kapetanaki
Technical University of Crete, Chania, Greece
e-mail: pmaravelaki@tuc.gr

I. Papayianni · M. Stefanidou
School of Civil Engineering, Aristotle University of Thessaloniki, Thessalonikes, Greece

I. Ioannou
Faculty of Engineering, University of Cyprus, Nicosia, Cyprus

P. Faria
Faculty of Science and Technology, NOVA University of Lisbon, Lisbon, Portugal

J. Alvarez
University of Pamplona, Pamplona, Spain

C. Nunes
Institute of Theoretical and Applied Mechanics, Czech Academy of Sciences, Prague, Czech Republic

M. Theodoridou
Newcastle University, Newcastle, UK

L. Ferrara · L. Toniolo
Politecnico di Milano, Milano, Italy

effects and testing of their performance have been properly tabulated. It is well known that a plethora of additives/admixtures are produced every year by chemical industries. However, when using them in lime-based mortars, compatibility and durability aspects are of primary importance. The introduction of additives/admixtures in lime mortars was imposed by the need to improve important properties of these composites in the fresh and hardened state, namely, workability, durability, early-age and long-term strength and to reduce defects, such as shrinkage and long setting time. In this review paper, the terminology proposed by EN 16572 is followed, designating additive as a constituent added in small quantity to the binder, and admixture as a substance in quantities at least 1% w/w added to the mix. The additives/admixtures are classified according to their action and their validation with specific testing methodologies highlights the dosage sensitivity and the need to develop further standardization. The combination of different additives proposed in several studies resulted as the most promising strategy to enhance the performance of lime mortars. However, recently developed additives and admixtures need to be further evaluated with reference to their compatibility with other mortar constituents, and their effects on the overall mortar and render durability need to be studied. Finally, adopting similar terminology for additives/admixtures in lime and cement-based mortars will facilitate better comparison and assessment issues.

Keywords Additives · Admixture · Lime mortars · Performance · Evaluation

1 Introduction

1.1 Definitions

The terms additives and admixtures (henceforth adds and adms) refer to ingredients that are added in mixtures where cement, lime, gypsum and clay are used as binders, with the aim of improving the fresh and hardened state physico-mechanical properties and the durability of mortars. According to definitions given in EN 16572 (2015): "additive (EN) or addition (ASTM) is a constituent usually added in small quantity to the *binder* to modify its manufacture or properties (for example accelerators, plasticizers, water repellents and air-entraining agents), whereas admixture is a substance other than the binder, aggregate or water, added in quantities of at least 1% weight/weight (wt/wt) to the *mix* to alter its properties" [1]. The same EN further denotes that: "pigments, pozzolana (as long as it is added in small quantities and not as a latent binder) and fibrous substances may be classified as admixtures". Therefore, based on EN 16572 (2015), an additive is added to the binder system (consider the case of factory-made binders), whereas admixtures are added to the mixture. It should be also clarified that, in many cases of lime-based mortars, pozzolans and cement act as latent binders. Additives and admixtures intend to enhance and ameliorate specific properties of the lime mortars, such as workability, durability, or early-age and final strength, and to reduce defects, such as shrinkage and long setting time. Nowadays, the increasing use of waste and by-product adds/adms in mortars may also decrease the embodied energy of the end-product.

In this review, the definitions of EN 16572 (2015) stated above will be adopted, even though there is a contradiction with terms frequently used in literature and cement technology. In the literature, the specification between additives/admixture derives from the terminology used in concrete technology: "admixtures are ingredients added to the concrete batch immediately before or during mixing" [2]. Similar definition can be found in ACI 116R-90 (ACI [3]) and in EN 934 (EN 934–2:2009 + A1:2012) for Cement and Concrete Terminology. In this case, the term "addition" is defined as a material that is inter-ground or blended in limited amounts into a hydraulic cement during manufacture, either as a "processing addition" to aid in manufacturing and handling the cement, or as a "functional addition" to modify the properties of the finished product [3, 4].

1.2 State of the art

The literature highlights the use of natural additives in mortar production throughout the centuries and all over the world [5, 6]. More specifically, Sickels [5], reported that analyses of old mortars, plasters, renders and other ancient materials, carried out in recent decades, provided insights into the use and composition of organic admixtures and their contribution to the performance of the end-product. Neuburger [6] discovered that substances of natural and organic origin, such as Arabic gum or tragacanth, animal glue from Rhodes, the blood of hippopotamus and the milky juice of figs mixed with egg yolk, all served as adhesives or binding substances. Other natural organic polymers like egg albumen, keratin, and casein were used in Egyptian artefacts as common binders [7]. In Vitruvius time, fig juice, rye dough, hogs' lard, curdled milk, blood and egg whites were employed to toughen and regulate mortar setting performance [8]. Blood and egg whites have also been used to retard the setting time of mortars, along with sugar. Different inclusions, either in fibrous form or as aggregates, were used in old mortars of different technology throughout history by researchers, aiming always to improve the properties of the end-product [9].

In the past decades, research on synthetic adds/adms was limited to concrete and cement-based mortars, but recently the use of traditional and modern adds/adms has been successfully extended to applications in lime-based mortars [10–12]. Lime-based mortars may have different applications, such as plasters and renders EN 998–1 [13], masonry bedding mortars EN 998–2 [14] or repointing mortars. The distinct functional roles of the previously mentioned lime-based mortars also involve different fresh- and hardened-state performance. In the surface of plastering/rendering lime mortar shrinkage and microcracking occurred that can be mitigated by applying hand and mechanical tools as the carbonation and hardening are evolving, thus eliminating the cracks and improving the coating overall performance and durability [15]. For plastering and rendering it is mandatory the mortars to have a good workability to ensure the applicability on relatively thin layers (in general from 0.5 to 2 cm), a good adherence to the substrate and between layers, but also a reduced final cracking. In the case of rendering mortars, the durability in the presence of the water is more important compared with mortars for plastering. The latter should present mechanical resistance compatible with the substrate, whereas for the bedding mortars adequate resistance to the compressive action of the masonry should be fulfilled. Adds/adms, vernacular and innovative, may provide a significative contribution for air lime-based mortars to fulfill their different requirements both in fresh and hardened state.

Nowadays, biopolymers are continuously used in the architectural sector of several countries, inspired from natural additives, which were identified with the aid of analytical techniques in traditional mortars which proved in the field their excellent durability and long-term performance [16]. It is worth remarking that most natural additives above-mentioned are mostly "edible" products, the use of which is no longer recommended. Therefore, efforts should be concentrated on the use of wastes and by-products along with synthetic materials to provide the same positive effects, thereby reducing the consumption of raw materials that are needed to feed humans and animals.

With the aim of further enhancing the performance of lime mortars, specifically expressed by low compressive strength, prolonged hardening time, low resistance to moisture, the use of adds/admsbecomes essential to take advantage of the benefits of these composite materials both in restoration and contemporary projects. There are several types of adds/adms, which may be classified according to their action, composition and morphology.

Regarding the lime-based mortars entailed for the repair of historic structures, compatibility with original material should be taken into account. In technical terms, compatibility of a repair mortar is governed by a list of characteristics [17]. This means no direct or indirect damage to the original material and ensuring the long-term stability of the intervention. Therefore, any assessment on the effectiveness of adds/adms in lime-based repair mortars for historic structures should also consider physico-chemical compatibility requirements [18–20].

In the following sections, sound knowledge concerning practice and research advances on the use of admixtures/additives will be presented and their benefits and drawbacks on the performance of lime mortars will be discussed. In particular, the most important properties for assessing the additive/admixture performance, with the aid of specific testing methodologies, are presented.

2 Classification of additives according to their action

The classification of additives according to their action is considered as the first step in an attempt to clarify the significance of introducing these materials in the design of mortars. Afterwards, the evaluation methods used for the introduction of additives in lime mortars

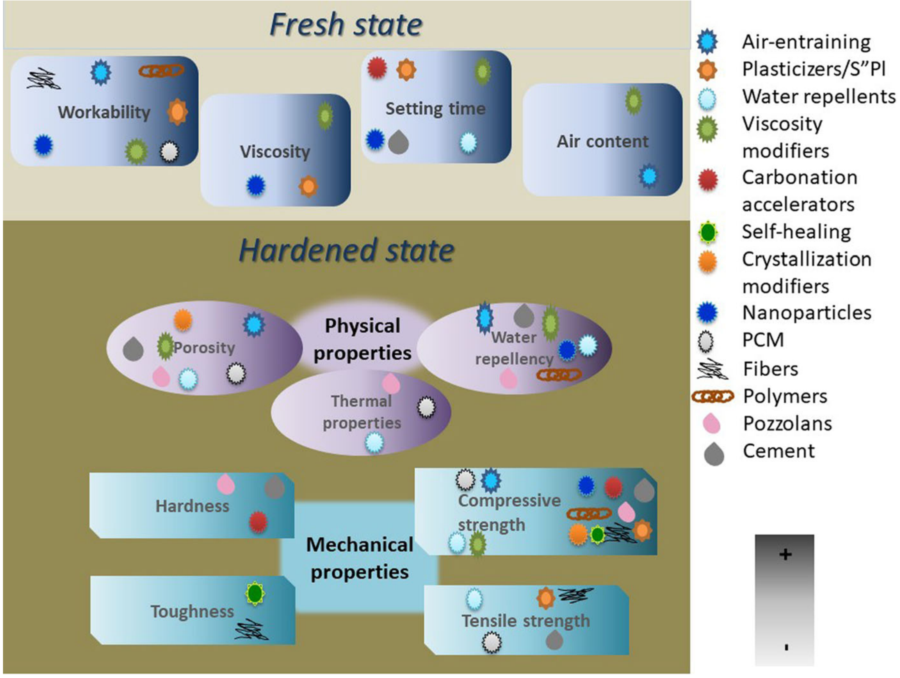

Fig. 1 Effect of additives/admixtures on various properties of lime-based mortars (The term nanoparticles refer to inorganic nanoparticles and the term polymers to natural and polymeric compounds)

and their contribution to the performance of the composite material are discussed and summarized at the end of almost each section. Furthermore, Fig. 1 highlights the action of each additive/ admixture in the fresh and hardened state properties. The scaling bar denoted by (+) and (−), with dark and light shading, respectively, refers to increasing and decreasing performance of each property.

2.1 Air-entraining agents

Air-entraining additives are organic substances that allow a controlled number of air bubbles to be incorporated in the fresh and hardened state of mortars. In the literature, various research works explicitly investigated the role of air-entraining agents in the setting and hardening of lime mortars. An air-entraining additive improves mortar workability through the formation of air bubbles, which are incorporated into the mortar and become part of the matrix that binds the aggregates together in the hardened state. These small air bubbles, which are produced in a controlled quantity, are uniformly distributed, and incorporated in the mortar during mixing and remain there after hardening. The molecule of an air-entraining agent is composed of a hydrophilic and a hydrophobic group. In an aqueous medium, considering that air is trapped inside the fresh mortar, these molecules orient themselves at the air–water interface, so that the hydrophilic end is in contact with the water and the hydrophobic one is outside it, thus forming stable spherical voids [21].

It has been demonstrated that the introduction of air bubbles dispersed uniformly through the mortar paste also increases total mortar porosity (closed and open porosity), thus minimizing damage caused by freezing in the masonry [21]. As the total porosity increase is generally associated with a decrease in the mechanical strength, care should be taken in the amount of the air-entraining agent added, so that the resistance of the mortars to external stress and weathering shall not be compromised. In addition, an air-entraining agent may assist in prolonging the service life of lime-based mortars subjected to salt crystallization.

The improvement of mortar workability via the formation of air bubbles could influence the textural quality and carbonation [22]. Cultrone [23] stated that the air-entraining agent Sikanol-MR (Sika, S.L) significantly altered the texture of the mortars by creating rounded pores and eliminating or reducing the

Table 1 Summary of studies for the use of air-entraining agents in lime-based mortars

Binder composition	Additive	% wt.	References	Purpose of use
CL + BSL or OPC	Air-entraining agent	0.05	Cerulli et al. [22]	Improvement of:
CL CL + NP	Sikanol-MR (Sika, S.L)	0.10	Cultrone et al. [23]	Workability
CL	Silipon –Aqualon	0.05–0.15	Seabra et al. [26]	Freeze–thaw resistance
CL	Sodium dodecyl sulfate (SDS)	0.01–0.05*	Silva et al. [25]	Salt decay resistance
CL	Sodium alfa olefin sulfonate (AOS)	0.005–0.020*		
CL	Cocamidopropyl betaine (CAPB)	0.01–0.50*		

CL: Calcitic lime, NP: Natural pozzolan, % addition: refers to % wt. addition to binder, BSL: Blast furnace slag, OPC: Ordinary portland cement

*% addition to the total dry weight of mortar (binder + aggregate)

drying cracks, but it seems that this had no effect on the carbonation process [23].

Surfactants are synthetic or natural molecules that can act as air-entraining compounds. Due to their bipolar nature, exhibiting a non-polar part usually oriented towards the aerial medium, and a polar fragment anchored in the lime matrix, the surfactants could stabilize the air bubbles inside the mortar [24].

Silva et al. [25] studied three different surfactants: sodium dodecyl sulfate (SDS), sodium alfa olefin sulfonate (AOS) and cocamidopropyl betaine (CAPB) [25]. The latter was the most effective air-entraining agent due to its amphoteric nature, but, at the same time, increased the porosity and pore diameter (pores from 4 to 500 microns), thus causing a dramatic drop in the mechanical resistance of the end-product. As opposed to that, SDS and AOS, which are anionic surfactants, increased pores in the range of 4–40 microns, leading to moderate changes in compressive strength, and positively affecting the hygric properties by reducing the capillary water absorption.

Air-entraining agents have been studied in lime and lime-pozzolan mortars. Results showed that those air-entraining agents do not affect carbonation, provide higher pores and permeability, but decrease the mechanical properties, compared to the pure lime mixtures [23, 26]. These agents could be used to increase mortar durability towards temperature stress and salt crystallization.

Table 1 classifies the studies of the literature for the use and purpose of air-entraining agents in lime-based mortars taking into account the binder composition, type and quantity of agent added.

2.2 Plasticizers-superplasticizers

Plasticizers or superplasticizers are polymers of low-molecular-weight in comparison to conventional ones and can reduce the water needed to achieve the desired mortar workability, without affecting its consistency. As a result, fluidity and workability, at a constant water/binder ratio, are improved with the use of plasticizers or superplasticizers, giving rise to higher mechanical strength and resistance to shrinkage. The chemical composition of plasticizers and superplasticizers comprises groups of lignosulphonates (anionic surfactants), polyglycol esters and carbohydrates [26].

The mechanism of action of plasticizers and superplasticizers relies on the reduction of the surface tension of water, as well as the electrostatic repulsion between the binder particles, as a result of the orientation of the additive molecule in the interface liquid–solid, which prevents the adsorption of water molecules on the surface of the binder particles, thus increasing the free water in the system. The influence of poly-parameters on the action of plasticizers is evidenced in several works referring to the water to lime ratio, the quantity and nature of admixture, such as pozzolans, cement, etc. Generally, the use of plasticizers improves the stability, water retention and rheological behavior of the material in the fresh state [26–29]. Indeed, according to Seabra et al. [26], the plasticizer or water-reducing agent Peramin SMF–Perstorp, even in small amounts, changed the rheological behavior of the fresh composite, as confirmed by the diminishing torque values due to the increase of free water in the system [26]. Polynaphthalene sulfonate (PNS), lignosulfonate (LS) and condensate of melamine–formaldehyde sulfonate (SMFC) plasticizers were found to be less effective than polycarboxylated ethers (PCE). Another drawback of sulfonates is related to the introduction of sulfate ions into the mortar mixture. PNS showed higher dispersing effect than LS on account of its higher adsorption onto portlandite, C-S-H, C-S-A-H, and C-A-H particles [28]. Plasticizers increase the compressive strength of mortars and may eventually reduce the setting time, depending on the dosage, without affecting the morphology of the composite. Therefore, plasticizers could be used in mortars that should reach high mechanical properties [26, 27, 29].

On the other hand, superplasticizers, without affecting the consistency, simultaneously permit a high reduction in the water content of a given mortar mix and a considerable increase in their slump/flow. Experience from practice has shown the dosage dependent performance of superplasticizers. In the cases e.g. of inclusions of fine material rich in calcium aluminates or pozzolans rich in carbonate content (e.g. fly ash), the effectiveness of superplasticizers is reduced and their dosage should be determined by trial mixes [30].

The chemical composition and molecular architecture of the superplasticizers are extremely important, with respect to the efficiency of these admixtures. Among the different families of superplasticizers, polycarboxylated ethers (PCE) have shown the best compatibility and improvement effects in lime mortars, with those compounds with longer side chains being particularly effective [31]. The slump retention ability of lime mortars with PCE was the highest. Steric hindrance was considered to be the main action mechanism. Fernández et al. [27] reported that the addition of a PCE superplasticizer in air lime mortars containing also nanosilica, affects positively the rheological properties of the mortar, increasing the flow-ability, whilst accelerating the setting process by avoiding the formation of agglomerates from the interaction of hydrated lime particles and nanosilica through a steric hindrance mechanism [27]. Moreover, the mechanical strength was also increased by the nanosilica and superplasticizer, which modified the microstructure favourably, as proved by pore size distribution and SEM observations.

Furthermore, superplasticizers could be mixed with photocatalytic agents to reinforce their action. The agglomeration of photocatalysts in water media was avoided with the use of PCE superplasticizers. Therefore, coatings with superplasticizers showed improved photocatalytic effect of TiO_2, since the superplasticizers enhanced TiO_2 distribution and percolation in the thin coating layers. In air lime mortars, coatings with polycarboxylate-based superplasticizers improved the NO removal rates, compared to superplasticizer free coatings: an average increase of NO degradation by 15% under UV and by 76% under solar light was found [28]. Accelerated weathering of these coatings showed that the NO removal was moderately reduced, and the TiO_2 nano-particles were slightly washed out, supporting long-run activity [28].

Table 2 classifies studies from the literature for the use and purpose of plasticizers-superplasticizers in lime-based mortars considering the binder composition, the type and quantity of agent added.

2.3 Water repellents

Water repellent agents reduce the water absorption by capillarity, hence improving the durability towards weathering agents involving, in some cases, water transport (e.g., freeze–thaw, salt crystallization, wetting–drying, biological colonization), without affecting the drying kinetics. Izaguirre et al. [24] found that the anionic surfactants sodium oleate and calcium stearate can function as water repellents into lime-based mortars, similarly to their already commercialized action as water repellents for cement-based mortars. They found clear improvement in water absorption by capillarity and durability against freeze–thaw, as a result of the formation of air voids, mainly due to the air-entraining ability of those water repellents [24]. Falchi et al. [32] proposed powdered silane and calcium stearates as water repellents for pozzolana-lime mortars [32]. Both additives provided good water-repellency, even if they modified some physical properties and the hydration kinetics of the composite materials. In particular, the silane strongly enhanced the mortar resistance to salt crystallization due to the complete water-repellent effect induced. Maravelaki-Kalaitzaki et al. [33] proposed the impregnation of hydraulic-lime mortars with an oligomeric organo-siloxane; the composites exhibited improved resistance to salt-decay and mechanical stress after treatment [33].

Low water absorption through capillarity, high permeability, enhanced durability of the material

Table 2 Summary of studies on the use of plasticizers-superplasticizers in lime-based mortars

Binder composition	Additive	% wt.	References	Purpose of use
CL	Peramin SMF—Perstorp	0.05–0.15	Seabra et al. [26]	Improvement of workability Increase of mechanical strength
CL	Melflux 2651 F (BASF)	0.50–1.00	Fernandez et al. [27]	Avoidance of agglomeration of nanoparticles
CL	52IPEG 5.8	1	Perez-Nicolas et al. [28]	
CL	23APEG	1		Improvement of Stability
CL	45PC6	1		Reduction of Water retention
CL	Melcret 500F, BASF	1		Control of Rheological behavior
CL + MK	Polycarboxylate	2.0*	Arrizi and Cultrone [31]	

CL: Calcitic lime, MK: Metakaolin, % addition: refers to % wt. addition to binder, * this % wt. refers to the addition to the total mass

towards freezing–thawing cycles and delay in setting time were achieved with the addition of water repellents [24, 32, 34].

Lime-metakaolin plasters and renders were treated with zinc stearate and the effect of hydrophobization was studied by Vejmelkova et al. [35]. Increasing the content of zinc stearate in the mortar mix rapidly led to a decrease in the coefficient of water absorption, together with a reasonable decrease in both flexural and compressive strength. Furthermore, a positive effect on vapor diffusion and thermal properties through a 1% wt. of zinc stearate addition was observed.

Čechová et al. [36] and Papayianni et al. [37] studied the addition of linseed oil in lime mortars as a traditional water-repellent agent. The addition of 1% wt. linseed oil proved to increase the mechanical strength and reduce the water absorption of mortar, without inducing significant effect on the total open porosity. The resistance to salt crystallization, as well as to freeze–thaw cycles, was also improved. On the contrary, a 3% wt. addition of linseed oil resulted in a hydrophobic mortar with lower mechanical strength.

Lime-based renders with multiple additives, such as an adhesion improver (ethylene–vinyl acetate copolymer, EVA), a water-repellent agent (sodium oleate), a viscosity modifier (a starch derivative) exhibited durability towards freezing–thawing cycles and sulfate attack, rendering them as effective candidates for application in cultural heritage and contemporary buildings [29].

Silva et al. [38] compared the behavior of sodium oleate, triethoxyoctyl-silane and alkylsilicone resin in dosages up to 0.6% [38]. For the silane derivative, the authors concluded that the low solubility of this product in water explains its uneven distribution in the mortar mix and this accounts for its moderate effectiveness. In the case of silicone resin, the interaction between functional groups gives rise to insoluble polysiloxane networks and a poor distribution in the lime mortar. The silicone resin exhibited a lower reduction in water absorption but increased mechanical strength. The sodium oleate reduced water absorption, induced insignificant changes in the pore size distribution without modifying the water vapour permeability, but decreased the mechanical strength of the composite.

The action of the water repellents is based on the properties of: (a) hydrophobicity, which prevents water ingress by reducing the surface tension, and (b) water repellence, which prevents water adhesion to the surfaces. Lime mortars contain electronegative elements that can easily absorb liquid polar water; the effect of hydrophobization relies on the existence of non-polar parts in the water repellents that are able to increase the water contact angle. Those agents did not completely fill the capillary pores but are rather deposited on the pore wall allowing the water vapor diffusion [39].

Table 3 reports studies from the literature on the use and purpose of water repellents in lime-based mortars exemplifying the type of additive and dosage.

Table 3 Summary of studies on the use of water repellents in lime-based mortars

Binder composition	Additive	% wt.	References	Purpose of use
CL + MK	Cellulose derivative	2.0	Arrizi 2012	Decrease of water absorption by capillarity
CL + NP, CL + NP + WC	Linseed oil	1.0	Papayianni et al. [37]	Increase of Hydrophobicity Increase of mechanical strength
CL + MK	Zinc stearate	1.1–15.0	Vejmelkova et al. [35]	Durability improvement
CL, CL + NP	Calcium Stearate	0.5–1.5	Falchi et al. [32]	
CL, CL + NP	Silres A	0.5–1.5		
CL	Sodium oleate	0.3–2.4	Izaguirre et al. [24]	
CL	Calcium stearate	0.3–2.4		
CL	Sodium oleate	0.05–0.6	Silva et al. [38]	
CL	Triethoxyoctyl-silane	0.2–0.5		
CL	Alkylsilicone resin	0.1–0.5		

CL: Calcitic lime, MK: Metakaolin, NP: Natural pozzolan, WC: White cement, % wt. refers to the addition to the total dry mortar's weight mass

2.4 Viscosity modifiers

The viscosity modifiers are compounds that influence the fresh state properties of mortars and are intended to improve homogeneity and cohesiveness of the mixture. Therefore, the mortar workability is enhanced, along with some of the mechanical properties, especially in mixtures rich in fines [26]. The commercial and natural organic additives that have been proposed as viscosity modifiers may also function as water retainers, plasticizers and air-entraining agents.

Those additives in grouts and flowable mortars exhibited a positive effect by modifying the air content and consequently the pore size distribution, thus enhancing the durability to freezing-thawing cycles. The dosage of the additive is very critical, since it affects the water demand and, as a result, the mortar's properties [24, 40].

Seabra et al. [26] demonstrated that a hydroxypropylmethyl cellulose, (HPMC–Walocel) with water-retaining properties, a plasticizer or water-reducing agent (Peramin SMF–Perstorp) and an air-entraining agent (Silipon–Aqualon) acted in a synergistic way that resulted in a significant change of the rheological behavior of lime mortars [26]. This can be explained by the thickening effect induced with the water-retaining agent, and after some agitation time, the thinning effect that followed due to the influence of the air-entraining agent in the mortar. Conversely, the plasticizer diminished torque values, since more free water has become available in the system. Therefore, both the plasticizer and water-retaining agent reduced the water needed to achieve the desirable workability, whereas the air-entraining agents reduced the flow resistance of mortar.

Izaguirre et al. [34] tested different dosages of potato starch into air lime-based mortars in order to check its efficiency as a rheology modifier [40]. This starch polymer was found to be strongly dosage-dependent: it acted as a thickener when the incorporated dosage was up to 0.30% of lime weight; conversely, above that dosage, it behaved as a plasticizer. Zeta-potential and particle size distribution results evidenced the thickening effect induced with the polymer molecules that were adsorbed onto lime particles functioning as a flocculant. In addition, when large amounts of polymer have been used the steric hindrance and electrostatic repulsive forces lead to a dispersion mechanism which explained the plasticizing effect observed in the fresh mortar behavior [40].

Izaguirre et al. [41] studied the performance of two different commercial viscosity modifiers, namely hydroxypropyl methylcellulose and a guar gum derivative (hydroxypropylguaran) into lime-based mortars [41]. The water retention, air content and setting time, density, shrinkage, water absorption through capillarity, water vapour permeability, long-term compressive strength, pore structure and durability were tested and evaluated. In cement-based

Table 4 Summary of studies on the use of viscosity modifiers in lime-based mortars

Binder composition	Additive	% wt.	References	Purpose of use
CL	Potato starch	0.03–0.80*	Izaguirre et al. [34]	Improvement of:
CL	Hydroxypropyl methylcellulose (Walocel)	0.05, 0.10, 0.15, 0.20	Seabra et al. [26]	Workability (flowable mortars)–Cohesiveness
CL	Hydroxypropyl methylcellulose	0.29	Izaguirre et al. [41]	No
CL	Guar gum derivative	0.29		Cracking appearance
CL	Potato starch Ethylene–vinyl acetate copolymer	0.5 5–10	González-Sánchez et al. [42]	Resistance to weathering
	Sodium oleate	0.5		

CL: Calcitic lime, % addition: refers to % wt. addition to binder; *this % wt. refers to the addition to the total dry mortar's weight mass

materials hydroxypropyl methylcellulose enhanced the viscous behavior; however, in air lime mortars showed a limited viscosity modification, attributed to an adsorption mechanism of this additive on the $Ca(OH)_2$ crystals. As opposed to that, the guar gum derivative with a larger quantity of ionized groups at alkaline pH, exhibited a reduced adsorption onto slaked lime particles, giving rise to an increase in viscosity, a larger water-retention capacity, and a delay of setting time [41]. Furthermore, the guar gum derivative raised the air content and changed the pore size distribution of the hardened mortars, thus improving durability in freezing–thawing cycles.

González-Sánchez et al. [42] have reported positive effects on air lime rendering mortars using simultaneous combination of a viscosity enhancer (potato starch), an adhesion booster (ethylene–vinyl acetate copolymer, EVA), a water repellent (sodium oleate) and mineral admixtures (metakaolin or nanosilica) [42]. These additives in mortars with msetakaolin enhanced the adherence to the substrate and lowered the superficial cracking. The mixtures including nanosilica presented better durability against aggressive atmospheric conditions [42].

Table 4 summarizes several literature studies on the use of viscosity modifiers in lime-based mortars taking into account the type of binder, type and dosage of additive and purpose of use.

2.5 Carbonation accelerators

The carbonation reaction in lime mortars is a very significant process for the development of the long-term performance of these composites [43]. The carbonation accelerators affect the carbonation of lime pastes and mortars, providing shorter setting time and higher mechanical strength [44, 45]. Shrinkage could be considered as a disadvantage in some cases. Ceramic dust [45], carbonic anhydrase enzyme [46], TiO_2 nano-particles [44, 47–50], synthesized aluminosilicates [51] could be used as carbonation accelerators. The addition of nano-TiO_2 to lime mortars provided a higher concentration of CO_2 in gas form on the surface of the material. The compressive and flexural strength were significantly improved, along with the resistance to weathering parameters.

Maravelaki et al. [47] and Kapetanaki et al. [50] studied the ability of hydrated lime and metakaolin, or natural hydraulic lime mortars with nano-TiO_2, to adhere fragments of porous limestone from the Acropolis monuments. The physico-chemical and mechanical properties of the nano-titania mortars were studied and compared to the respective ones without that addition. This study highlighted that the addition of nano-TiO_2 decreases the setting time and increases the modulus of elasticity, carbonation and hydration, comparing to mortars without that addition [48].

Cizer et al. [46] studied the effect of carbonic anhydrase enzyme on the precipitation kinetics and phase transformations of calcium carbonate, and on the strength development of lime mortars [46]. The carbonic anhydrase catalyzes the reaction between carbon dioxide and aqueous lime and increases: (a) the rate of calcium carbonate crystallization, (b) the yield of the carbonation reaction and (c) the mortar strength

Table 5 Summary of studies on the use of carbonation accelerators in lime-based mortars

Binder composition	Additive	% wt. or application	References	Purpose of use
CL	Nano-TiO_2	6% CL replacement	Karatasios et al. [44]	Acceleration of setting time
CL + Mt, NHL	Nano-TiO_2	3–6%	Maravelaki et al. [47]	Accelerated carbonation rate
CL putty	Diethyl carbonate	Sprayed after 15 days	Ergenc et al. [45]	Increased depth of carbonation
CL	Synthesized aluminosilicates	5.0–10.0[b]	Loganina et al. [51]	Increase of mechanical strength
CL putty	Carbonic anhydrase enzyme	0.6 µM[a]	Cirez et al. [46]	Self-cleaning effect
CL	Nano-sized dolerite quarry waste and the olivine basalt	15% CL replacement	Kyriakou et al. [53]	
CL	Nano-sized dolerite quarry waste	5 and 15% CL replacement	Rigopoulos et al. [52]	

CL: Calcitic lime, MK: Metakaolin, NHL: Natural hydraulic lime, % addition: refers to % wt. addition to the binder, a: CA enzyme was dosed at a 0.6 µM concentration to the saturated lime solution and lime putty, b: The synthesized aluminosilicates replaced 5 and 10% of the binder

at early ages, due to the increasing rate of carbonate ions supplied to the solution by the enzyme. In addition, this enzyme favors the formation of stable calcite and significantly modifies its morphology by developing new crystal faces.

Ergenc et al. [45] investigated the effect of the diethyl carbonate on the carbonation of two types of lime mortars, one using lime putty and standard sand and the other one also containing dust and fragments of ceramic [45]. The mortar samples with the diethyl carbonate had steadier carbonation and slight changes in their microstructure.

Rigopoulos et al. [52] and Kyriakou et al. [53] used nano-sized dolerite quarry waste and olivine basalt to enhance the carbonation reaction in lime renders. The incorporation of both additions at 15% w/w resulted in mortars with denser microstructure and higher compressive strength. This was attributed to the enhanced diffusion of atmospheric CO_2 through the mortar pore structure and the improved CO_2 uptake of the nano-sized additions. Thermogravimetric (DTA/TG) and powder X-ray diffraction (XRD) analyses, as well as the phenolphthalein indicator test, confirmed that the degree of carbonation of the modified lime renders was indeed notably enhanced. This also had a positive effect on the setting and hardening time of the nano-modified end-products, which could be adopted not only for restoration purposes, but also in contemporary sustainable construction.

Table 5 summarizes the additives used to accelerate the carbonation of lime-based mortars from the literature.

2.6 Biological self-healing agents, autogenous self-healing stimulators

Self-healing materials are able, once damage occurs, to repair themselves to restore their original properties or limit further deterioration, without human intervention. Such a function is a vital development in reducing the significant maintenance costs in building structures and the insidious deterioration of valuable heritage structures.

The self-healing agents are proprietary chemical compounds, generally known under the category of “crystalline admixtures” [54], added to the cement/lime mixtures; they are highly hydrophilic and can react with calcium hydroxide to promote the carbonation and formation of non-soluble compounds, which deposit into cracks and seal them, also contributing to recover the pristine level of performance (e.g., compressive strength) of the pre-damaged mortar [55, 56]. To the same purpose, the addition into the mortar of hydraulic lime encapsulated into either organic or non-organic shells has also been attempted [55]. As for the encapsulated lime, the cracks break the capsules by intercepting them and the fresh hydraulic lime mortar cargo becomes available to react with the calcium hydroxide and outdoor available agents (water, moisture, air), thus promoting delayed hydration and carbonation reactions, whose products contribute to the sealing of the cracks.

Biological self-healing incorporates naturally occurring microorganisms, such as bacteria, along with chemical precursors which can lead to biomineralization, e.g., of calcite. Although, the alkaline environment in lime mortars is considered harsh for microbial organisms, there are considerable opportunities for successful application on geological materials, due to their bioreceptivity and suitability for biomineralization. Different ways in which the bacteria, along with the precursor chemicals, can be introduced into porous geological construction materials are currently under study, aiming at defining a protocol for the application and evaluation of the results [57]. In biomineralization, spores trapped within a mineral are exposed by damage and germinate into cells, which heal the damage, re-encapsulating themselves and resetting the cycle. Calcite biomineralization via urea hydrolysis can be used as the basic mechanism for assessing self-healing in lime mortars, due to its common mineralogy, porosity and moisture content. The hydrolysis of urea produces ammonia and carbonate; ammonia release acts to raise the pH of the medium which is a favorable condition for the precipitation of calcium carbonate. Carbonate binds calcium ions, which are present in the medium, resulting in the formation of calcium carbonate crystals. The potential of the mechanism to function and modify the microstructure of the materials under study could be detected through simple water absorption tests and the application of other analytical methods and microscopic techniques [57]. Alternatively to the use of bacteria that produce calcite biomineralization, other bioproducts have been studied with the same aim of reducing water absorption of air lime mortars, such as iron supplemented Escherichia coli cultures and microbial mixed cultures grown with crude glycerol [58]. The use of these bioproducts decreased the mortar compressive strength but improved its water-mediated weathering behaviour.

Table 6 lists several studies from literature on the effects of self-healing agents on lime-based mortars.

2.7 Crystallization modifiers

The addition of crystallization modifiers to lime mortars is a novel approach aimed at eliminating damage induced by salt crystallization in historic masonry. Crystallization modifiers are ions or molecules which exhibit a multi-functional action in the salt crystallization process: they can inhibit nucleation, promote efflorescence instead of damaging crypto-efflorescence, and/or modify the shape of the crystals [59]. Lubelli et al. [59] studied whether the incorporation of sodium ferrocyanide in different percentages in cement-lime mortars influence the durability of these mortars to salt crystallization [59]. It was proved that the addition of sodium ferrocyanide in the fresh mortar contributed to the reduction of damage induced by sodium

Table 6 Summary of studies on self-healing lime-based mortars

Binder composition	Additive	% wt. or application	References	Purpose of use
CL + NHL5 + Cem	Coated granules with sodium fluoride	3.0–6.0	De Nardi et al. [56]	Healing capacity
CL + NHL5 + Cem	Coated granules with maleic anhydride	3.0–6.0		Healing capacity and recovery of compressive strength
CL + NHL5 + Cem	Coated granules with phthalic anhydride	3.0–6.0		Reduce water absorption
CL + NHL5	Crystalline admixture	3.0	De Nardi et al. [55]	
CL	E.coli + Fe	Lyophilized, suspended in water and used as mixing liquid	Oliveira et al. [58]	
CL	MMC			

CL: Calcitic lime, NHL:5 Natural hydraulic lime5, Cem: Cement, E. coli + Fe: *Escherichia coli*-based bioproduct, MMC: Crude-glycerol-based microbial mixed cultures bioproduct, % addition by weight of mortar

Table 7 Summary of studies on the use of crystallization modifiers in lime-based mortars

Binder composition	Additive	% wt	References	Purpose of use
CL	Sodium ferrocyanide	0.94	Lubelli et al. [59]	Resistance to salt crystallization
CL	Borax	1.6–3.2		
Cem + CL	Sodium ferrocyanide	0.001–0.02%*	Granneman et al. [60]	

CL: Calcitic lime, Cem: Cement, % addition refers to % wt. addition to binder

*This addition refers to % wt. of the dry weight of the mortar

chloridecrystallization, since a change in the crystal morphology of sodium chloride has occurred. Granneman et al. [60] reported sodium ferrocyanide and borax, as modifiers for sodium chloride and sodium sulfate crystallization, respectively [60]. The results showed that sodium ferrocyanide changed the crystal morphology yielding elongated crystals that can less adhere to the pore walls; borax, on the other hand, modified the status of anhydrous/hydrated state of sodium sulfate from prisms to elongated needles, thus favoring the formation of efflorescence rather than the damaging crypto-efflorescence. Therefore, the selected modifiers may mitigate salt crystallization even after going through the carbonation process of the mortar. Besides, no major effects of the modifiers on the fresh and hardened mortar properties were observed, rendering them as potential crystallization modifiers in restoration mortars. The addition of these agents that can reduce crystallization damage of lime mortars needs more studies to assess their behavior in the presence of other salts in the mix.

Table 7 summarizes the above-mentioned studies on the use of crystallization modifiers in lime-based mortars.

2.8 Inorganic nano-particles

Inorganic or mineral particles of 10^{-9} m (nano-dimensions) are mixed with lime binders, providing mortars with self-cleaning properties, as well as improved mechanical properties. Nano-TiO_2 and SiO_2 are common nanoparticles used as additives in lime mortars [27, 48, 49, 61–65].

Nanosilica is one of the most studied types of nanoparticles used as an additive in cementitious composites to improve their characteristics in the plastic and hardened state [66–68]. Investigating the effect of nanosilica on the properties of lime-based systems, the main drawback was the water/binder ratio increase with increasing amount of nanosilica, whereas the workability and setting time decreased. Superplasticizers have, therefore, been added to tackle this problem [27, 68]. Researchers also reported that the mechanical strength increases with increasing amount of nanosilica, whereas the capillary porosity decreases, hence improving the durability of lime mortars [60, 62, 63, 69]. Graphene oxide (GO) was also proposed as additives into natural hydraulic lime mortars in different concentrations. The influence of GO percentage and the type of mixing was investigated in terms of changes induced in the composite microstructure, mechanical and physical properties [65]. The best results were obtained with dispersed GO at concentrations of 0.05% and 0.1% wt/wt of binder, slightly improving the mechanical and physical characteristics and enabling those mortars to be used for building rehabilitation.

The development of photocatalysts as self-cleaning coatings is an innovative field to mitigate the decay of surfaces induced by atmospheric pollution. TiO_2 nanocomposites have been investigated for their photocatalytic properties. These nanocomposites produce photoactive coatings when applied, without compromising the mortar hardened state properties [44, 48, 49, 61, 67]. Some efforts have been made to enhance the sensitivity of the photocatalysts towards the visible light spectrum. Doped-TiO_2 (Fe–TiO_2 and V–TiO_2) nanoparticles have been tested as effective agents to be applied as active coatings [28, 70]. The photocatalytic assessment of the Fe-doped TiO_2 nanoparticles indicated that doping titania with a low iron content (0.05 and 0.10 w/w % to binder) makes it possible to degrade organic pollutants under visible radiation [70]. In this study, another interesting finding was the enhancement of carbonation in lime mixtures at early stages.

Table 8 Summary of studies on the use of nanoparticles in lime-based mortars

Binder composition	Additive	% wt.	References	Purpose of use
CL	SiO_2	0.5–6.0	Fernandez et al. [27]	Increase of mechanical strength Photocatalytic activity Self-cleaning effect Acceleration of setting time Slight increase of mechanical strength Decrease of water absorption
CL + CB		3	Theodoridou et al. [49]	
CL + P		1.5	Nunes et al. [76]	
CL		6–20	Duran et al. [64]	
CL, CL + Cem, CL + Gyp, CL + MK, NHL	TiO_2	0.5–6.0 6	Lucas et al. [61], Maravelaki-Kalaitzaki et al. [47], Kapetanaki et al. [50]	
CL + CB		3	Theodoridou et al. [49],	
CL		3–11	Karatasios et al. [44]	
CL	Fe-TiO_2	0.05–1	Kapridaki et al. [70]	
NHL	GO	0.05–1.0	Faria et al. [65]	

CL: Calcitic lime, P: Natural pozzolan, MK: Metakaolin, NHL: Natural hydraulic lime, Cem: Cement, Gyp: Gypsum, % addition: refers to % wt. addition to binder

Table 8 reports several studies from the literature on the addition of nano-particles in lime-based mortars and the effects induced on the mortars' performance.

2.9 Natural and polymeric compounds

Polymers consist of large molecules or macromolecules and are composed of many repeated subunits. Depending on their composition, polymers are used in lime mortars to provide mixtures with improved properties, such as adhesion, resistance to moisture and weathering. In past centuries, people used to add different organic components in building materials, such as plants´ mucilages, animal fats, etc. A vernacular example of the use of natural polymers is the extinction of calcium oxide together with animal fat with abundant water, resulting in a water-repellent lime putty. Nowadays, natural organic compounds are still used in India with lime-based mortars [71]. Recently, the influence of natural additions on the physical and mechanical properties of lime mortar was investigated in order to better understand the mechanism of ancient mortars [11, 16, 72–75].

Various organic additives have been used as additions, according to their properties and historical use: polysaccharides (e.g. opuntia used either as a powder or as mucilage), proteins (e.g. animal glue and casein) and fatty acids (e.g. olive oil, linseed oil). They were all considered compatible with traditional building materials [11]. The presence of proteins and carbohydrates influences the carbonation and hydraulic reaction in lime mortars and can enhance mechanical properties. Some types of organic additives can also reduce the porosity and the water affinity of the mortar [72]. Fatty acids are mainly used to impart water repellent properties to mortars and improve their durability in severe weathering conditions involving water transport [76]. The mucilaginous juice extracted from nopal cladodes constitutes a natural organic additive that has been incorporated in the slaking lime water for the formulation of lime mortars and plasters [77]. This study evidenced that the adsorption of polysaccharides on $Ca(OH)_2$ crystals prevents the development of large particles, resulting in a very reactive, nanosized portlandite slurry. It also promotes steric stabilization, which favors the colloidal nature of the lime putty and limits aggregation. Overall, these effects enhance the plasticity and related properties of lime mortars. It should be highlighted that these potential effects occurred whether the nopal juice was added to a slaking lime, and not in an already slaked one as it is the case of commercial dry hydrated lime [77].

Kang et al. [78] studied the effect of addition of fully refined sugar (sucrose 99.9%) to the hydration of quicklime and the development of carbonation process and compressive strength of derived lime-based mortars [78]. Water solution containing 3.5 and 7% of sugar were tested and the observed are related to a delayed quicklime hydration yielding significantly

Table 9 Summarizing studies for natural compounds and polymers in lime-based mortars

Binder composition	Additive	% wt.	References	Purpose of use
NHL	Cissus Glauca Roxb (CGR)	15–20^{a}	Ravi et al. [72]	Increase of mechanical strength Water repellency Resistance to salt crystallization Resistance to frost Enhancement of carbonation rate
CL	Animal glue	5.0	Ventola et al. [11]	
CL	Casein	5.0		
CL	Nopal as powder	5.0		
CL	Nopal as mucilage	5.0		
CL	Olive oil	5.0		
CL	Sugar (sucrose)	3.5 and 7	Kang et al. [78]	
CL	Areca nut extract	3–5%	Gour et al. [73]	
CL	Fermented organic extracts (aloe vera, cactus, kadukkai, hibiscus, jaggery, and neelamari)	Up to 5%	Shivakumar et al. [74]	
CL	Ficus Carica (Moraceae)	2, 4 and 6% in volume	Jayasingh et al. [75]	
SL	Nopal juice	The quicklime slaked in aqueous nopal juice extract;	Rodrigues-Navarro et al. [77]	
CL, CL + MK	Linseed oil	1.5	Nunes et al. [76]	

NHL: Natural hydraulic lime, CL: Calcitic lime, SL: Slaked lime putty, MK: Metakaolin, % addition: refers to % wt. addition to binder, a: Content of CGR % by wt. of water

smaller hydrated lime particles. This is attributed to the molecular structure of sucrose that effectively retained absorbed water for a long time, alleviating dry shrinkage and the resulting cracks in lime–based materials. The beneficial effect is also reflected to the higher compressive strength developed at 28 and 56 days in the mortars prepared with the sugar additive. Moreover, it seems that the presence of sugar in the hydrated lime assists the CO_2 diffusion inside the mortar because of the slower carbonation that considerably delays the clogging of pores around the surface of mortar [78].

Table 9 summarizes the studies from the literature on the use and effects of natural compounds and polymers in the lime-based mortars.

3 Classification of admixtures according to their action

In the following sections, the most commonly used admixtures, which represent substances added in the mortar mixture, are classified into categories according to their multi-function characteristics that influence various mortar properties. More specifically, phase change materials (PCMs), natural and synthetic fibers, pozzolans and white cement are hereby presented and discussed.

3.1 Phase change materials (PCMs)

PCMs can function as latent heat storage systems, absorbing or releasing heat upon changing their phase from solid to liquid or vice-versa [12]. This ability of PCMs makes them suitable to be used as admixtures in the production of thermally efficient composite building materials and products, such as lime-based plasters [79]. To avoid unintended movements in the binding matrix during the liquid phase of PCMs, and also to prevent microstructural changes that could compromise the performance of the mortars, the use of microencapsulated materials has been developed [80].

The fresh state characterization of lime mortars with PCM additions was studied by Lucas et al. [81], who found that there was an improvement in workability and mechanical strength when PCM capsules

Table 10 Summary of studies on the use of PMC in lime-based mortars

Binder composition	Admixture	% wt.	References	Purpose of use
CL	Micronal DS 5008, (BASF)	5, 10, 15, 20*	Lucas et al. [81]	
CL + Cem				
CL + Gyp				
CL	PCM DS 5001 Micronal® (BASF) (wax with a specific alkane chain)	5, 10, 15*	Ventola et al. [12]	Improvement of
CL	PCM DS 5001 Micronal (BASF)	9.1*	Pavlik et al. [83]	Thermal properties
CL	Paraffin core with wall in melamine–formaldehyde	21.26	Cunha et al. [82]	
NHL5		45.38		Thermal insolation
CL	Micronal DS 5038 X (BASF)	5*	Theodoridou et al. [79]	
NHL				
CL	Poly(ethylene glycol)	6.375	Frigione et al. [84]	
		69		
NHL	Poly(ethylene glycol)	23.02	Sarcinella et al. [85]	

CL: Calcitic lime, NHL: Natural hydraulic lime, Cem: Cement, Gyp: Gypsum, % addition: refers to % wt. addition to binder, *this addition: refers to % wt addition to binder and aggregates

were incorporated in air lime mortars [12, 81]. PCMs addition results in a better compaction of mixtures, as the friction between particles reduced, and consequently the flow resistance. Ventola et al. [12] further investigated the use of PCMs as an admixture in traditional lime mortar [12]. This led to improved thermal insulation properties, compressive strength and carbonation rate. According to Cunha et al. [82], the use of microencapsulated PCMs in lime-based mortars leads to an increase in water demand during the mixing processes; this is attributed to the fineness of the PCM microcapsules [82]. Furthermore, PCMs induce an increase in the microporosity, as well as a decrease in the flexural and compressive strengths of the hardened end-product. Theodoridou et al. [79] also studied the effect of microencapsulated PCM addition on the performance of traditional lime-based plasters [79]. Their results suggest that the use of PCMs enhances the thermal properties of air and hydraulic lime composites, but reduces their compressive and flexural strength. In addition to the most commonly tested paraffin waxes (such as the commercial Micronal of BASF [83]), Frigione et al. [84] and Sarcinella et al. [85] tested in air lime and hydraulic lime mortars PCMs based on poly-ethylene glycol supported onto fragments of a calcareous stone. According to these studies the proposed PCMs leave unchanged the thermal conductivity when added to aerial lime mortars, whilst reducing the heating and cooling needs of the hydraulic lime mortars.

Table 10 reports several studies from the literature on the type of PCM admixture used and the effects in lime-based mortars.

3.2 Fibers: natural/synthetic

Fibers are mostly used in mortars in order to improve their engineering properties, such as fracture toughness, flexural strength, control of plastic shrinkage cracking, and, in general, cracking [86]. The use of natural fibers (straw, wooden fibers, animal hair) has been widespread in the past, as it has been found by the analysis of old mortars, in particular when clay was used as the binder, in the case of lime plasters serving as substrate of frescos or other decoration to mitigate cracking [71] or in the case of gypsum plaster substrates.

Fiber-reinforced hydraulic lime mortars can be used for repairing historic structures. The use of glass, basalt and even polymer fibers from recycled bottles has been reported, with known effects on improvement of the mortar mechanical behavior (flexural strength, toughness) in the post-cracking condition [86, 87].

Table 11 Summary of studies on the use of fibers in lime-based mortars

Binder composition	Admixture	% wt.	References	Purpose of use
CL	Glass fibre	1.0–2.0	Iucolano et al. [86]	Increase of flexural strength
CL	Basalt fibre	1.0–2.0		
CL + Cem	PET fibre	0.5–1.5^{a}	Pereira de Oliveira et al. [87]	Improvement of toughness
			Izaguirre et al. [88]	Effect on the consistency
NHL	Nano-fibrils	0.17–0. 80	Rosato [91]	Increase of fracture energy
CL + MK	Palygorskite	4.16–5.26	Andrejkovičová et al. [90]	
CL	End-of-Life tires	0.25–1.0	Gil et al. [94]	Reduction of cracks
CL + Cem	Natural fibers of jute, coconut and kelp	1.5^{a}	Kesikidou and Stefanidou [92]	
NHL	Nano-structured cellulose fibers	0.17, 0.4, 0.8	Rosato et al. [91]	
CL	Electrospun CA fibers	0.59 and 1	Chousidis et al. [89]	

CL: Calcitic lime, NHL: Natural hydraulic lime, Cem: Cement, MK: Metakaolin, % addition: refers to % wt. addition to binder, ^{a} % addition: per volume in dry mortar

The effect of polypropylene fibers on the behavior of air lime-based mortars was studied by Izaguirre et al. [88]. It was found that, when the fibers were added in low dosage in air lime-based mortars, there was an improvement in various properties, such as strength, permeability, macroscopic cracks reduction or freeze-thawing durability. However, the aforementioned authors also noticed that the addition of fibers in lime-based mortars hindered workability, which required a larger amount of water that gave rise to the formation of larger pores. It was, thus, concluded that only crack reduction and material durability were improved with the addition of fibres in larger amounts.

Electrospun cellulose acetate (CA) polymer fibers have been used by Chousidis et al. [89] to reinforce lime pastes [89]. Their addition resulted in a tremendous improvement of the mechanical properties of the latter, thus pointing towards the use of electrospun fibrous additives in the development of advanced composite lime-based building materials for traditional and contemporary structures.

Another type of fibrous material, such as the particles of needle-like palygorskite, has been incorporated in air lime binder with metakaolin [90]. It was observed that needle-like palygorskite particles are functional as reinforcing fibers. Another study investigated the influence of nano-structured cellulose fibers (bio-fibrils) on the properties of natural hydraulic lime pastes [91]. A slight decrease of flexural and compressive strength was registered as the percentage of bio-fibrils increased. The only beneficial effect was the decrease in the total capillary absorbed water, as a function of the admixture increase.

Finally, reduced workability and inhomogeneity have been mentioned by authors as a drawback of fiber addition in lime and cement-based mortars [92–94]. In addition, research has shown that in lime mortars hemp fibers could achieve the same strength in compression and flexure with polypropylene fiber reinforced mortars [95]. Gil et al. (2016) investigated the addition of fibers from End-of-Life tires. It was found that 1% addition of fibers in lime mortars slightly decreased the consistency, without changing the mechanical properties [88].

Table 11 reports several studies from the literature on the type of fibers used and the effects in lime-based mortars.

3.3 Pozzolans: natural/artificial

Even from the Late Bronze age period a common practice was to add natural or artificial pozzolans to traditional lime mortars to improve their mechanical performance and durability, especially the ability to harden in the presence of water. Their effectiveness and mortar performance are well-documented and proven by several applications [37, 96–102]. Artificial and natural pozzolanic materials added to lime mortars yielded calcium aluminosilicate composites featuring hydraulic activity, and a microstructure with reducing pore radii and increasing apparent density. $Ca(OH)_2$ was consumed by the pozzolanic reaction enabling a decreased hardening time of the new materialsand an increased mechanical strength.

The effect of artificial pozzolanic admixtures on the mechanical, thermal and hygric properties of lime mortars were investigated by Papayianni [103], and Cerny et al. [104]. In both studies, significant mechanical and physico-chemical properties of lime mortars, such as compression and flexural strength, heat conductivity capacity, diffusivity of moisture, water sorptivity, co-efficient of water vapor diffusion, sorption isotherms, and coefficients of heat and hygric linear expansion were determined. They concluded that lime-pozzolan mortars exhibited significantly improved mechanical, thermal and hygric properties. Therefore, they suggested the use of lime pozzolan mortars as plasters and renders, instead of the traditionally used pure lime mortars.

The specific surface area of the pozzolan was demonstrated to be the parameter mostly affecting the water demand of the paste, amorphousness being the most relevant property that determines the pozzolan reactivity to a much greater extent than any other pozzolan property [105]. Conversely, the chemical composition of the pozzolan seems to influence either its reactivity or the strength of the paste insignificantly. However, according to Mehta [108], the mineralogical composition plays a significant role on the reactivity of the pozzolan [106]. Therefore, more than the silica and alumina content, the amorphous fractions are important when comparing pozzolans with similar high surface area.

Vejmelkova et al. [107] studied the mechanical, fracture-mechanical, hygric, heat and durability properties of lime-metakaolin plasters for renovating historical buildings [107]. They concluded that, when compared with the reference lime plaster, there was an improvement in the mechanical parameters, along with a decrease of 25% in the diffusion coefficient of water vapor. The reduced liquid water transport was attributed to the presence of metakaolin. They suggested this formulation as adequate for renovating a wide range of historical buildings. Xu et al. [110] studied natural hydraulic lime-based mortars using diatomite/fly ash as mineral admixtures, with aggregates of masonry waste [108]. The properties of the designed mortars were improved because of the pozzolanic reaction between diatomite, fly ash and $Ca(OH)_2$. Diatomite was found to have a stronger and better pozzolanic effect; it also improved the compressive strength of mortar, compared to fly ash.

While several studies on air lime natural pozzolan, fly ash, metakaolin and fired red clay investigated the mechanical properties of pozzolanic mortars, they have not included triaxial testing results [107, 109–111]. The determination of the triaxial behavior of hydraulic mortars is important when considering that they are brittle materials with an elastic behavior, in contrast to lime mortars which exhibit a plastic behavior [112]. The evaluation of hydraulic mortars under triaxial conditions becomes more important when considering their application with respect to infill joint fragments of historic structures, where shear stresses develop along the mortar-stone interface, which may result into lateral confinement of the mortar layers. Kaklis et al. [113] investigated the triaxial behavior of a pozzolanic mortar consisting of hydrated lime and metakaolin, which was used as a filler between metallic connectors and marble blocks during restoration activities of ancient monuments in Greece [113]. They highlighted the plastic behavior of the mortars under study, enabling them to perform well in the intended conditions.

3.4 Cement: white, Portland

Mosquera et al. [114] studied the effect of cement addition to hydraulic lime mortars, in partial replacement to the lime binder [114]. Mercury Intrusion porosimetry results in the aforementioned study showed that the diffusivity values were comparatively higher for the lime/cement mixes, compared to the mixes which contained only hydraulic lime as binder.

The research of Arandigoyen et al. [115] showed that 0–40% addition of cement in lime mortars resulted in a slight increase of mechanical strength, whereas the addition of a small amount of lime in cement mortars led to a high decrease of the mechanical strength [115]. Papayianni [116] presented that the addition of cement influences essentially the early strength and porosity of the end-products [116].

However, lime-rich mortars exhibit a more plastic behavior, being able to undergo higher deformation. Similar results were also obtained by Cizer et al. [117], who studied mortars composed of 30%, 50% and 70% hydrated lime and lime putty as cement replacement [117]. As the percentage of lime increases, the total porosity also does so, whereas the compressive strength decreases. However, the addition of lime resulted in more plastic behavior.

Silva et al. [118] investigated the addition of cement in lime mortars used in restoration projects, as substitution to lime-natural hydraulic lime binder [118]. The water transport properties, porosity and mechanical strength of the mortars were studied as these properties influence compatibility. Cement-lime mortars proved to be less porous and therefore less permeable, compared to lime-hydraulic lime mortars. Additionally, it was also mentioned that the presence of soluble salts in cement should be considered as a cause that initiates chain reactions yielding efflorescence and sub-efflorescence in lime mortars.

4 Evaluation tests/analyses

The most important properties and the analysis that should be performed to study the effect of additives and admixtures in the fresh and hardened state of mortars are reported in Tables 11, 12, 13, 14 and 15. The evaluation of adds/adms in lime mortars requires the use of specific tests and techniques that are differentiated concerning the fresh/hardened state of mortars. The tests and the relevant standards that are proposed for the majority of the properties evidence the need for further standardization that could elucidate both the optimum amount of additive/addition and the method of assessment. This review has as objective to summarize the state of the art and portray future developments in the use and assessment of adds/adms in lime mortars.

The influence of adds/adms on the basic properties of fresh and hardened state mortars is illustrated in Fig. 1. Apart from the individual contribution of each agent to the improvement of each property, it becomes evident that combining certain additives and/or admixtures, the beneficial effect is enhanced in both the fresh and hardened state. Supporting evidence is the combined effect of plasticizers, nano-particles, polymers and carbonation accelerators in the increase of compressive strength and decrease of porosity, as illustrated in Fig. 1. Furthermore, all the properties shown in Fig. 1 exemplify the influence of additives/admixture in diverse gradients to the fresh and hardened state properties of mortars. Several inorganic nanoparticles, carbonation accelerators and cement, for example, remarkably reduce the setting time.

5 Conclusions

The preservation and restoration of historical structures are mostly based on the use of lime-based mortars. Additives and admixtures have been found in lime mortars of different historical periods, originating mostly from locally available materials. They occur in all types of lime-based mortars, while their proportions by weight and volume are differentiated according to the type of mortar and the building technology of each historical period. They were deliberately added in lime mortars in order to improve either the fresh or the hardened state properties of the mixtures. This knowledge was considered as the “secret” of ancient builders, which allowed them to construct different types of structures and buildings, using water-proofed mortars for aqueducts and baths, and occasionally lime-based “concrete”. During discussions for this paper, a topic has arisen concerning the terminology used in the literature and the definitions of EN 16572. The terms additives and admixtures refer to the additions to the lime and in the lime mortar mixtures, respectively, with the aim of improving workability, physico-mechanical properties and durability. According to definitions given in EN 16572, additive (EN) or addition (ASTM) is a constituent usually added in small quantity to the binder to modify its manufacture or properties (for example accelerators, plasticizers and air-entraining agents), whereas admixture is a substance other than the binder, aggregate or water, added in quantities of at least 1% wt/wt to the mix to alter its properties.

Nowadays, additives and admixtures are available in a great variety, due to the evolvement of the chemical industry, and can thus be used very effectively in contemporary lime mortars. They are being developed into much more user-friendly forms, based on modern technology, namely waste recycling, and are no longer making use of products that can be exploited for feeding humans and animals. The

Table 12 Fresh state properties and relative analysis of lime mortars with additives

Property studied		Workability	Rheological behavior	Water retention capacity	Air content of fresh mixture	Reactivity	Saturation moisture content	Setting time	Carbonation-Hydration reaction
Analysis/ test		Flow table test	Rheology and Viscosity	Water retention measurement	Pressure method	-pH metering - Conductivity	Gravimetric method	Vicat test	-DTA/DSC - FTIR - XRD - BET - Particle size distribution - Zeta Potential Analyzer
Standard		EN 1015-3 (1999)		UNE 83-816-93 (1993)		RILEM CPC-18 (1980)		EN 196-3 (2016)	
Additives	Air entraining	☑	☑		☑				☑
	Plasticizers-Superplasticizers	☑	☑		☑			☑	☑
	Water-repellents	☑	☑	☑	☑		☑	☑	☑
	Viscosity modifiers	☑	☑	☑				☑	☑
	Carbonation accelerators					☑			☑
	*Self-healing								☑
	Crystallization modifiers	☑							
	Inorganic nanoparticles	☑	☑						☑
	Natural/polymeric compounds	☑				☑			☑

addition of additives and admixtures in lime mortars is essential to gain improved properties, as well durability, compared to pure lime mortars These materials can promote freeze–thaw resistance, repel water absorption, and allow mortars to be cured more efficiently, making mortars much more plastic and workable, with increased mechanical properties. They can also reduce the overall water content in a mortar, thus significantly reducing problems of drying shrinkage. Therefore, additives and admixtures can be important for both the application (fresh state properties) and for the long-term performance of mortars. Based on their action mechanism, they offer significant benefits to lime-based mortars, giving technicians the ability to apply them in broader application fields.

To examine the efficiency of additives/admixtures in a lime mixture, a protocol should be established containing tests of the fresh and harden state properties, as well as tests related to durability. The tests are necessary in order to decide upon the correct dosages that should be used or even to select in the most efficient way between different products. This important gap that is referred to the lack of standardized methodology for assessing some important properties that depend on the additive/admixture dosage and efficiency opens new research possibilities in this field.

Therefore, in this review paper, various agents, classified according to their action or shape or nature, were presented along with the most important properties that were analyzed. Some additives, such as superplasticizers, air-entraining agents, have been used successfully in repair mortars for more than 20 years. Most of the additives presented in this work have been under investigation mainly in the last 20 years, whereas other additives such as vernacular natural polymers, and admixtures such as fibers, pozzolans and cement, are well established in lime-mortar technology. Regarding the assessment of air-entraining agents, plasticizers, superplasticizers, water repellents, viscosity modifiers, crystallization modifiers, nanoparticles of SiO_2 and TiO_2, research

Table 13 Hardened state properties and relative analysis of lime mortars with additives

Property studied	**Morphology-structure**				**Hygric-physical properties**					**Mechanical properties**	
	Morphology Homogeneity Microstructure	Shrinkage	Density	Porosity	Water absorption	Hydrophobicity	Water vapour permeability	Colorimetric measurements	Thermal properties	Mechanical strength	Pencil hardness
Analysis/ test	OM, SEM, BET	Shrinkage test	Density measurement	MIP RILEM CPC 11.3	Capillarity test	Contact angle measurement	Water vapour permeability test	UV-VIS, chromatometer	-Thermal conductivity measurement, -Specific heat capacity measurement	-Compression test - Flexural test	Hardness measurement
Standard		ASTM C596 (2001)	EN 1015-10 (1999)	EN 1936 (2006), EN 623-2 (1993)	EN 1015-18 (2002), EN 15801 (2010)	EN 15802 (2010)	DIN 52615 1987, EN 1015-19 (1998)			EN 12390-3 (2019), EN 1015-11 (2019)	ASTM D 3363 (2020)
Air entraining	☑			☑	☑		☑			☑	
Plasticizers-Superplasticizers	☑	☑		☑	☑		☑			☑	
Water-repellents		☑		☑	☑		☑	☑	☑	☑	☑
Viscosity modifiers	☑									☑	
Carbonation accelerators	☑			☑	☑					☑	☑
*Self-healing	☑									☑	
Crystallization modifiers	☑		☑	☑	☑					☑	
Inorganic nanoparticles	☑	☑		☑		☑		☑		☑	
Natural/Polymeric compounds	☑				☑			☑		☑	

Table 14 Fresh state properties and relative analysis of lime mortars with admixtures

Property studied		**Workability**	**Rheological behavior**	**Water retention capacity**	**Saturation moisture content**	**Setting time**	**Carbonation-Hydration reaction**
Analysis/ test		Flow table test	Rheology and Viscosity	Water retention measurements	Gravimetric method	Vicat test	DTA/DSC, FTIR, XRD, BET, Particle size distribution, Zeta Potential Analyzer
Standard		EN 1015-3 (1999)		UNE 83-816-93 (1993)		EN 196-3 (2016)	
Admixtures	PCM	☑	☑				
	Fibers	☑					
	Pozzolans	☑		☑	☑	☑	☑
	Cement	☑					

Table 15 Hardened state properties and relative analysis of lime mortars with admixtures

Property studied		**Morphology-structure**			**Hygric-physical properties**				**Mechanical properties**	
		Morphology Homogeneity Micro-structure	Density	Porosity	Water absorption	Water vapour permeability	Thermal conductivity	Specific heat capacity	Mechanical strength	Pencil hardness
Analysis/ test		OM SEM BET	Density measurement	MIP RILEM CPC 11.3	Capillarity test	Water vapour permeability test	Thermal conductivity measurement	Specific heat capacity measurement	Compression test Flexural test	Hardness measurement
Standard				EN 1936 (2006), EN 623-2 (1993)	EN 1015-18 (2002), EN 15801 (2010)	DIN 52615 (1987), EN 1015-19 (1998)			EN 12390-3 (2019), EN 1015-11 (2019)	ASTM D 3363 2020
Admixtures	PCM	☑							☑	
	Fibers	☑	☑	☑	☑				☑	☑
	Pozzolans	☑		☑	☑	☑	☑	☑	☑	
	Cement	☑	☑	☑		☑			☑	☑

advances in the literature, based on evaluation criteria and durability studies, suggest their use in lime mortars, as they enhance the physico-mechanical properties of the end-product. Besides, more innovative admixtures, such as PCMs, along with self-sensing and self-healing stimulators, need to be further investigated regarding certain aspects of their performance evaluation and durability.

This review opens new thoughts and insights concerning further elaboration in the CEN adopted terminology regarding additives/admixtures. Even though the definitions widely used in the cement industry consider the air-entraining agents, the superplasticizers and the viscosity modifiers as admixtures, nevertheless, the EN standardization introduced the term additives for those agents. Therefore, the adoption of this term is imperative in the cultural heritage field. Given that lime-based mortars can include hydraulic mortars, namely air lime-pozzolan mortars, when limes with hydraulic properties are used and when cement is added to lime mortars, a similar terminology to the cement/concrete mortars would better indicate the improvement achieved after the use of additive/admixture and allow comparisons between different types of mortars.

Funding Open access funding provided by HEAL-Link Greece.

Declarations

Conflict of interest The authors declare no competing interest.

References

1. EN 16572 (2015) Conservation of cultural heritage–Glossary of technical terms concerning mortars for masonry, renders and plasters used in cultural heritage. European Committee for Standardization (CEN)
2. Ramachandran VS (1995) Concrete admixtures handbook. Properties, science and technology. 2nd Edition, Noyes Publications
3. ACI 116R-90 Cement and concrete terminology, American Concrete Institute
4. BS EN 934–2:2009+A1:2012, Admixtures for concrete, mortar and grout part 2: concrete admixtures–Definitions, requirements, conformity, marking and labelling, European Committee for Standardization (CEN).
5. Sickels LB (1981) Organics vs. synthetics: their use as additives in mortars, Symposium on mortars, cements and grouts used in the conservation of historic buildings. 3-6.11.1981 Rome, p 25
6. Neuburger A (1930) The technical arts and sciences of the ancients. Nature 126:949–950
7. Sayre V (1976) Deterioration and restoration of plaster, concrete and mortar. In Preservation and conservation: principles and practices. Proceedings of the North American international regional conference. Williamsburg, Virginia, 10–16 September 1972 pp. 191–201.
8. Bankart G (1908) The art of the plasterer. Routledge, New York
9. Stefanidou M, Papayianni I, Pachta V (2012) Evolution of Inclusions in historic mortars. Archaeometry 54(4):737–751
10. Papayianni I (1997) Effectiveness of superplasticizers and air-entraining agents on lime-based repair mortars. In: VM Malhotra (ed) Proceedings of 5th CANMET/ACI International Conference on Superplasticizers and other Chemical Admixtures in Concrete, pp 235
11. Ventolá L, Vendrell M, Giraldez P, Merino L (2011) Traditional organic additives improve lime mortars: New old materials for restoration and building natural stone fabrics. Constr Build Mater 25:3313–3318
12. Ventolà L, Vendrell M, Giraldez P (2013) Newly-designed traditional lime mortar with a phase change material as an additive. Constr Build Mater 47:1210–1216
13. EN 998–1 (2010) Specification for mortar for masonry-part 1: rendering and plastering mortar
14. EN 998–2 (2016) Specification for mortar for masonry-part 2: masonry mortar
15. Veiga R (2017) Air lime mortars: What else do we need to know to apply them in conservation and rehabilitation interventions? A review. Construct Build Mater 157:132–140. https://doi.org/10.1016/j.conbuildmat.2017.09.080
16. Ramamurthi DS, Sophia M (2016) A review on modified lime based mortars–An alternative to cement mortar. IJIRST Int J Innov Res Sci Technol 2(12):350–356
17. Van Balen K, Papayianni I, Van Hees R, Binda L, Waldum A (2005) Introduction to requirements for an functions and properties of repair mortars. Mater Struct 28(282):781–785
18. Ramachandran VS, Malhotra VM, Jolicoeur C, Spiratos N (1998) Superplasticizers: properties and applications in concrete, CANMET, Canada
19. Flatt RJ, Houst YF (2001) A simplified view on chemical effects perturbing the action of superplasticizers. Cem Concr Res 31:1169–1176. https://doi.org/10.1016/S0008-8846(01)00534-8
20. Spiratos N, Page M, Mailvaganam NP, Malhotra VM, Jolicoeur C (2006) Superplasticizers for concrete fundamentals, technology and practice. Handy Chemicals, Canada
21. Chatterji S (2003) Freezing of air-entrained cement-based materials and specific actions of air-entraining agents. Cem Concr Res 25:759–765
22. Cerulli T, Pistolesi C, Maltese C, Salvioni D (2003) Durability of traditional plasters with respect to blast furnace slag-based plaster. Cem Concr Res 33(9):1375–1383. https://doi.org/10.1016/S0008-8846(03)00072-3
23. Cultrone G, Sebastian E, Ortega Huertas M (2005) Forced and natural carbonation of lime-based mortars with and without additives: mineralogical and textural changes. Cem Concr Res 5(12):2278–2289
24. Izaguirre A, Lanas J, Álvarez JI (2009) Effect of water-repellent admixtures on the behaviour of aerial lime-based mortars. Cem Concr Res 39:1095–1104
25. Silva BA, Ferreira Pinto AP, Gomes A, Candeias A (2020) Suitability of different surfactants as air-entraining admixtures for lime mortars. Constr Build Mater 256:118986
26. Seabra MP, Paiva H, Labrincha JA, Ferreira VM (2009) Admixtures effect on fresh state properties of aerial lime based mortars. Constr Build Mater 23:1147–1153
27. Fernández JM, Duran A, Navarro-Blasco I, Lanas J, Sirera R, Alvarez JI (2013) Influence of nanosilica and a polycarboxylate ether superplasticizer on the performance of lime mortars. Cem Concr Res 43:12–24
28. Pérez-Nicolás M, Plank J, Ruiz-Izuriaga D, Navarro-Blasco I, Fernández JM, Alvarez JI (2018) Photocatalytically active coatings for cement and air lime mortars: Enhancement of the activity by incorporation of superplasticizers. Constr Build Mater 162:628–648
29. González-Sánchez JF, Taşcı B, Fernández JM, Navarro-Blasco I, Alvarez JI (2020) Combination of polymeric superplasticizers, water repellents and pozzolanic agents to improve air lime-based grouts for historic masonry repair. Polymers 12(4):887
30. Burgos-Montes O, Palacios M, Rivilla P, Puertas F (2012) Compatibility between superplasticizer admixtures and cements with mineral additions. Construct Build Mater 31:300–309. https://doi.org/10.1016/j.conbuildmat.2011.12.092
31. Arizzi A, Cultrone G (2012) Aerial lime-based mortars blended with a pozzolanic additive and different admixtures: a mineralogical, textural and physical-mechanical study. Constr Build Mater 31:135–143. https://doi.org/10.1016/j.conbuildmat.2011.12.069
32. Falchi L, Müller U, Fontana P, Izzo FC, Zendri E (2013) Influence and effectiveness of water-repellent admixtures on pozzolana–lime mortars for restoration application. Constr Build Mater 49:272–280

33. Maravelaki-Kalaitzaki P (2007) Hydraulic lime mortars with siloxane for waterproofing historic masonry. Cem Concr Res 37(2):283–290
34. Izaguirre A, Lanas J, Álvarez JI (2010) Ageing of lime mortars with admixtures: durability and strength assessment. Cem Concr Res 40(7):1081–1095
35. Vejmelkova E, Konakova D, Cachova M, Keppert M, Cerny R (2012) Effect of hydrophobization on the properties of lime-metakaolin plasters. Constr Build Mater 37:556–561
36. Čechová E, Papayianni I, Stefanidou M (2010) Properties of lime-based restoration mortars modified by the addition of linseed oil. In: 2nd historic mortars conference HMC2010 and RILEM TC 203-RHM final workshop, 2010, Prague, Czech Republic.
37. Papayianni I, Pachta V, Stefanidou M (2013) Analysis of ancient mortars and design of compatible repair mortars: the case study of Odeion of the archaeological site of Dion. Constr Build Mater 40:84–92
38. Silva BA, Ferreira Pinto AP, Gomes A, Candeias A (2020) Comparative analysis of the behavior of integral water-repellents on lime mortars. Constr Build Mater. https://doi.org/10.1016/j.conbuildmat.2020.120344
39. Karapanagiotis I, Hosseini M (2018) Superhydrophobic coatings for the protection of natural stone. Adv Mater Conserv Stone 1:1–25
40. Izaguirre A, Lanas J, Álvarez JI (2010) Behaviour of a starch as a viscosity modifier for aerial lime-based mortars. Carbohyd Polym 80:222–228
41. Izaguirre A, Lanas J, Álvarez JI (2011) Characterization of aerial lime-based mortars modified by the addition of two different water-retaining agents. Cement Concr Compos 33:309–318. https://doi.org/10.1016/j.cemconcomp.2010.09.008
42. González-Sánchez JF, Fernández JM, Navarro-Blasco Í, Alvarez JI (2021) Improving lime-based rendering mortars with admixtures. Constr Build Mater 271:121887. https://doi.org/10.1016/j.conbuildmat.2020.121887
43. Alvarez JI, Veiga R, Martínez-Ramírez S et al (2021) RILEM TC 277-LHS report: a review on the mechanisms of setting and hardening of lime-based binding systems. Mater Struct 54:63. https://doi.org/10.1617/s11527-021-01648-3
44. Karatasios I, Katsiotis MS, Likodimos V, Kontos AI, Papavassiliou G, Falaras P, Kilikoglou V (2010) Photo-induced carbonation of lime-TiO_2 mortar. Appl Catal B-Environ 95:78–86
45. Ergenc D, Fort R, Santos Silva A, Veiga R, Sanz Arauz D (2018) The effects of DiloCarB as carbonation accelerator on the properties of lime mortars. Mater Struct 51:10
46. Cizer Ö, Ruiz-Agudo E, Rodriguez-Navarro C (2017) Kinetic effect of carbonic anhydrase enzyme on the carbonation reaction of lime mortar. Int J Arch Herit Conserv Anal Restorat. https://doi.org/10.1080/15583058.2017.1413604
47. Maravelaki-Kalaitzaki P, Agioutantis Z, Lionakis E, Stavroulaki M, Perdikatsis V (2013) Physico-chemical and mechanical characterization of hydraulic mortars containing nano-titania for restoration applications. Cement Concr Compos 36(1):33–41
48. Maravelaki N, Kapridaki C, Lionakis E, Verganelaki A (2014) Improvement of properties of hydraulic mortars with addition of nano-titania. In: Croccolo D (ed) Adhesives: mechanical properties, technologies and economic importance, chapter 4. Nova Science Publishers Inc, Bologna, pp 79–92
49. Theodoridou M, Charalambous E, Maravelaki-Kalaitzaki P, Ioannou I (2016) Amelioration of crushed brick-lime composites using nano-additives. Cement Concr Compos 68:77–87
50. Kapetanaki K, Kapridaki C, Maravelaki PN (2019) Nano-TiO_2 in hydraulic lime-metakaolin mortars for restoration projects: physicochemical and mechanical assessment. Buildings 9:236
51. Loganina VI, Sadovnikova ME, Jezierski W, Małaszkiewicz D (2017) Modification of lime mortars with synthesized aluminosilicates. IOP Conf Ser Mater Sci Eng (Online) 245(2):9. https://doi.org/10.1088/1757-899X/245/2/022023
52. Rigopoulos I, Kyriakou L, Vasiliades MA, Kyratsi T, Efstathiou AM, Ioannou I (2021) Improving the carbonation of air lime mortars at ambient conditions via the incorporation of ball-milled quarry waste. Constr Build Mater 301:124073. https://doi.org/10.1016/j.conbuildmat.2021.124073
53. Kyriakou L, Rigopoulos I, Ioannou I (2020) Use of ultra-fine mafic rocks for the enhancement of carbonation reaction in lime renders. Int J Arch Herit 14(7):1087–1097. https://doi.org/10.1080/15583058.2019.1709583
54. de Souza Oliveira A, da Fonseca MGO, Ferrara L, de Moraes Rego Fairbairn E, Toledo Filho RD (2021) An overview of a twofold effect of crystalline admixtures in cement-based materials: from permeability-reducers to self-healing stimulators. J Build Eng 41:102400. https://doi.org/10.1016/j.jobe.2021.102400
55. De Nardi C, Cecchi A, Ferrara L, Benedetti A, Cristofori D (2017) Effect of age and level of damage on the autogenous healing of lime mortars. Compos Part B Eng 124:44–157
56. De Nardi C, Bullo S, Ferrara L, Ronchin L, Vavasori A (2017) Effectiveness of crystalline admixtures and lime/cement microcapsules in engineered self-healing capacity of lime mortars. Mater Struct 50(4):1911–19112
57. Theodoridou M, Harbottle M (2018) Biological self-healing for preventing and repairing damage in geological construction materials. RAMS 2018-Recent appointees in materials science, pp 10–11/9/2018, Cardiff, UK
58. de Souza OA, Pereira AS, Lemos P, Guerra JP, Silva V, Faria P (2021) Effect of innovative bioproducts on air lime mortars. J Build Eng 35:101985
59. Lubelli B, Nijland TG, van Hees RPJ, Hacquebord A (2010) Effect of mixed in crystallization inhibitor on resistance of lime–cement mortar against NaCl crystallization. Constr Build Mater 24:2466–2472
60. Granneman SJC, Lubelli B, Van Hees RPJ (2018) Characterization of lime mortar additivated with crystallization modifiers. Int J Arch Herit Conserv Anal Restorat 12(5):849–858. https://doi.org/10.1080/15583058.2017.1422570
61. Lucas S, Ferreira VM, Barroso de Aguiar JL (2013) Incorporation of titanium dioxide nanoparticles in

mortars—Influence of microstructure in the hardened state properties and photocatalytic activity. Cem Concr Res 43:112–120

62. Stefanidou M (2010) The role of nanoparticles to salt decay of mortars, proceedings. In: 8th International Symposium on the Conservation of monuments in the Mediterranean Basin, Petra 2010
63. Nunes C, Slížková Z, Stefanidou M, Němeček J (2016) Microstructure of lime and lime-pozzolana pastes with nanosilica. Cem Concr Res 83:152–163
64. Duran A, Navarro-Blasco I, Fernández JM, Alvarez JI (2014) Long-term mechanical resistance and durability of air lime mortars with large additions of nanosilica. Constr Build Mater 58:147–158
65. Faria P, Duarte P, Barbosa D, Ferreira I (2017) New composite of natural hydraulic lime mortar with graphene oxide. Constr Build Mater 156:1150–1157. https://doi.org/10.1016/j.conbuildmat.2017.09.072
66. Singh LP, Karade SR, Bhattacharyya SK, Yousuf MM, Ahalawat S (2013) Beneficial role of nanosilica in cement-based materials–A review. Constr Build Mater 47:1069–1077
67. Zhang R, Cheng X, Hou P, Ye Z (2015) Influences of nano-TiO_2 on the properties of cement-based materials: Hydration and drying shrinkage. Constr Build Mater 81:35–41
68. Stefanidou M, Papayianni I (2012) Influence of nano-SiO_2 on the portland cement pastes. Compos B Eng 43(6):2706–2710. https://doi.org/10.1016/j.compositesb.2011.12.015
69. Theodoridou M, Kyriakou L, Ioannou I (2014) Salt crystallization resistance of nano- modified repair lime mortars. In: H de Clerq (Ed.) Proceedings of the 3rd international conference on salt weathering of building and stone sculptures, 2014.
70. Kapridaki C, Xynidis N, Vazgiouraki E, Kallithrakas-Kontos N, Maravelaki-Kalaitzaki P (2019) Characterization of photoactive Fe-TiO_2 lime coatings for building protection: the role of iron content. Materials 12(11):1847. https://doi.org/10.3390/ma12111847
71. Satish C (2003) History of architecture and ancient building materials in India. Tech Books International, New Delhi, India, p 315
72. Ravi R, Rajesh M, Thirumalini S (2018) Mechanical and physical properties of natural additive dispersed lime. J Build Eng 15:70–77
73. Gour Khalid A, Ramadoss R, Selvaraj T (2018) Revamping the traditional air lime mortar using the natural polymer–Areca nut for restoration application. Construct Build Mater 164:255–264. https://doi.org/10.1016/j.conbuildmat.2017.12.056
74. Shivakumar M, Singh A, Selvaraj T, Thangaraj S (2022) Production of the traditional organic mortars of padmanabhapuram palace—A characterization study on the simulated mortars for their compatibility. Buildings 12:1466. https://doi.org/10.3390/buildings12091466
75. Simon J, Thirumalini S, Simona R (2022) Evaluating the impact of organic addition and aggregate gradation on air lime mortar: new compatible green material for heritage application. Int J Arch Herit 16(5):681–691. https://doi.org/10.1080/15583058.2020.1836287
76. Nunes C, Slížková Z (2016) Freezing and thawing resistance of aerial lime mortar with metakaolin and a traditional water-repellent admixture. Constr Build Mater 114:896–905. https://doi.org/10.1016/j.conbuildmat.2016.04.029
77. Rodriguez-Navarro C, Ruiz-Agudo E, Burgos-Cara A, Elert K, Hansen EF (2017) Crystallization and colloidal stabilization of $Ca(OH)_2$ in the presence of nopal juice (opuntia ficus indica): implications in architectural heritage conservation. Langmuir 33:10936–10950
78. Kang SH, Kwon YH, Moon J (2020) Controlling the hydration and carbonation in lime-based materials: Advantage of slow carbonation in CO_2 curable construction materials. Constr Build Mater 249:118749
79. Theodoridou M, Kyriakou I, Ioannou I (2016) PCM-enhanced lime plasters for vernacular and contemporary architecture. Energy Procedia 97:539–545
80. Liu H, Wang X, Wu D (2019) Innovative design of microencapsulated phase change materials for thermal energy storage and versatile applications: a review. Sustainable Energy Fuels 3:1091
81. Lucas S, Senff L, Ferreira VM, Barroso de Aguiar JL, Labrincha JA (2010) Fresh state characterization of lime mortars with PCM additions. Appl Rheol 20:63162. https://doi.org/10.3933/ApplRheol-20-63162
82. Cunha S, Aguiar JB, Tedeu A (2016) Thermal performance and cost analysis of mortars made with PCM and different binders. Constr Build Mater 122:637–648
83. Pavlík Z, Trník A, Keppert M et al (2014) Experimental investigation of the properties of lime-based plaster-containing PCM for enhancing the heat-storage capacity of building envelopes. Int J Thermophys 35:767–782. https://doi.org/10.1007/s10765-013-1550-8
84. Frigione M, Lettieri M, Sarcinella A, Barroso de Aguiar J (2020) Sustainable polymer-based phase change materials for energy efficiency in buildings and their application in aerial lime mortars. Const Build Mater 231:117149. https://doi.org/10.1016/j.conbuildmat.2019.117149
85. Sarcinella A, De Aguiar JLB, Lettieri M, Cunha S, Frigione M (2020) Thermal performance of mortars based on different binders and containing a novel sustainable phase change material (PCM). Materials 13:2055
86. Iucolano F, Liguori B, Colella C (2013) Fibre-reinforced lime-based mortars: a possible resource for ancient masonry restoration. Constr Build Mater 38:785–789
87. Pereira de Oliveira LA, Castro-Gomes JP (2011) Physical and mechanical behaviour of recycled PET fibre reinforced mortar. Constr Build Mater 25:1712–1717
88. Izaguirre A, Lanas J, Álvarez JI (2011) Effect of a polypropylene fibre on the behaviour of aerial lime-based mortars. Constr Build Mater 25(2):992–1000
89. Chousidis N, Charalambous O, Zymaride M et al (2021) Mechanical reinforcement of lime pastes by electrospun cellulose acetate polymer fibers. Fibers Polym 22:676–684. https://doi.org/10.1007/s12221-021-0378-2
90. Andrejkovičová S, Velosa A, Gameiro A, Ferraz E, Rocha F (2013) Palygorskite as an admixture to air lime–metakaolin mortars for restoration purposes. Appl Clay Sci 83–84:368–374
91. Rosato L, Stefanidou M, Milazzo G, Fernandez F, Livreri P, Muratore N, Terranova LM (2017) Study and evaluation

of nano-structured cellulose fibers as additive for restoration of historical mortars and plasters. Mater Today Proc 4(7):6954–6965

92. Kesikidou F, Stefanidou M (2019) Natural fiber-reinforced mortars. J Build Eng. https://doi.org/10.1016/j.jobe.2019.100786
93. Çomak B, Bideci A, Salli Bideci Ö (2018) Effects of hemp fibers on characteristics of cement based mortar. Constr Build Mater 169:794–799
94. Gil L, Bernat-Masó E, Cañavate FJ (2016) Changes in properties of cement and lime mortars when incorporating fibers from end-of-life tires. Fibers 4(1):7. https://doi.org/10.3390/fib4010007
95. Stefanidou M, Papachristoforou M, Kesikidou F (2016) Fiber reinforced lime mortars. In: proceedings of 4th historic mortars conference (October 10–12 2016), Santorini, Greece
96. Moropoulou A, Bakolas A, Bisbikou K (2000) Investigation of the technology of historic mortars. J Cult Herit 1:45–58
97. Moropoulou A, Bakolas A, Moundoulas P, Aggelakopoulou E, Anagnostopoulou S (2005) Strength development and lime reaction in mortars for repairing historic masonries. Cement Concr Compos 27:289–294
98. Veiga RM, Fragata A, Velosa AL, Magalhaes A, Margalha G (2010) Lime-based mortars: viability for use as substitution renders in historical buildings. Int J Arch Herit 4:177–195
99. Pavlidou E (2012) Systematic analysis of natural pozzolans from Greece suitable for repair mortars. J Therm Anal Calorim 108:671–675. https://doi.org/10.1007/s10973-011-2039-y
100. Theodoridou M, Ioannou I, Philokyprou M (2013) New evidence of early use of artificial pozzolanic material in mortars. J Archaeol Sci 40(8):3263–3269. https://doi.org/10.1016/j.jas.2013.03.027
101. Groot C (2016) Repair mortars for historic masonry; Effects of the binder choice on durability. HERON 61(1):33–56
102. TC 203-RHM (Main author: Rob van Hees). RILEM TC 203-RHM: Repair mortars for historic masonry (2012) Mater Struct 45:1295–1302. doi https://doi.org/10.1617/s11527-012-9917-z
103. Papayianni I (1999) Utilization of high-volume fly ash concrete for roller compacted concrete dams. In: International symposium on concrete for sustainable development in the twenty-first century pp 126–142
104. Cenry R, Kunca A, Tydlitat V, Drchalova J, Rovnanikova P (2006) Effect of pozzolanic admixtures on mechanical, thermal and hygric properties of lime plasters. Constr Build Mater 20:849–857
105. Walker R, Pavía S (2011) Physical properties and reactivity of pozzolans, and their influence on the properties of lime–pozzolan pastes. Mater Struct 44:1139–1150. https://doi.org/10.1617/s11527-010-9689-2
106. Mehta PK (1987) Natural pozzolans: supplementary cementing materials for concrete. CANMET Spec Publ 86:1–33
107. Vejmelkova E, Keppert M, Kersner Z, Ravnanikova P, Cerny R (2012) Mechanical, fracture-mechanical, hydric, thermal, and durability properties of lime-metakaolin plasters for renovation of historical buildings. Constr Build Mater 31:22–28
108. Xu S, Wang J, Jiang Q, Zhang S (2016) Study of natural hydraulic lime-based mortars prepared with masonry waste powder as aggregate and diatomite/fly ash as mineral admixtures. J Clean Prod 119:1–10
109. Charola AE, Faria-Rodrigues P, McGhie AR, Henriques FMA (2005) Pozzolanic components in lime mortars: correlating behaviour, composition and microstructure. Restorat Build Monum 11(2):111–118. https://doi.org/10.1515/rbm-2005-5942
110. Mohammed S (2017) Processing, effect and reactivity assessment of artificial pozzolans obtained from clays and clay wastes: a review. Constr Build Mater 140:10–19
111. Morsy MS, Al-Salloum YA, Almusallam TH, Abbas H (2017) Mechanical properties, phase composition and microstructure of activated metakaolin-slaked binder. KSCE J Civ Eng 21(3):863–871
112. Costigan A and Pavía S (2010) Influence of mechanical properties of lime mortar on the strength of masonry. In: Válek J, Groot C and Hughes JJ (ed) Proceedings of 2nd Conference on Historic Mortars - HMC 2010 and RILEM TC 203-RHM final workshop, e-ISBN: 978–2–35158–112–4, 457–466, RILEM Publications SARL
113. Kaklis K, Maurigiannakis S, Agioutantis Z, Maravelaki-Kalaitzaki P (2018) Characterization of pozzolanic lime mortars used as filling material in shaped grooves for restoring member connections in ancient monuments. Int J Arch Herit 12(1):75–90. https://doi.org/10.1080/15583058.2017.1377313
114. Mosquera MJ, Silva B, Prieto B, Ruiz-Herrera E (2006) Addition of cement to lime-based mortars: effect on pore structure and vapor transport. Cem Concr Res 36:1635–1642
115. Arandigoyen M, Alvarez JI (2007) Pore structure and mechanical properties of cement–lime mortars. Cem Concr Res 37:767–775
116. Papayianni, I. and Stefanidou, M. 2003. Mortars for interventions in monuments and historic buildings. Advances in architecture, Vol 15, pp. 57–64 - 642003 eighth international conference on structural studies, repairs, and maintenance of heritage architecture, STREMAH VIII, 7–9 May 2003, Code 63223
117. Cizer O, Van Balen K, Van Gemert D (2008) Blended lime-cement mortars for conservation purposes: Microstructure and strength development. In: Ayala D, Fodde E (eds) Structural analysis of historic construction. Taylor & Francis Group, London
118. Silva BA, Ferreira Pinto AP, Gomes A (2015) Natural hydraulic lime versus cement for blended lime mortars for restoration works. Constr Build Mater 94:346–360