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Influence of leaching on the local mechanical properties of an aggregate-cement paste composite

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ABSTRACT

This paper presents an experimental study to characterize the mechanical behaviour, at the local scale, of sound and degraded concrete after leaching. An experimental protocol was developed to study the effects of the calcium leaching mechanism at the interfacial transition zone (ITZ) on the mechanical properties of the cement-aggregate interface and bulk paste of model material. The process of lixiviation with deionised water occurs very slowly. The experimental study in the laboratory was accelerated by replacing the water with an ammonium nitrate solution. To quantify the development and kinetics of degradation at the cemented bond, the concrete leaching fronts were characterized at different levels of degradation using phenolphthalein and local mechanical tests (compression and tensile) and performed on samples consisting of two aggregates bound by cement paste. Tests were performed on sound samples at different hydration times and on chemically degraded samples. The results show the effect of leaching on the mechanical properties of the samples and making it possible to correlate the progress of leaching to the evolution of these locally considered properties. The experimental results show that there is an ITZ effect on the alteration of the mechanical properties due to leaching.

1. Introduction

The chemical degradation of concrete by leaching is the result of the liquid attack of (water with a low pH compared with the pore fluid) leading to the dissolution of the cement paste hydrates [1]. This is particularly true when the hydrates contain calcium (Ca^{2+}) as the main chemical component of the cement paste. For this reason, simplified approaches avoid the need to consider all the elementary chemical mechanisms and focus on the evolution of calcium ion concentration in the liquid and solid phases. Cement degradation is produced by several dissolution fronts depending on the relative solubility of each hydrate. The portlandite (CH) contained in cement paste is the first mineral to be completely dissolved, followed by the progressive decalcification of (C—S—H) over time [1,10,32]. This degradation, which develops from the surface by the diffusion of products dissolved towards the outside, leads to a significant increase in porosity. This, in turn, accelerates the process of material transport and the dissolution of hydrates, leading to modification of the microstructure of concrete. The physicochemical and mechanical properties of the concrete are gradually altered with the evolution of the degradation rate, so a loss of stiffness and strength of the concrete are observed [28]. Most experimental studies of the leaching effect on the mechanical properties of cementitious materials are investigated for bulk materials [20,24,34] and show that the strength of the samples is significantly reduced after dissolution of the hydrates. It is also noted that the change in microstructure increases the ductility of chemically degraded material. However, the effect of leaching on the interfacial transition zone remains little studied in the scientific literature despite the fact that the presence of aggregates should influence the leaching process and mechanical behaviour of leached concrete [29]. So it is of interest to conduct investigations at the local scale in order to provide a better understanding of the mechanical behaviour of leached concrete and to characterize the effect of leaching at the Interfacial Transition Zone (ITZ) on the mechanical concentration at this scale.

The Interfacial Transition Zone (ITZ) is considered an additional phase introduced by the presence of aggregates [27]. It is

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characterized by a higher porosity than the bulk paste and a high concentration of portlandite [4,11,16,31,40] which increases calcium leaching. The work of Zheng and al. [39] and Winslow and al. [37] have also shown that ITZ provides a preferential path for the transport of water and ions. In order to characterize the lixiviation effect of the ITZ on mechanical properties, a series of experiments was carried out by Carde and François [9] on a cement paste and mortar, with or without silica fume, demonstrating the effect of the ITZ on the mechanical properties of concrete. A recent experimental study by Buzzi and al. [8] showed that a leached rock-concrete interface at fixed leaching depth presents a radical change of behaviour because of the local loss of mechanical strength.

The process of lixiviation with deionised water occurs very slowly [1]. It has been shown that for concrete with a water/cement ratio of 0.4, a degradation depth of 4 cm would necessitate a leaching time about of 300 years and very few data are available in the literature about long term degradation of concrete by pure water [35]. In order to collect experimental data in a reasonable period of time, it is necessary to use an accelerated procedure. There are three methods to accelerate calcium leaching of cementitious materials: application of an electric field [17,33], using temperature [21] and using different chemical solutions to increase concentration gradients between the interstitial solution and the aggressive environment [13,32]. Ammonium nitrate (NH_4NO_3) has been chosen by several authors as a representative aggressive solution for these accelerated experiments [3,10,19,25,26,30,34]. The principle of accelerated degradation with ammonium nitrate is the penetration of NH^{4+} and NO^{3-} ions stemming from the dissolution of ammonium nitrate (Eq. (1)), in the interstitial solution by diffusion. The NH^{4+} ion reacts with the OH^- of the interstitial solution (Eq. (2)) and the NO^{3-} ion reacts with the calcium ions (Eq. (3)). This consumption of Ca^{2+} ions disturbs the chemical balance of the calcium between the liquid and solid phases, causing the dissolution of hydrates to free new calcium ions (Eq. (4)).

$$NH_4NO_3 \Leftrightarrow NH_4^+ + NO_3^- \tag{1}$$

$$NH_4^+ + OH^- \Leftrightarrow NH_4OH \Leftrightarrow NH_3 + H_2O \tag{2}$$

$$Ca^{2+} + 2NO_3^- \Leftrightarrow Ca(NO_3)^2 \tag{3}$$

$$Ca(OH)_2 \Leftrightarrow Ca^{2+} + 2OH^- \tag{4}$$

These accelerated degradation tests make sense when it comes to studying coupled processes: degradation-cracking [6,34], degradation-permeability [15,38] or degradation-mechanical behaviour [10,17,18,36]. This paper presents the effect of degradation on mechanical behaviour.

The proposed study is a contribution to the experimental characterization of the impact of chemical degradation by leaching of cementitious materials on mechanical properties at the cement paste-aggregate interface and the cement bond. In this context, a preliminary study was carried out on sound samples to characterize the behaviour of concrete during the hydration process. Interactions on this scale were experimentally analysed through mechanical testing of compression and tensile by means of a specific device.

1.1. Sample design and materials selected

For this study, the specimens consisted of two spherical “calcite” aggregates, the calcareous aggregates having been chosen for their high adhesion with the cement paste [22]. The aggregates were of the same diameter (8.3 mm), and were bound by a cement paste (cement CEM II/B 32.5 N) with a ratio water/cement of 0.5 with a circular cross section. Aitcin [2] found a reduction in the porosity and thickness of the interfacial transition zone in the High-Performance Concrete (HPC). Generally, the thickness of the ITZ of the HPC is limited to 12 μm, whereas that of ordinary concretes is about 50 μm. The distance between two aggregates was fixed at 1.7 mm, this being the average distance between two large grains observed in a concrete [12]. According to the definition of De Larrard [23], the distance between these two large adjacent aggregates separated by the cement paste is called the Maximum Paste Thickness (MPT). The MPT affects the compressive strength of concrete. When MPT decreases, the compressive strength increases. the interstitial space between aggregates mobilized by the ITZ also increase. Another explanation is crystal orientation of C–S–H, more the MPT is lower, more the hydrates are oriented, what improve resistance to compressive [23].

The geometric configuration of the sample is presented in Fig. 1 and the “material” data in Table 1. Simplified form of aggregates is used. These were perfectly spherical and of the same size, as originally proposed by Bisschop and Van Mier [5] to study the effect of aggregates on drying shrinkage in cement-based composites and by Burlion and al. [7] to study microcracking in leached cementitious materials. Demoulding was carried out after 24 h. The sample obtained had a diameter of 8 mm and a height of about 18 mm: This sample was regarded as representative. The samples were stored in water saturated with lime at 21 °C to reduce the leaching with water and the effects of drying shrinkage.

The specific sample geometry selected allowed us to consider the concrete as a three-phase material: aggregates, cementitious matrix and ITZ (Fig. 2). This allowed us to follow the direct effect of leaching at the ITZ interface and also in the bulk paste. Two sets of samples were examined: the first was kept in water saturated with lime at 21 °C (control series) and the second was immersed in the aggressive ammonium nitrate solution (degraded series), after

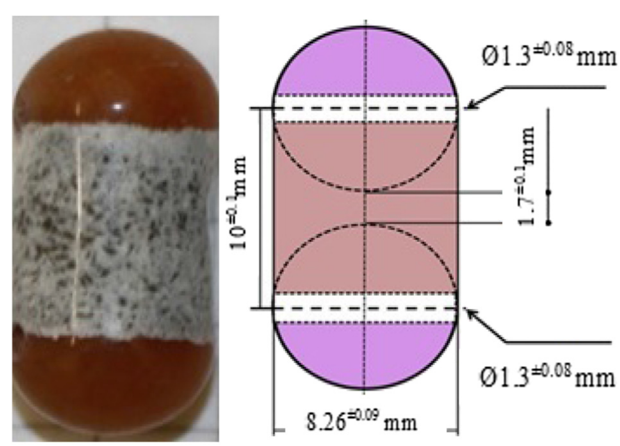

Fig. 1. Description of concrete samples at local scale.

Table 1
Material data of components used.

Aggregates	
Type	Calcite $CaCO_3$
Density	2.6 g/cm^3
Diameter	$8.26^{\pm 0.09}$ mm
Cement paste	
Formula	CEM II/B-LL 32.5 N
Water/Cement ratio	0.5