Microstructural Characterization of Mortars

Mónica Sofia Lopes de Almeida Gominho

Civil Engineering Department, Instituto Superior Técnico, Universidade Técnica de Lisboa, Portugal

Abstract

The variability of the internal structure of the coating mortar makes the analysis and comparison of the various performance properties a complex task. To facilitate it, the use of advanced characterization and diagnostic techniques contribute to a better analysis of the chemical, mineralogical and microstructural mortars, and the establishment of the relevant performance parameters.

In the scope of thermal mortar properties, various formulations with light weight aggregates (expanded cork, expanded clay and silica aerogel) were used. The methodology of the study includes X-ray Microtomography (micro-CT), Electronic Scanning Microscope (SEM), X-ray Diffraction (DRX) and Infrared Spectroscopy Fourier Transform (FTIR), and Stereo Microscopy. Thus, it is intended to study the solid structure in terms of the aggregate, binder and aggregate/binder interface, and the porous structure regarding to the amount, shape, size and connecting pores. In addition, information was obtained on the components used and the products formed under the chemical reactions involved in the production and curing of the mortar.

The method proposed is based on the evaluation of the applicability of each microstructural characterization techniques and their potential for interpretation of results, in mechanical and physical tests, that are commonly performed in the laboratory in the hardened state. Based on this methodology, it was possible to identify a number of relevant parameters for the microstructural characterization of mortars and propose microstructure groups according to the type of connection and porous network. This systematization has shown to be useful for distinguishing the performance of thermal and reference mortar.

Keywords: Thermal mortars, microstructure, X-ray microtomography, electronic scanning microscope
1. Introduction

Modern society has created new challenges in the construction sector, requiring the use of more suitable materials in terms of performance, durability and sustainability (economic and environmental). Regarding to mortars, the challenge is to create new formulations with sustainable and innovative compounds in order to meet those needs.

In general, mortar characterization consists on the knowledge of physical, mechanical, chemical and mineralogical properties. This work gives emphasis to microstructural characterization, an issue that has increasingly been study by several researchers [2, 3, 5, 6, 12]. Microstructural characterization includes a detailed study of structural features considered at the microscopic level [1, 9, 10, 17, 18].

The concept of microstructure is not universal and varies in with the different fields of science and technology. In metallurgy and ceramics, the microstructure can be defined by the spatial distribution of elements and defects in a solid crystalline structure, comprising the grain orientation, defects and chemical composition [17, 8]. In the study by Costa et al. [3] the microstructure for cementitious materials is defined by the chemical reaction of cement with water, being influenced by several factors such as: type of cement, chemical and mineralogical composition, water/cement ratio, mixing process, conditions healing nature, amount and size of aggregates or other additions. Other authors consider the microstructure of a material as a structure of small size, with characteristics that can be observed under the microscope [7, 14, 16]. In this study, it is considered the definition of Gottstein [8] "microstructure is all internal structural features that affect the properties of the material", in other words how the constituents of the mortar are arranged on the microscale (below 1 mm) and its own nature [14].

What sets the difference between macrostructural characterization and microstructural characterization are techniques and the observation scale. In a macrostructural level, the techniques are used to provide general information with low magnification in the order of millimetre (photographic record, stereo microscopy) [19]. At the microstructural level techniques are used to provide great discrimination of materials, making use of high magnification and resolution (nano scale to millimetre), which require specialized equipment (Micro-CT, SEM) [4, 14]. These two observation levels are complementary, since most aspects observed at the macrostructural level originates or depends on the information at a microstructural level.

Silva and Libório [20] emphasize that most characterization studies covers the analysis of the macroscopic properties in detriment of microscopic properties. But the microscopic properties are the basis to understand the performance and the macroscopic properties of mortars. Elaqra et al. [11] mention that despite being known the existence of a relationship between the microstructure and the performance of a mortar, this relationship is not well understood. Mehta [15] and Burlion et al. [13] also mention that it is important to relate microstructure with macrostructure, to study and model the mechanical behaviour and properties of materials.

For these reasons, the microstructural characterization of mortars are highly relevant to analyse the composition, structure and interaction of the constituents (aggregates, binder and porous network structure), and it’s the basis for the study and optimization of new formulations with high performance.
In this study it’s present a methodology for microstructural characterization of mortars with lower thermal conductivity 0.2 W / mK with different insulating aggregates (expanded cork, expanded clay and silica aerogel). Industrial thermal mortars and sand reference mortars with and without admixtures, for comparison purposes.

The main goals of this study are:

- Characterize light weight mortars in terms of mineralogical and microstructural composition, taking into account the aggregate, the binder, aggregate/binder interface and the pore space, using Micro-CT, MEV, stereo microscope, FTIR and DRX;
- Establish microstructural parameters for comparison between mortars to facilitate their distinction;
- Grouping the different mortars according to their microstructure, in order to establish a correlation between their microstructural characteristics and performance parameters.

## 2. Materials and methods

A set of observation techniques was used to characterize different thermal mortars (Table 1.), taking into account the aggregate, the binder, the aggregate interface / binder and the pore space.

### Table 1. Composition of the studied mortars (produced in the laboratory)

<table>
<thead>
<tr>
<th>Mortars</th>
<th>Binder</th>
<th>Aggregate/ % replacement (in volume)</th>
<th>Dimensions of Isolant aggregated (mm)</th>
<th>water/cement</th>
<th>Additions /Admixtures</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>CEM II 32,5 N</td>
<td>60% aerogel + 40% expanded clay</td>
<td>0,5 to 2</td>
<td>1,01</td>
<td>Yes</td>
</tr>
<tr>
<td>B</td>
<td>CEM II 32,5 N</td>
<td>60% aerogel + 40% expanded clay</td>
<td>0,5 to 2</td>
<td>0,86</td>
<td>Yes</td>
</tr>
<tr>
<td>C</td>
<td>CEM II 32,5 N</td>
<td>100% aerogel</td>
<td>0,5 to 2</td>
<td>0,66</td>
<td>Yes</td>
</tr>
<tr>
<td>D</td>
<td>CEM II 32,5 N</td>
<td>100% expanded clay</td>
<td>0,5 to 2</td>
<td>0,55</td>
<td>Yes</td>
</tr>
<tr>
<td>E</td>
<td>CEM II 32,5 N</td>
<td>100% expanded clay</td>
<td>0,5 to 2</td>
<td>0,85</td>
<td>Yes</td>
</tr>
<tr>
<td>F</td>
<td>CEM II 32,5 N</td>
<td>100% sand</td>
<td>0,063 to 2</td>
<td>0,40</td>
<td>Yes</td>
</tr>
<tr>
<td>G</td>
<td>CEM II 32,5 N</td>
<td>100% sand</td>
<td>0,063 to 2</td>
<td>1,10</td>
<td>No</td>
</tr>
<tr>
<td>H</td>
<td>CEM II 32,5 N</td>
<td>100% sand</td>
<td>0,5 to 2</td>
<td>0,95</td>
<td>No</td>
</tr>
<tr>
<td>I</td>
<td>CEM II 32,5 N</td>
<td>100% sand</td>
<td>0,5 to 2</td>
<td>0,40</td>
<td>Yes</td>
</tr>
</tbody>
</table>
Table 2. Composition of industrial mortars

<table>
<thead>
<tr>
<th>Mortar</th>
<th>Binder</th>
<th>Replacement Volume (%)</th>
<th>Aggregate dimensions (mm)</th>
<th>Other Aggregates</th>
<th>Amount of water by bag (L/kg)</th>
<th>Additions /Admixtures</th>
</tr>
</thead>
<tbody>
<tr>
<td>J</td>
<td>Natural hydraulic Lime</td>
<td>Cork (SI %)</td>
<td>≤ 3</td>
<td>Diatomaceous Earth / Clay</td>
<td>0.55</td>
<td>Natural additions; polipropilene fibers; air introducers</td>
</tr>
<tr>
<td>K</td>
<td>lime/ white cement and syntetic binders</td>
<td>70-80% EPS</td>
<td>1.5 to 2</td>
<td>Sand (calcarious and silicious)</td>
<td>0.7</td>
<td>No specified</td>
</tr>
</tbody>
</table>

It is shown in a flow chart (Figure 1.), the procedure set to analyse the mortars as well the techniques applied and the objectives for this methodology.

Each of the techniques applied has a particular applicability and in each one, different aspects may be observed in different scales of observation. The stereo microscopy and the photographic record allow us to perform surface observations with low magnification. MicroCT allows us to make observations of the surface and in the interior of the sample with medium magnification (maximum pixel size around two micron). However this technique has limitations in distinguishing extremely porous materials, from the
air pores. SEM observations are limited to the surface but allow us to identify objects in the order of few nanometers. Some of the possible outcomes from each technique were compiled in the Figure 2.

**Photographic record**
- Analysis of the components’ spatial distribution
- Rough evaluation of the amount of de pores
- Rough evaluation of the amount of binder between the aggregates
- Identification and characterization of the aggregates (color, shape, size)
- Evaluation of the cohesion of the mortars through disaggregation by touch

**Stereo microscope**
- Analysis of the components’ spatial distribution
- Evaluation of the amount of the pores of macropore and micropores if the enlargement is high
- Evaluation of the amount of the binder between the aggregates
- Identification and characterization of the aggregates (colour, shape, size, cutting surface, amount)
- Identification and characterization of the porous structure (pores type, size, shape, location, connection and depth)
- Observation of the interface between materials

**MicroCT**
- Analysis of the components’ spatial distribution
- Evaluation or strict analysis of the amount of the pores
- Evaluation or strict analysis of the amount of binder
- Identification and characterization of aggregates (shape, size, amount and compactness)
- Identification and characterization of the porous structure (pores type, size, shape, location, connections and depth)

If the aggregate is too porous, it is not always possible distinguish the aggregates from the existing pores in the mortar. There’s a range of pores that are not covered by this technique, because they are too small.

**MEV**
- Analysis of the amount of pores in a define zone with a large scale up (up to 10000x)
- Analysis of the interface zone between the materials
- Analysis and identification of the crystals present in the binder
- Identification and characterization of aggregates (shape, texture, size and cutting surface)
- Identification and characterization of the porous structure (type, size, shape, location, connection and depth)

**FTIR e DRX**
- Determination of the binder’s mineralogical composition
- Identification of the phases or cristaline structures (DRX)
- Identification of the phases or amorph structures (FTIR)

Figure 2. Outcomes from the different techniques

The goal of this study is to collect data about the microstructure of thermal mortars and propose parameters for the microstructural characterization that are useful to compare their performance and that allow to interpret the results in physical and mechanical tests. The formulations of mortars studied in Table 1 (produced in the laboratory with a volumetric trace 1: 4) and Table 2 (thermal industrial mortars) vary with the insulating aggregates (expanded cork, expanded clay and silica aerogel). Also evaluated the replacement 40% of aerogel by volume of expanded cork or expanded clay aggregates. In the sand cementitious mortars, we evaluated the effect of grain size distribution of sand and addition.
3. Results and discussion

This section presents a comparative summary of mortars, which is based on the following set of parameters: i) the size of the aggregate pore; ii) cutting surface; iii) the thickness of the binder and mass percentage of binder; iv) aggregate binding/linker; v) estimate of the amount of pores and pore size in the binder; vi) indicator portlandite/calcite. In Tables 3 and 4 are exemplified some of the results obtained. Based on these parameters, we evaluate the influence of the type and grain size of the aggregates, the influence of the partial replacement of aerogel by other aggregates, and the influence of the addition of admixtures in the microstructure of the mortar. In the final part, it was set out a systematization of the studied mortars, taking into account the connection of the aggregate particles and the type of porous network, whose proposal aims to contribute to the understanding of its performance.

<table>
<thead>
<tr>
<th>Table 3. Parameters for microstructural analysis, comparison between the mortars A, B and C</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>A&lt;sub&gt;AG+AE&lt;/sub&gt;</strong></td>
</tr>
<tr>
<td><strong>Dimension of pores’ aggregate</strong></td>
</tr>
<tr>
<td><strong>Contact surface</strong></td>
</tr>
<tr>
<td><strong>Binder’s thickness and % of the binder in mass</strong></td>
</tr>
<tr>
<td><strong>Connection aggregate/binder</strong></td>
</tr>
<tr>
<td><strong>Evaluation of the amount and dimension of the pores in the binder</strong></td>
</tr>
<tr>
<td><strong>Portlandite/calcite indicator</strong></td>
</tr>
</tbody>
</table>

3.1. Influence of admixtures and the grain size distribution

The introduction of admixtures was evaluated only in pairs of sand mortar F-G and I-H. It’s quite clearly that the introduction of admixtures (F and I mortars) contributes to increase porosity, helps to a better distribution of the pores and change the morphology of the pores (Fig.3a and 3c).
Table 4. Parameters for microstructural analysis, comparison between the mortars D, H and F

<table>
<thead>
<tr>
<th></th>
<th>D&lt;sub&gt;AH&lt;/sub&gt;</th>
<th>H&lt;sub&gt;Control&lt;/sub&gt;</th>
<th>F&lt;sub&gt;Control&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pores’ dimension in the aggregate</strong></td>
<td>Expanded clay: 1-200 µm</td>
<td>Sand: pores couldn’t be identified</td>
<td>Sand with air intruducers: pores with imperceptible dimensions</td>
</tr>
<tr>
<td><strong>Contact surface</strong></td>
<td>Expanded Clay: Rough</td>
<td>Sand: Smooth</td>
<td>Sand: Smooth</td>
</tr>
<tr>
<td><strong>Binder’s thickness and % of the binder in mass</strong></td>
<td>40-50 µm</td>
<td>50-200 µm</td>
<td>40-50 µm</td>
</tr>
<tr>
<td></td>
<td>31.52 %</td>
<td>15.73%</td>
<td>15.73%</td>
</tr>
<tr>
<td><strong>Connection aggregate/binder</strong></td>
<td>Connection in “bridge”</td>
<td>Connection “with close links”</td>
<td>Connection in “bridge”</td>
</tr>
<tr>
<td><strong>Evaluation of the amount and dimension of the pores in the binder</strong></td>
<td>High amount of macropores; most of them connected; medium dimensions between 1-2 mm; High amount of pores with dimensions ≤1µm (observed in MEV)</td>
<td>Medium amount of macropores, most of them connected; average dimensions between 1-2 mm; High amount of pores with dimension ≤1µm (observed in MEV)</td>
<td>High amount of macropores, most of them connected; Average dimensions between 100-1000 µm; high amount of pores with dimension ≤1µm (observados no MEV)</td>
</tr>
<tr>
<td><strong>Portlandite/calcite indicator</strong></td>
<td>Medium level of carbonation (14)</td>
<td>Low level of carbonation (25)</td>
<td>High level of carbonation (9)</td>
</tr>
</tbody>
</table>

In mortars where air introducers weren’t used (H<sub>Control</sub> and G<sub>ref</sub>), we observe air bubbles with small dimensions (mm), sometimes erratically distributed (Fig.3b and 3d). In this case, we observe a network of intermediate pores following broadly the contour of the particles. In the same mortars, we notice that grain size distribution curve changes the pore network when aggregates have dimensions between (63 µm to 2 mm), pores tend to be smaller. This aspect may be attributed to differences in crystal nucleation of cementitious phases and particle compression. A well calibrated sand corresponds to the least number of crystallization nucleus, with more developed crystals and more connections between particles (Figure 3a and 3c). The cutting surface (rough or smooth) influences aggregate/binder interface. Particles with a rough cut surface at the interface have greater overlapping area and allow growth of acicular and tabular crystals, which in principle benefits adhesion values.

### 3.2. Type of aggregate

Comparing the three formulations C<sub>AG</sub>, D<sub>AH</sub> and E<sub>GC</sub>, it is possible to observe significant differences at microstructural level, taking into account that all have the same grading curve and the same volumetric trace. One of the differences between this mortars is the binder volume, which determines the space between aggregate particles. The volumetric differences of binder, can be related to aggregate density that settles the amount of cement introduced in the formulation (C-52.01%, 51.22% and D-E-31.52%). Other aspect is the link between binder and the aggregate, since it depends on the roughness of the surface and the hydrophobic characteristics of the aggregate.

Aerogel binding occurs through very smooth surfaces, observing sometimes detachment of particles. In the case of cork, there is a strong imbrication of the binder and the particles, given to the cellular nature
of the surface. These differences may strongly condition the physical properties of these mortars. In the case of expanded clay mortar, there is a low volume of binder, so it forms a thin layer on particles and concentrate a small amount of binder in limited areas (Fig. 4a).

3.3. Aerogel replacement by expanded clay or expanded cork
The substitution of 40% aerogel by expanded clay \((A_{AE})\) or expanded cork granulate \((B_{AG+GC})\) in the aerogel mortar, doesn’t change significantly the overall structural of the mortar. It’s possible to see that particles are separated by a significant amount of binder. The mixture of aerogel and cork provides the greatest amount of binder, this is counterbalanced by increasing the size of the intermediate pore size (Figs. 6a, b and 6c).

3.4. Composition of the binder paste
According to DRX analysis the presence of original or hydrated cementitious components is not very significant. However, it recognizes the formation of portlandite \((\text{Ca(OH)}_2)\) and calcium carbonate (calcite and vaterite) in variable quantities, as intermediate products or final carbonation of the cement. In the case of sand, there is a direct relationship between the type of porous network and the degree of carbonation. The existence of an open porous network promote the gas exchange necessary to the carbonation reaction.

3.5. Comparison with industrial mortars
The observations made at different scales in these two mortars \((J_{GC} \text{ and } K_{EPS})\) enable the identification of some components, referred in the product datasheets. It’s possible to see a good dispersion of aggregates. The volume of binder in \(J_{GC}\) (industrial cork mortar) is the largest when compare to all other mortars in the group (Fig. 6d). This volume can be related to the mass of binder (information not provided by the supplier). In the EPS mortar, the thickness of the binder paste is smaller, which may also be associated to the reduced mass of binder. This mortar \((K_{EPS})\) is extremely porous and the shape of large pores is adapted to the contours of the EPS particles (Fig. 6e).

3.6. Systematization taking into account the connection of the aggregate particles
The parameters listed in Tables 3 and 4 allow to group mortars, according to the connections of aggregate particles. It was settle three categories of mortars connection: (1) “in bridge”, (2) “with close links” and (3) “dispersed connection” of the aggregates.

The connection "in bridge" - connection established between particles where the contact area is small and limited to a certain number of points. The expanded clay mortar \((D_{AE})\) and the two sand mortars I and F are mortars with this type of connection (Fig. 4).This connection appears to be stronger (more compact), where the binder’s crystals have higher development and a least acicular morphology (needle shaped). In some mortars it’s difficult to see particles boundaries in tomographic images, in general it depends on the compacity and the thickness of the binder (Fig. 4). Mortars with this connection appear to have a solid interconnected skeleton.
The connection “with close links” – connection between particles held through a thin layer of binder paste. This connection has a reduced thickness of binder (tens of microns) and involves the whole aggregate. Mortars with this type of connection are mortars like $H_{\text{control}}$ (with admixtures and coarse particle size) and mortar $G_{\text{ref}}$ (without admixture and coarse particle size) (Fig. 5). The crystals formed in this connection have acicular or tubular extend morphology (Fig. 5a). Sometimes, the proximity of the grains is so high that it becomes impossible to distinguish their contours in tomographic images (Fig. 5b).

The “dispersed connection” of aggregates - connection between particles held by a thick layer of binder. This connection has a submillimetric thickness and constitutes a solid paste, usually made up of microcrystals acicular and tabular. The thickness of binder in mortars with smaller aggregates (Figs 6a, 6b and 6c) is thinner than in mortars with higher aggregates (Fig 6d and 6e). This type of connection is typically cohesive and involves the entire particle.

**3.6. Systematization taking into account the type of porous network**

The parameters listed in Tables 3 and 4, allowed to set three group mortars according to its porous structure. The “open porous network” is characterized by the coalescence of millimetric and submillimetric pores in the interstitial space. Mortars with this type of network are: expanded clay mortar ($D_{AE}$) and the two mortars with sand and admixtures ($I_{\text{control}}$ and $F_{\text{control}}$). These three mortars have a connection “in bridge” between the aggregates, which helps the existence of voids (Fig. 7). This type of network is determined by the arrangement of particles, so it is very influenced by the grading curve of the aggregate.
The “pore-channel network” – a number of pores connected to each other through channels with a small diameter (Fig.8). These channels appear because the binder doesn’t fill the entire space between the aggregates. Mortars with this type of network have large pores inside, generally they correspond to air bubbles or inter-particle spaces unfilled with binder paste. Mortars with “continuous microporous network” have the aggregates completely surrounds by binder and this particles are relatively separated from each other by a homogeneous layer completely interconnected by pores (Fig.9)

![Figure 7. Mortars with open porous network (Micro-CT images)](image)

![Figure 8. Mortars with “pore-channel network” (Micro-CT images)](image)

![Figure 9. Mortars with “continuous microporous network” (Micro-CT images)](image)

4. Conclusions

The microstructure analysis of mortars allowed to study its solid and porous structure, by the level of aggregate, binder-paste, aggregate/binder interface and pore space. Through this analysis, it was possible to determine the influence of type of aggregate, grain size and the presence of admixtures in mortars.

The proposed methodology using a set of observation techniques (with different scales) and the two mineralogical characterization tests, enabled the identification of several distinct parameters: i) the size of the aggregate pore; ii) cutting surface; iii) the thickness of the binder and mass percentage of binder iv) aggregate/binder connection; v) evaluation of the amount and dimension of the pores in the binder; vi) indicator portlandite/calcite.

The parameters above allow to distribute the microstructures according to the type of connection between particles of aggregate: (1) “in bridged”; (2) with “close links; and (3) dispersed connection. Regarding the type of porous network the following categories were distinguished: (1) open porous network; (2) the pore-type channel network; and (3) continuous microporous network. These groups have distinct characteristics, which should determine the performance of these mortars, a subject a bit explored in this work but that will be more developed in future work.
References


