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Original Research Article

# Impedance spectroscopy study of the effect of environmental conditions in the microstructure development of OPC and slag cement mortars

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ABSTRACT

In this work, the microstructure of mortars made with an ordinary Portland cement and slag cement has been studied. These mortars were exposed to four different constant temperature and relative humidity environments during a 180-day period. The microstructure has been studied using impedance spectroscopy, and mercury intrusion porosimetry as a contrast technique. The impedance spectroscopy parameters make it possible to analyze the evolution of the solid fraction formation for the studied mortars and their results are confirmed with those obtained using mercury intrusion porosimetry. The development of the pore network of mortars is affected by the environment. However, slag cement mortars are more influenced by temperature while the relative humidity has a greater influence on the OPC mortars. The results show that slag cement mortars hardened under non-optimal environments have a more refined microstructure than OPC mortars for the studied environmental conditions.



## 1. Introduction

The use of wastes in the cement industry is becoming more and more popular [1–5], because they provide a very important ecological and economical benefit. The reduction of $CO_2$ emissions and the lower energy consumption during the cement production are some of the most important advantages obtained by the use of wastes in cement manufacture.

Ground granulated blast-furnace slag, and its effect on the properties of cementitious materials has been the topic of a number of studies [6–9]. Many studies show that in laboratory conditions this kind of material has good service properties, even better than Portland cement [6]. This fact is due to the development of the hydration reactions of slag. These reactions densify the pore network of concrete due to formation of additional CSH phases, so the pore size distribution is shifted towards smaller pores (pore refinement) [10]. This microstructure refinement improves the service properties of mortars and concretes made with slag cements since early ages [11,12].

It is very important to study the microstructure of cementitious materials due to the relationship between microstructure and mechanical and durability properties [2,13]. There are some

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techniques for microstructural characterization, most of them destructive, such as mercury intrusion porosimetry (MIP) [14] and nitrogen sorption tests [15]. In recent years, the non-destructive technique of impedance spectroscopy (IS) has become quite popular for the study of the microstructure of cement paste, mortars and concrete. The main advantage of this technique is that it is non-destructive, allowing one to follow the development of the microstructure of the sample with time. This is not possible using destructive techniques.

The use of impedance spectroscopy is based on the idea of correlating dielectric and mechanical properties of a solid material [16,17]. The interpretation of the impedance spectra is made by fitting the measurement to an equivalent electrical circuit, where each one of the electrical elements in the circuit (resistances and/or capacitances) represents one of the aspects of the microstructure of the material. This technique has been widely used for OPC pastes, mortars and concretes [18–20]. Nevertheless, regarding materials prepared with cements which contain additions, this technique has rarely been used for fly ash cements [2,21,22], slag cements [22,23] or cements with other admixtures [22,24]. All of the above studies have been performed under optimal laboratory conditions, or in controlled drying processes [18]. Then, the technique of impedance spectroscopy has been never used for characterizing the development of the microstructure of slag cement mortars exposed to non-optimal hardening environments.

Real concrete structures are hardened in environmental conditions that depend on their geographical location. The temperature and relative humidity present in the environment may influence the development of slag and clinker hydration [25]. As a consequence, the microstructure of concretes and mortars made with slag cements [26,27] or with OPC must be dependent also on the environmental characteristics, mainly due to the presence or lack of water and the effect of the temperature on the hydration rate. This influence of temperature and relative humidity on the microstructure of these materials has been studied separately during early hardening stages, but rarely over longer time periods [25,28]. The influence of the combined effects of both temperature and relative humidity has scarcely been reported as well. The impedance spectroscopy technique has been proved to be very sensitive to the amount of water in the pores in cementitious materials [18,29,30], as well as to possible changes in the internal structure of the solid phase [2].

The objective of this work is to study the influence of the combined relative humidity and temperature environmental conditions on the microstructure, specially the solid phase, of OPC and slag cement mortars for long hardening periods, using the impedance spectroscopy and mercury intrusion porosimetry as a contrast technique.

## 2. Experimental setup

### 2.1. Sample preparation

The tests were performed on mortar samples, which were prepared using an ordinary Portland cement (OPC), CEM I 42.5 R (CEM I from now on), and a ground granulated blast-furnace slag (GGBS) cement (with a content of GGBS between 66 and 80% of total binder), III/B 42.5 L/SR (CEM III from now on), according to the standard UNE EN 197-1 [31].

Two different water to cement ratios, 0.4 and 0.5, were used. Fine siliceous aggregate was used according to the standard UNE EN 196-1 [32]. The aggregate to cement ratio was 3:1 for all the mortar samples.

The specimens were cast in cylindrical moulds of 10 cm diameter and 15 cm height. Samples were kept in 95% RH chamber and 20 °C for 24 h. After that time they were removed from their moulds and cut into slices of approximately 1 cm thickness.

### 2.2. Environmental conditions

Four different environmental conditions were studied, as has been indicated in Table 1. Environmental condition A was the optimum laboratory condition, of 20 °C and 100% relative humidity (RH). This condition was taken as a reference in order to compare the influence of non-optimal hardening conditions on the microstructure of the studied mortars. The other environmental conditions studied are representative of the average conditions of different climates present in the Iberian Peninsula or surrounding areas. The values of temperature and RH of these conditions are the annual average values for each climate. Condition B represents the Atlantic climate, present in the northern part of the Iberian Peninsula, at 15 °C and 85% RH. Environmental condition C (20 °C and 65% RH) is representative of a Mediterranean climate, present in the eastern part of Iberian Peninsula. Finally, a more extreme condition was studied, at 30 °C and 40% RH, called condition D.

These environmental exposure conditions (temperature and relative humidity) were achieved by using hermetically sealed recipients containing water or glycerol solutions, and these containers were placed into chambers with controlled temperatures. The appropriate concentrations of the glycerol solutions were selected in order to achieve the target relative humidity values, according to the standard DIN 50 008 part 1 [33]. The mortar samples were placed into the containers, avoiding contact with the solutions.

### 2.3. Mercury intrusion porosimetry

In order to check the validity of the interpretation of the impedance spectroscopy results, mercury intrusion porosimetry was used. This is a well-known and extensively used

**Table 1 – Characteristics of environmental conditions studied.**

| Condition | Temperature (°C) | Relative humidity (%) | Represented climate |
|---|---|---|---|
| Condition A | 20 | 100 | Laboratory condition |
| Condition B | 15 | 85 | Atlantic climate |
| Condition C | 20 | 65 | Mediterranean climate |
| Condition D | 30 | 40 | Extreme condition |