



Evolution of the microstructure of lime based mortars and influence on the mechanical behaviour: The role of the aggregates

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HIGHLIGHTS

- Air lime mortars with optimized sands improve packing enhancing mechanical performance.
- Angular shape sands allow a better packing density. Stiffness was not influenced.
- Siliceous sand mortars have higher volume of macropores reducing strength.
- Aggregates with calcite increase mesopores and decrease macropores volume.
- Chemical reactions between aggregates and lime improve cohesiveness of the matrix.

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ABSTRACT

The pore structure of air lime mortars, formulated with aggregates of different mineralogy and grain size distribution, was evaluated with the aim of studying the influence of sand type on the microstructural modification over time and of establishing the relationship between these modifications and the mechanical behaviour.

The microstructure of the mortars was determined by mercury intrusion porosimetry at 28, 90 and 360 days, and the results were compared with its mechanical characteristics, namely compressive strength, flexural strength and modulus of elasticity.

A remarkable increase of mechanical strength over time was found for all mortars. An optimized grain size distribution of the sand (optimized compactness) proved to be favourable to air lime mortars compared with grain size distribution defined by standard EN 196-1, showing a good behaviour of the interlocked structure but also adequate porosity that favours lime carbonation.

A relationship between the mechanical properties and the porous structure was also established: large volumes in the range of macropores tend to reduce the mechanical strength while gel pores, related with the presence of hydraulic compounds, are generally linked to an increase in strength. Overall, the assessment of the microstructure indicates that the packing density and the shape of the aggregates are the dominant parameters influencing the pore size distribution of macropores; however, the mineralogy and related characteristics also have an important impact on capillary porosity.

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1. Introduction

Pore structure is a very important microstructural characteristic in building materials, not only because it influences the physical

and mechanical properties but also because it controls their in-service durability [1].

It is well known that the constituents of mortars (namely binders and aggregates), the binder/aggregate and water/binder ratios, the mixing procedures as well as the application affect the pore structure during the hardening process. Moreover, the physical and mechanical characteristics depend on the compactness, which is closely related to the porosity, in turn affected by pore size, pore space distribution and pore structure connectivity [1–6]. Therefore, materials with the same total pore volume can

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exhibit different performance, depending on the pore structure and more specifically on the relation between small and large pores [2,3].

Aggregates are, by weight or by volume, the major component of mortars. These granular materials are not involved, in general, in the hardening reaction of the mortar, but are referred as the “skeleton” of the mortars’ systems, with direct influence on their properties, such as mechanical strength and shrinkage [6–22].

In some previous works, the mineralogical composition of the aggregates is considered to affect the strength of the mortars [6–18,23–25]. *Tasong et al.* [7] refer that the mechanical behaviour of a given mortar depends on the mineralogical compounds that result from the hardening process of the binder and on the mineralogy and particle shape of the used aggregates.

Aggregates can be a source of silica, which can react in certain conditions with lime, leading to the formation of reaction rims at the edge of the grains and recrystallization along the pre-existing cracks in the grains with calcium silicate hydrates (C-S-H) formation, which are known to improve the mortars strength [23–25]. Siliceous sands are quite usual in formulation of the mortars and are mainly made of quartz, a silica mineral; however, in non-hydraulic lime mortars, limestone aggregates yield higher strengths than those containing siliceous aggregates [6,9–13]. This behaviour is attributed to the limestone aggregates’ structure, which is similar to the calcitic binder matrix, leading to a reduction of the discontinuities between the lime matrix and the aggregates [6,10]. *Arizzi and Cultrone* [11] point out that there is a noticeable difference in the textural properties of limestone and siliceous aggregates, with the former having more angular, rough and porous surface. It has been suggested that this affects the degree of cohesion between aggregate surface and binder, possibly leading to the higher strengths resulting from the use of limestone aggregate. However, *Pavía and Toomey* [14] found that mortars with natural hydraulic lime (NHL) and limestone aggregates were slightly weaker than those produced with siliceous and quartzitic aggregates, suggesting that an increase in the aggregate’s calcite content lowers the flexural and compressive strength of the mortar.

On the other hand, *Quiroga and Fowler* [16] concluded that the use of fines of basalt tend to yield higher strength than other aggregates studied (limestone and granite), while *Tasong et al.* [17] pointed out that basaltic rock produces (compared to limestone, quartzitic and siliceous ones) the most reactive aggregates studied, hence being able to improve the strength over time.

Lanas et al. [15] correlated the morphology of the aggregates with the strength and porosity of the mortars, concluding that round-shaped aggregates increase large pores thus causing a strength reduction. This is in agreement with the study of *Pavía and Toomey* [14], which concluded that angular aggregates tend to increase the mechanical strength and bulk density of mortars, simultaneously reducing porosity, water absorption and capillary suction.

The particle size distributions (PSD) of the raw materials influence some intrinsic properties including porosity, permeability and mechanical strength [1,19,22,26]. An adequate PSD allows the development of high mechanical strength in the mortars. The performance is enhanced through two mechanisms: a filling action, where the voids between grains of average size are filled by smaller grains, and a chemical reaction that allows an improvement of the interfacial transition zone (ITZ).

The study of *Pavía and Toomey* [14] indicates that aggregates with a small average particle size contribute to reduce porosity and water absorption and also to enhance the mechanical strength of mortars.

The amount and type of fine aggregates also have an effect on the drying shrinkage of mortars [20,21]. The results of *Felixkala et al.* [27] and *Ilangovana et al.* [28] clearly show that granite fines,

as a partial siliceous river sand replacement, have positive effects on the mechanical properties of high-performance concrete. However, it is important to point out that the fineness of granite fines could promote durability problems, such as alkali-silica reaction [28].

In addition, the indirect role of aggregates in the carbonation process is evident: the shape and grading of the aggregates influence the pore system of the mortars, while the speed and the degree of carbonation depend on the pore structure [11].

The carbonation is the primary chemical reaction that takes place in air lime mortars. During the carbonation process, the microstructure changes, as a result of the transformation of portlandite into calcite. Since calcite has different crystal structure and higher molar volume (Table 1) the porosity will depend on the polymorph formed during the precipitation of CaCO₃.

Arandigoyen et al. [29] demonstrated the influence of carbonation on the porosity of mortars prepared with hydrated lime and cement. These authors obtained a porosity decrease of ~10% as result of carbonation; it was also found that fewer pores of the range of 1 µm can be found and the highest increase is between 0.03 µm and 0.01 µm in lime pastes.

On the other hand, *Lawrence et al.* [30] assert that the pore size distribution is likely to have an influence on the rate of the carbonation reaction: in fact, pores under 0.1 µm do not interfere with carbonation and the increase of that range of pores during carbonation originates a decrease of the large pores that have a major contribution for carbonation, so reducing the rate of carbonation. *Lanas et al.* [10] demonstrated that an increase in porosity reduces the strength of mortars; however, they claim that, when the binder is not in excess, an increase in porosity can also enhance the carbonation and produce the opposite effect by increasing the strength.

Differences in the porosity of the aggregates can also have an impact on the overall mortar strength, due to differences in the diffusion of CO₂ through the specimen, which can lead to different rates of carbonation [13].

The formulation of replacement repair mortars used for preservation of ancient buildings is a complex process that must take into account the compliance with their functional demands, as well as of the construction elements where these mortars will be incorporated.

This study aims at determining the aggregates influence (grading and mineralogical composition) on the pore structure of lime binder mortars at 28, 90 and 360 days. The assessment of the pore structure in air lime matrixes is extremely important in order to fully understand their mechanical behaviour and to enhance their performance. In this paper, five aggregates with two size distributions were used, in a total of ten different mixes, which were prepared using the same binder. The flexural and compressive strength results of the air lime mortars will be compared with the pore size distributions obtained at the same ages.

In the next section, a description and a classification of the porosity of the mortars are presented, to help in the results’ discussion done in the following sections.

Table 1
Properties of the minerals that can be involved in the carbonation process [29].

Mineral	Crystal shape	Density (kg/m ³)	Molar volume (cm ³)	Δ volume (%)
portlandite	lamellar	2230	33.20	–
aragonite	fibrous	2930	34.16	2.9
calcite	prismatic	2710	36.93	11.2
vaterite	spherical	2540	39.40	18.7

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