LITERATURE STUDY

# Microstructure, Chemical Processes and Experimental Investigation of Lime-Based Mortars

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### Introduction

The use of lime as a binder in mortars involves well-known inconveniences (i.e. slow setting and carbonation, high drying shrinkage, low mechanical strength) [1] that, in the last 50 years, have been overcome with the use of Portland cement. On the other hand, the negative effects of the use of Portland cement in the Architectural Heritage [2, 3] have forced workmen, restorers and scientists to find out alternative materials apt to improve the performances of lime-based mortars. In this sense, it is opportune to use specific admixtures, such as air-entraining and water-retaining agents and pozzolans, which improve workability in the fresh state, and mechanical strength, water permeability and frost resistance in the hardened state of lime-based mortars.

An extensive damage to the ancient masonry due to the use of cement-based repair mortars in the restoration works has been reported [4, 5, 6]. For this particular application the cement mortars are inadequate, due to their excessive brittleness, low plasticity, high elastic modulus and high content in soluble salts that leach out over time [7, 8, 3]. These salts produce anaesthetic layers on the surface but they can also introduce large crystallization pressures, damaging the structure [9].

The suitable repair mortar should be capable of accommodating movements [10]. Lawrence et al. [11] suggested that non-hydraulic lime mortars accommodate movement and better retain their integrity when subjected to stresses. Maravelaki-Kalaitzaki et al. [12] criticized cement-based mortars, which were used for restoration purposes over a substantial part of the 20<sup>th</sup> century, for their limited physico-chemical, mechanical, and aesthetic compatibility with the old masonry and old archeological surfaces. The adhesion of cement-based to the old historic materials was found poor, their thermal conductivity usually much higher, and the open porosity lower than of the lime-based mortars [13].

The authorities of cultural heritage in the Czech Republic discourage from using the common commercial renovation render systems in historical buildings a forbid their use if possible. They encourage using strictly the materials compatible with the original ones [14]. The increased hydrophobicity of render was objected to for the possible damage of the original historical masonry due to the accumulation of water in area close to rendered part and its freezing in the winter period. In general, mortars should be more permeable to vapor transport than the other masonry materials used so that water containing damaging ions, can evaporate before it spreads throughout the structure [15]. The use of small size aggregates in new render mortars and plasters was also criticized because of the optical properties of a façade. Therefore, in general, mortars based on lime or hydraulic lime without artificially increased volume of pores or cement additions, were recommended for repairs of historical buildings [16].

### **Constituents of Lime-Based Mortars**

The additions into modern concrete or lime-based mortars can be divided into two groups: *additives* (i.e. pozzolans, mineral fillers, ceramic powder) that are used in lime-based mortars with the aim to improve certain properties or obtain special performances mainly related to the increase of mortar strength, and *admixtures*, added in low amounts (i.e. not higher than a 5% of the total mass) in order to produce a permanent modification in the fresh or hardened mortar, such as density decrease, workability improvement or waterproofing [1].

#### **1.1 Lime**

Purity of commercially produced limes is regulated by the standard EN 459-1, which is not very strict. The mass of CaO + MgO in commonly used CL-90 lime hydrate should be higher than 90%. The presence of impurities does not have to be harmful, Vejmelková et al. [16] obtained the best mechanical properties of lime-pozzolana mortars containing lime of higher purities<sup>1</sup>.

The content of impurities, such as silica  $(SiO_2)$  and alumina  $(Al_2O_3)$ , was mainly responsible for hydraulic character of lime mortars. The reaction between lime and  $SiO_2$  and  $Al_2O_3$  leads to the formation of calcium silicates and aluminates [17]. However, contemporary high-purity limes without any additives lack the required durability [16] and therefore these minerals have to be added to lime in form of pozzolans.

#### **1.2 Pozzolans**

Pozzolans were used in combination with lime to improve the resistance to moisture of rendering mortars (and also freezing resistance [18]), the compactness of floor bedding mortars and the mechanical strength of structural mortars [19, 20]. Nowadays, the addition of pozzolanic additives (e.g. fly ashes, silica fumes and calcined clays) to aerial lime mortars is recommended because they are responsible for good properties in the early age, high values of mechanical strength, low water permeability, good cohesion between binders and aggregates and durability [1, 8].

In the study of Vejmelková et al. [16] the biggest strength and fracture energy was obtained in case of mortars containing white air-slaked lime (CL90) Čertovy schody having a great purity, 98% of CaO + MgO (97.4% CaO, 0.6% MgO, 0.13% SO<sub>3</sub> and 0.1% SO<sub>2</sub>)

Pozzolanic mortars are able to harden in high relative humidity (that is why they are called *hydraulic*) or when the access of  $CO_2$  is limited as in case of mortars supporting glazed tiles [8].

The lime reactivity of a pozzolanic material depends on its particle size and surface area and on its mineralogical composition. Pozzolans containing large proportions of amorphous silico-aluminates, having particles of small average mean diameter and relatively high specific surface, exhibit high reactivity [21].

#### 1.2.1 Metakaolin

Metakaolin is an artificial pozzolan obtained by the calcination of kaolinitic clays over a specific temperature range. It is a material with pozzolanic properties and can be added to lime mortar mixes to provide improved mechanical and water behavior characteristics for use in conservation mortars. From a chemical point of view, the metakaolin consits of minerals enabling hydraulic reactions – siliceous content is always high (around 60%) as well as  $Al_2O_3$  content (around 30%) [8].

The fact that mortars prepared with a lower amount of metakaolin (10% of MK on the total binder) show slightly higher peaks of unreacted lime (i.e. portlandite) in their X-ray diffraction patterns confirms that a faster lime consumption does not indicate a quicker carbonation but only the development of hydrated phases [1].

Vejmelková et al. [22] reported that the suitable amount of metakaolin<sup>2</sup> in the lime-pozzolana binder is 20%. According to their study this dosage of metakaolin makes the mortar significantly stronger in comparison with pure lime, but the water vapor transport properties are not much negatively influenced by the metakaolin addition.

#### 1.2.2 Crushed Bricks

Powdered ceramic material from tiles and pottery was used since early Hellenistic time and later on it has been used in Europe especially in the Mediterranean countries. The use of crushed bricks in the joints of load-bearing masonry walls became frequent during the latest part of the Roman Empire as pozzolanic material when there was no volcanic material in the region available. This was probably the case for the Roman mortars used for the Hadrian's wall in Britain, where crushed ceramic material was added to the lime binder [23]. The use of crushed bricks was continued during the Byzantine period [7], and it has probably been less common during medieval times. The use of crushed or finely ground bricks as an aggregate in ancient mortars were known as *Horasan* in Turkey [24], *Surkhi* in India, *Homra* in Arabic countries [25], and *cocciopesto* in Roman times [26].

From early Hellenistic to early Byzantine times crushed ceramics seem also to be preferred in mortars related to water-bearing constructions and to protect the inside of walls from moisture, typically in baths, canals and aqueducts [27, 28]. Fine brick particles were mainly used for rendering and for the upper layers of floors, whereas larger crushed brick particles were recommended not only for masonry walls, arches, and foundations, where high humidity or water were present, but also to improve the performance of mortars under normal conditions [29]. Crushed pottery and tiles have been used but clay minerals themselves can gain a distinct pozzolanic activity when fired at temperatures

<sup>&</sup>lt;sup>2</sup> finely ground metakaolin Mefisto K05,  $D_{50} = 4.82 \ \mu m$ ,  $D_{90} = 9.31 \ \mu m$ , specific surface 13.06 m<sup>2</sup>/g, with main components SiO<sub>2</sub> (58.7%) and Al<sub>2</sub>O<sub>3</sub> (38.5%), K<sub>2</sub>O (0.85%), Fe<sub>2</sub>O<sub>3</sub> (0.72%), TiO<sub>2</sub> (0.5%), MgO (0.38%), and CaO (0.2%), produced by České lupkové závody Inc., Nové Strašecí [16]

between 600 and 900°C [30].



Figure 1.1: Reactivity rim around brick fragment indicating pozzolanic reaction [31]

The amorphous components of the crushed brick aggregate are mainly represented by aluminosilicates, which react with lime (the lime makes the interfacial surface alkaline and causes chemical reaction) to produce calcium silicate hydrate and/or calcium aluminate hydrate at the bricklime interface, giving the mortars a hydraulic character [32]. The penetration of lime into the ceramic and the consequent reaction, transform the microstructure of the ceramic by shifting the pore radii into smaller pores [33]. The rim of hydration products around the brick fragment can be seen in Figure 1.1 [31]. The size of crushed brick particles directly influences its hydraulic reactivity – smaller particles have a bigger surface per volume, resulting in higher porosity.

### **Chemical Processes**

The main phase formed in mortars is calcite because of the carbonation of lime. The portlandite amount found in mortars after only 28 days of carbonation is low compared to the quantity that is likely to be found in aerial lime-based mortars without admixtures. This is because a part of the portlandite dissolved in water transforms into calcite (i.e. carbonation) while another part is involved in the lime-pozzolan reactions (i.e. hydration) favored by the alkaline environment [1].

#### 2.1 Carbonation

Romans forbade the use of any lime putty that was less than three years old. The effect of extended exposure of lime putty to water causes that portlandite crystals,  $Ca(OH)_2$ , undergo both a significant size reduction and a shape change from prism to platelike crystals [34, 5].

Carbonation is a chemical process whereby slaked lime, or portlandite,  $(Ca(OH)_2)$  reacts in solution with atmospheric carbon dioxide  $(CO_2)$  to form calcium carbonate, or calcite  $(CaCO_3)$  which is significantly stronger and less soluble that the portlandite it replaces:

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$$
 (2.1)

Therefore, for carbonation the presence of water is essential, since it requires the dissolution of  $CO_2$ and  $Ca(OH)_2$  is accessed by the  $CO_2$  in its dissolved state [35]. There are five stages involved: 1. diffusion of gaseous  $CO_2$  through pores of mortar, 2. dissolution of the  $CO_2$  in the pore water, 3. dissolution of  $Ca(OH)_2$  in the pore water, 4. solution reaction between  $Ca(OH)_2$  and  $CO_2$ , and 5. precipitation of solid  $CaCO_3$  [11].

Carbonation is more extended on the surface than in the core of mortar samples [1]. The uncarbonated calcium hydroxide is highly alkaline and phenolphthalein stain very well indicates where the carbonation process is not complete.

At a medium term (since 28 to 182 days, approximately), the mortars start to gain weight (Figure 2.1). This is due to the fact that the water excess has been eliminated and the mortars have begun the carbonation process. However, this weight increment is low and the carbonation is not complete after 1 year.

Carbonation changes the microstructure of the mortar, improves mechanical properties and affects pore structure, and hence water transport characteristics [11].



Figure 2.1: Weight losses in dependence on curing time in hydraulic lime mortars with different aggregates [17]

### 2.2 Hydration

Calcium aluminate and silicate hydrates of variable steichiometry are formed after activation of the alluminate and silicate phases of metakaolin, in presence of calcium hydroxide (i.e. portlandite) and water [36]. By means of XRD analysis, three general hydrated phases were detected: CSH, ( $CaOSiO_2H_2O$ ) or calcium silicate hydrates; CASH, ( $CaOAl_2O_3SiO_2H_2O$ ) or calcium alumina silicate hydrate; and CACH, ( $Ca_4Al_2(CO_3)(OH)_12 \cdot 6H_2O$ ) or monocarboalluminate. The latter is one of the mono-phase calcium hydrates and derives from the reaction between the reactive alluminates of metakaolin and the  $CO_3^{2-}$  ions present in mortars [37].

Calcium silicate hydrates (CSH) and calcium alumina silicate hydrate (CASH), which are among the main hydrated phases formed at room temperature after pozzolanic reaction of metakaolin [36, 38, 39].

Lanas et al. [17] found that at early ages  $C_3S$  hydration is responsible for the strength increment, at medium term compressive strength lightly changes and at long term a new strength increment can be expected due to hydration of  $C_2S$  and portlandite carbonation. However, mixes with binder to aggregate ratios lower than 1 : 3 do not show any strength increment due to low amount of hydraulic lime in these mortars.

Vejmelková et al. [16] reported that a pure lime mortar without addition of pozzolans exhibited about 10% higher porosity than those containing metakaolin (20% of binder mass). It can be explained by formation of hydration products, such as CSH gels,  $C_4H_{13}$ ,  $C_3AH_6$  and  $C_2ASH_8$  as a result of the reaction of silicate and aluminate ions with Ca<sup>2+</sup> ions [40].

### Microstructure

#### 3.1 Morphology of Hydrated Phases



Figure 3.1: Calcite crystals in a lime paste [41]

When hydrated lime in powder form is mixed with water, hydrated crystals are formed which later produce thick plates of carbonated lime. The structure of this matrix is not very condense. Many discontinuities are present. In the early stages, the hydrated lime pastes cured at high humidity are loose in comparison with those cured under dry conditions. However, at later stages the latter pastes show more intense cracking than those of the former wet cured lime pastes. SEM analysis of this paste showed crystals of size less than  $10 \,\mu\text{m}$  at the age of 28 days (Figure 3.1). The strength of lime pastes at that age ranges from 0.5 to 2 MPa. It is obvious that it is a soft binder of low strength capacity, very susceptible to cracking due to shrinkage because of the carbonation process and removal of the absorbed water from the capillary pores. The presence of autogenous stresses in the lime matrix is higher than its tensile strength capacity, and cracking easily occurs.

The products of pozzolanic reaction can be seen in Figure 3.3, showing metakaolin-lime mortar at the age of 28 days [8].

Aluminates (as CACH) crystallize as hexagonal platelets similar to those of portlandite but normally larger and thinner [36] (see Figure 3.4a-f). Generally, they appear tangled in the mortar matrix, in some cases randomly oriented (Figure 3.4a and b) and in others in form of overlapping sheets ori-



Figure 3.2: SEM micrograph of well carbonated mortar (with bioclastic aggregates) [11]



Figure 3.3: Evidence of pozzolanic reaction (CSH formation) in metakaolin-lime mortar [8]

ented perpendicularly to the basal plane (Figure 3.4c-e). Bigger quantity of aluminates was found in CCM at higher magnifications (Fig. 4a and c). The common morphologies that calcium silicate hydrate (CSH) phases adopt after precipitation are: fibres (Figure 3.4g), flakes (Figure 3.4h), honeycomb structure (Figure 3.4i), and reticular network (Figure 3.4j). In some mortars all the calcium silicate morphologies can be observed whilst in the other only fibres and flakes of CSH can be recognized [1].

#### 3.2 Porosity

It has been shown by Arandigoyen et al. [42] that carbonated lime-pastes have two pore size peaks – one peak between 0.5 and 1.0  $\mu$ m in diameter, varying according to kneading water used in mortar preparation, and a smaller peak between 0.1 and 0.2  $\mu$ m, independent of the water content in the fresh mortar. The peak seen between 0.1 and 0.2  $\mu$ m results from the change in microstructure of the binder due to recrystallization of portlandite crystals to smaller carbonate crystals. It is likely that the peak



Figure 3.4: FESEM images of hydrated phases (aluminate and silicate) formed after pozzolanic reaction in mortars: hexagonal plates randomly oriented observed in (a) and (b), overlapping sheets of aluminate observed in (c) and (d), aluminate crystals oriented in one direction (e), CSH fibres in (g) and CSH isolated flakes in (h), honeycomb structure (i) and reticular network (j) of silicated hydrates [1]

at 1.0  $\mu$ m is structural, created in the binder by the dispersion of the binder in the matrix [11].

Arizzi and Cultrone [1] found that the main peak in pore size distribution curves (PSDs) obtained in lime and metakaolin-based mortars corresponds to pores whose radius is comprised between 0.1 and 1.0  $\mu$ m and whose volume is the most influent on the total porosity of mortars (see 3.5).

The presence of metakaolin makes the dependence between water content and pore size distribution not as clear as in lime mortars, since a part of the water is consumed during the pozzolanic reaction [1]. The peak of pores whose radius is in range between 0.01 and 0.1  $\mu$ m is present due to presence of metakaolin in the mix [43] forming the network of hydrated calcium silicates (having the pores in range 0.01 < r < 0.04  $\mu$ m, according to Pandey and Sharma [44]). The amount of these small pores is increasing with the increasing amount of metakaolin in a mortar [1]. This is in agreement with findings of Vejmelková et al. [16].



Figure 3.5: Pore size distribution curves of external (Ex) and internal zones of mortar (lime : metakaolin = 8:2, binder : sand = 1:3 volumetrically) – investigated by mercury intrusion porosimetry [1]

An increase of pores having approximately 0.1  $\mu$ m in diameter can be observed in all mortar types which is attributed to the transformation of portlandite (Ca(OH)<sub>2</sub>) to calcite (CaCO<sub>3</sub>). Also an increase of pores smaller than 0.03  $\mu$ m can be observed [11].

The above stated is in agreement with findings of Kramar et al. [31], Vejmelková et al. [16] and Lawrence et al. [11] who claims that formation of pores about 0.1  $\mu$ m is irrespective of the type of aggregate.

According Lawrence et al. [11], pores smaller than  $0.03 \,\mu\text{m}$  present in pure lime mortars are not present in pure lime pastes because the carbonate crystals in the mortars are attaching themselves to aggregate particles and create tiny pores at the interface between aggregate and calcite.

Mosquera at al. [15] reported about the peak indicating presence of 15  $\mu$ m pores in pure lime mortars. They found that addition of Portland cement contributes to reduction of these big pores and also of the total porosity. In natural hydraulic mortars the presence of 15  $\mu$ m pores was not reported (see Figure 3.6).

Lanas et al. [17] investigated hydraulic mortars and they found a correlation between pore size distribution and aggregates used. From their results it can be concluded that hydraulic lime contributes with pores  $0.3 \mu m$  in diameter. Such peaks were sharper with decreasing amount of aggregates in the



Figure 3.6: Pore size distribution curves for pure lime, hydraulic lime and lime-cement mortars – investigated by mercury intrusion porosimetry [15]



Figure 3.7: Results from mercury intrusion porosimetry: pore size distribution for hydraulic mortars with different aggregates after 365 days [17]

mix, irrespective of the aggregate used (see 3.7). They also reported that large binder amounts cause porosity increment but also strength increment, because more CSH phases can be formed and the carbonation is faster and more complete. Another finding of Lanas et al. [17] is that round-shaped aggregates increase large pores (> 50  $\mu$ m) due to bad cohesion between the binder and the aggregate, causing the strength reduction. Angle-shaped aggregates lead to a better packed structure, a large pore decrease and consequently a strength increment.

Clearly visible cracks can be observed in case of pure lime mortar (see Figure 3.8a). These drying cracks are not present in such a big amount in case of hydraulic mortars (Figure 3.8b) [15].



Figure 3.8: Backscatter SEM images of mortars: (a) aerial lime-based (b) hydraulic lime-based [15]

The pore structure of a mortar influences its performance and durability. In cement and hydraulic lime mortars, the compressive strength decreases with increasing porosity, whereas in air lime mortars this relationship is less evident [45].

### **Mechanical Properties**

#### 4.1 Influence of Metakaolin

Arizzi and Cultrone [1] claim that 20% of metakaolin content produces a larger enhancement of the mechanical strengths, compared to a 10% metakaolin content, which has produced values of compressive strength between twice and three times lower. This is in agreement with a study carried out by Wild et al. [46] on metakaolincement pastes. They found that the optimum amount of metakaolin replacing ordinary Portland cement for the strength enhancement was about 20%. For comparison, indicative values of strength and porosity of mortars based on cement, pure lime and lime-pozzolan combination are given in Table 4.9.

Table 4.1: Mechanical and physical characteristics of mortars with different binding systems at the age of 90days [47]

binder	compressive strength [MPa]	open porosity [%]
lime	0.25	22.81
lime + pozzolan $(1:1)$	6.56	19.66
lime + pozzolan + cement $(1:0.5:0.5)$	7.34	16.72

Vejmelková et al. [16] also concluded that in comparison with pure lime mortars a remarkable improvement of mechanical properties can be achieved if the 20% is replaced by metakaolin. According to their study the compressive strength increased up to five times, the bending strength up to three times and fracture-mechanical parameters even more. They also found that the water vapor diffusion coefficient decreased only about 25% in comparison with pure lime mortar, which is a positive finding since vapor permeability is needed for a movement of moisture from the original masonry.

Velosa et al. [8] tested samples of metakaolin-lime mortars (lime : MK : sand = 1 : 0.5 : 2.5) and compared the results with pure lime and cement mortars (see Figures 4.1 and 4.2. They used 3 kinds of metakaolin, MK1 was richer in quartz whereas MK2 and MK3 in kaolinitic minerals. MK1 showed evidences of disordered kaolinite whereas MK3 more ordered kaolinite and MK2 showed an intermediate composition. MK2 was also richer in Al<sub>2</sub>O<sub>3</sub> and poorer in alkalies (sodium and potassium); MK1 and MK2 showed the opposite characteristics. It can be concluded, that mortars using metakaolin richer in well ordered kaolinite and richer in Al<sub>2</sub>O<sub>3</sub> (also being poorer in alkalies)

	$\mathrm{SiO}_2$	$Al_2O_3$	$\mathrm{Fe}_2\mathrm{O}_3$	MgO	CaO	Na <sub>2</sub> O	$K_2O$	$\mathrm{TiO}_2$	LOI
	[%]	[%]	[%]	[%]	[%]	[%]	[%]	[%]	[%]
MK1	62.62	28.63	1.07	0.15	0.06	1.57	3.46	0.36	2.00
MK2	59.90	32.29	1.28	0.17	0.04	0.24	2.83	0.36	2.80
MK3	62.48	28.72	1.01	0.13	0.03	2.45	3.55	0.34	1.20

Table 4.2: Elemental analysis (XRF) results of metakaolin samples [8]

showed better mechanical properties [8]. Table 4.2 provides an information about composition of the individual metakaolin samples and Table 4.3 informs about their pozzolanic reactivity.

Table 4.3: Pozzolanic activity of metakaolin samples [8]

sample	OH <sup>-</sup> concentration	CaO concentration
	[mmoles/l]	[mmoles/l]
MK1	38.80	7.19
MK2	43.20	7.70
MK3	52.19	5.56



Figure 4.1: Flexural strength of lime-metakaolin mortars (MK) compared with pure lime mortar (L) and cement mortar (C) [8]

The lower strength in mortars containing MK1 can be explained by lower firing temperature or inadequate grinding during production of metakaolin MK1, which acted partially as an aggregate.

The elastic modulus of lime-metakaolin mortars, tested by Velosa et al. [8] is low, as desired, compared to cement mortar (see Figure 4.3). The variations from 28 to 90 days are not significant. Only mortar containing metakaolin MK3 yielded relatively high Young's modulus, almost 5 GPa after 90 days.



#### Compressive strength

Figure 4.2: Compressive strength of lime-metakaolin mortars (MK) compared with pure lime mortar (L) and cement mortar (C) [8]



### Elastic Modulus

Figure 4.3: Elastic modulus of lime-metakaolin mortars (MK) compared with pure lime mortar (L) and cement mortar (C) [8]

#### 4.2 Influence of Crushed Bricks

The brick-mortar interface, investigated in depth over a range of  $120 \ \mu\text{m}$ , going from mortar to brick and considering as the interface the range  $30 - 50 \ \mu\text{m}$ , revealed the following significant features: Ca element decreases, while Si and Al compounds increase from mortar matrix to brick [4].

Mortar samples containing crushed bricks (usually of a bigger diameters) also show self-healing effects and proved to be resistant to continuous stresses and strains due to the above-mentioned presence of the amorphous hydraulic CSH formations, which allow for greater energy absorption without initiations of fractures and explain the good performance of the historic composites in resisting earth-quakes [4, 48].

In the case of large sized brick pebbles (even if reaction layers can be detected along the contact surface between the binder and the pebbles) the reaction cannot penetrate very far into the pebble. Hence, the reaction can only realize a better adhesion between the binder and the aggregate, a modification of the external surface of the pebble, but does not affect in a complete modification of the pebble itself [29].



Figure 4.4: Reaction ring at brick–mortar interface [4]

Silva et al. [32] investigated the influence of fine ceramics in cement-based mortars and found that its additions improves mechanical properties and contributes to a reduction of mortar shrinkage.

#### 4.3 Influence of Aggregates

The presence of aggregates causes the shrinkage reduction and improvement of mechanical resistance. However, too high content of aggregate may result in a mortar with poor cohesion between the grains and, consequently, with a lower strength [1].

In concrete, the interlocking of the aggregate and paste forms a transition zone, which is considered weak due to high porosity, formation of micro-cracks and different crystal sizes of the binder [49]. The incompatibility between the aggregate and the paste moduli of elasticity causes the development of micro-cracks at the aggregatematrix interface. However, it is well recognized that coarse aggregate particles act as crack arresters, as they restrict the shrinkage of the matrix, so that under an increasing load, extra energy is absorbed for the formation of a new crack, resulting in the gradual failure of the concrete [50, 49]. The demand for water is lower in case of bigger aggregates [41].

Arizzi and Cultrone [1] obtained increasing values of compressive and flexural strengths with increasing amount of aggregate (sand) in the mortar, but it is valid only to binder to sand ratio 1 : 6, then the trend was opposite. They also found that the use of a binder-to sand proportion higher than 1 : 6 does not lead to any change in the pore system (open porosity value and pore size distribution).

On the other hand, Lanas et al. [17] obtained the highest strength of hydraulic mortar with increasing binder content (see Figure 4.5 irrespective of the type of aggregate used. They disagreed with many authors claiming that binder to aggregate ratio equal to 1 : 3 is the most suitable for repair mortars since it results in the biggest strength and mortars having higher ratios suffer from shrinkage cracking. They also claim that use of siliceous aggregates results in mortar strength reduction, while limestone aggregates contribute to a higher strength.

Stafanidou and Papayianni [41] also obtained higher mortar strength for lime mortars of low binder to aggregate ratios (1 : 1.5, 1 : 2.5 and 1 : 3) containing fine sand 0–4 mm. However, the highest strength was obtained for mortars having the binder to aggregate ratio 1 : 2.5 (see Figure 4.7). Coarse aggregates have contributed to volume stability of the mortars since they restricted the volume changes – it has an impact on long term strength. When the coarse aggregates are used, good compaction is



Figure 4.5: Strength development in time for hydraulic mortars with different aggregates [17]

necessary to reduce voids and increase bond of lime paste with pebbles. The compaction results in an increased strength (see Figure 4.6) reduced water penetration and increased resistance of mortars to weathering [41].



Figure 4.6: Influence of compaction on the strength of lime mortars with coarse aggregates (L-Compac. = low compaction, H.-Compac. = high compaction) [41]

For pure lime mortars Lanas and Alvarez [45] suggested that siliceous aggregates provide small



Figure 4.7: Development of compressive strength in lime mortars with different binder to aggregate ratios [41]

radius pores, which obstruct an adequate  $CO_2$  flow through the mortar. As a consequence, the mortar carbonation decreases and the strength decrease. Nevertheless, limestone aggregates, increasing the amount of medium and large radius pores, allow mortar carbonation and improve mortar strength. They investigated the relationship between binder to aggregate ratio, porosity and compressive strength (see Figure 4.8).



Figure 4.8: Porosity and compressive strength versus percentage of lime in lime mortars tested after 365 days [45]

A similar study of pure lime mortar porosity and strength development in time for different aggregate sizes was done by Stefanidou and Papayianni [41], but the investigated mortar was rich in binder, having the binder to aggregate ratio 1 : 1.5. Their results can be seen in Figure 4.9.





#### 4.4 Influence of Mortar Age

Veiga and Carvalho [51] attributed the reduction of strength from the age of 28 days to the age of 90 days in the case of lime-metakaolin mortars (sometimes observed in other mortars containing pozzolans) to microcracking due to shrinkage, to which flexural strength is very sensitive. The same phenomena was observed by Vejmelková et al. [16] – they found that the highest values of the flexural strength were reached after 28 day in case of lime-metakaolin mortars while in case of pure lime mortars the strength was continuously increasing with time (see Tables 4.4 and 4.5). Similar results of early strength gain, followed by a slight reduction in strength (compressive and flexural) and stiffness were also reported by Giavarini et al. [52].

age	pure lime mortar	lime-metakaolin (4 : 1) mortar
14 days	0.73	5.50
28 days	0.67	6.73
56 days	0.72	6.64
90 days	1.43	7.03

Table 4.4: Compressive strength [MPa] of mortar samples [16]

Table 4.5: Flexural strength [MPa] of mortar samples [16]

age	pure lime mortar	lime-metakaolin (4 : 1) mortar
14 days	0.2	1.6
28 days	0.2	1.9
56 days	0.6	1.8
90 days	0.6	1.6

### Shrinkage

Lanas et al. [17] reported that measured shrinkage in hydraulic mortars with binder to aggregate ratio equal to 1:1 was ranging from 0.5% to 0.7%.

Stefanidou and Papayianni [41] found that in case of pure lime mortars the rate of shrinkage is dependent on the amount of aggregates (Figure 5.1) and their size (Figure 5.2). Probably, at later ages, cracks appeared to be blocked by the coarse aggregate.



Figure 5.1: Shrinkage of mortars having different binder to aggregate ratios [41]



Figure 5.2: Shrinkage of mortars having different size of aggregates [41]

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