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X-ray microtomography: Application to microstructure analysis of a cementitious material during leaching process

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Abstract

Leaching of cementitious materials leads to an increase in porosity, which has important consequences on transport and mechanical properties. The present study outlines the characterization of microstructural evolution in a mortar subjected to a chemical attack, by means of a powerful non-intrusive experimental method, namely synchrotron X-ray computed microtomography. This innovative method is used to measure the variation of porosity in a leached mortar. Having described the effects of leaching in mortar (influences on microstructure and on mechanical properties), we present the accelerated leaching process and the microtomographic analysis that have been used to monitor it. We then investigate the capacity of this method to quantify the evolution of porosity during the leaching process. The method is validated by comparison of the results obtained with data available in the literature.

Keywords: Leaching; X-ray absorption; Synchrotron X-ray microtomography; Mortar; Porosity evolution

1. Introduction

Mortars and concretes are non-homogeneous materials and their macroscopic physical properties depend on their local characteristics (physical properties of elementary components, micro-geometry, transport properties at a micro-level, etc....). For instance, porosity and permeability of concrete are intimately related to the microstructure. Thus it is of importance to link the local and the macroscopic scale in order to study and model the mechanical behaviour and the transport properties of cementitious materials. This requires the knowledge of the 3D micro-geometry.

Characterization of concrete or mortar microstructure, and its evolution, often raises problems: very small observation scales, interference of measurement with the material (thin section obtained by mechanical abrasion, scanning electron microscope analysis made under vacuum after material desiccation, etc....). In order to avoid these problems, various innovative techniques are in development as, for example, acoustic emission analysis, infrared thermography or X-ray computed microtomography (XCMT).

Synchrotron XCMT finds applications in the study of sample microstructure without damaging it (biological samples, geological or industrial materials) [1–3]. The principle, similar to the medical scanner, consists of acquiring digital images of the material's X-ray absorption. This acquisition is undertaken at various angles: a three-dimensional image is then obtained by numerical reconstruction from the set of 2D-images. This technique has already been used for cementitious materials, particularly mortars: to measure internal crack growth in small specimens under uniaxial compressive loading [4,5]; to quantify damage parameters of asphalt concrete specimens [6]; to evaluate sulphate attack effects on cement paste [7] and mortar [8]; to observe water evaporation with respect to time during drying [9,10]; to characterize the early hydration of calcium aluminate's cement [11]; or to validate numerical

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simulations on mathematical analysis of concrete aggregate shapes [12] and on evolution of microstructure and transport properties of porous bricks [13]. The objective of the present study is to evaluate the increase in porosity induced by leaching of mortar by using synchrotron XCMT.

Concrete leaching is often the results of a fluid attack (pure water or water with very low pH compared to that of the pore fluid), and leads to the hydrolysis of cement paste hydrates [14–22]. This hydrolysis, which develops from the surface by diffusion of the aggressive agents through the material and diffusion of the dissolved products to the exterior, leads to an important increase in the cement paste porosity. As a consequence, liquid and gas transport properties are increased and the mechanical strength decreases [17,19–27]. Classical experimental methods used to study leaching effects [19,21,22,24–27] are often conducted by uncoupling the leaching process and other tests: for example, mechanical tests are usually performed after complete leaching. Furthermore, it remains very difficult to measure the evolution of porosity due to the leaching process, as numerous interactions occur between the experimental method and the leached material. One of the objectives of the present study is to obtain, with minimum interaction between the measurement and the degradation process, quantitative data about the effects of leaching on mortars. These unaffected data will then provide valuable validation of existing models [21–23,28–31] that predict loss of density of cement paste, evolution of porosity, or creation of porosity around aggregates.

Using synchrotron XCMT as a non-destructive characterisation method and performing the accelerated leaching with an ammonium nitrate solution, it is possible to carry out the microtomographic analysis on the same specimen during the leaching process. The evolution of the cement paste linear attenuation coefficient, the degradation kinetics, the leaching front position and the porosity increase can be determined without interfering with the material.

This paper is composed of three parts: first, we briefly describe mortar degradation mechanism caused by an aggressive solution, showing the importance of porosity increase and its mechanical consequences. In the second part, the chosen leaching process and the principles of synchrotron X-ray computed microtomography (XCMT) are clearly described. In the third part, the results obtained are depicted, demonstrating the capacity of the method to quantify the evolution of porosity and movement of the degradation front during leaching. Potential contributions of X-ray microtomography to a durability analysis of cementitious materials are emphasized.

2. Influences of porosity evolution due to leaching of cement paste

Calcium leaching is a coupled diffusion–dissolution process involving dissolution fronts that propagate through the sample. Under the influence of an aggressive solution (deionised water or ammonium nitrate solution), hydrolysis phenomena develop from the surface by diffusion of aggressive spices into the material and of dissolved products to outside, allowing the development of the leaching process by a front of dissolution/precipitation. With a decrease in pH of the pore solution, the first hydrate likely to be leached is the calcium hydroxide, also known as Portlandite. After complete dissolution of Portlandite, the calcium-silicates hydrates (C-S-H) are then decalcified [14,15]. Portlandite dissolution is associated with an important increase in capillary porosity, while the decalcification of C-S-H porosity results in only slight changes. Leaching kinetics is related to the material transfer properties: the lower the porosity, and the lower the degree of interconnection between pores, the lower will be the penetration of the aggressive solution.

Furthermore, there are some important differences between the bulk of the hydrated cement paste and the interface with aggregate (the so called interfacial transition zone—ITZ) in terms of porosity, hydration, and chemical composition. It can be assumed, in case of leaching, that the particular properties of this zone could contribute to an increase in mortar sensitivity compared to cement paste. Bourdette [16] estimated the role of aggregates in leaching a mortar, and identified only a weak influence of the ITZ. The chemical mechanisms are similar to those described by Adenot [14], as far as hydrated cement is the only reactive constituent. These results were confirmed by Carde and François [17,22]. An explanation of it could be the physical equivalence, in terms of porosity and transport properties, between the leached cement paste and this interfacial zone. On the other hand, the ITZ mechanical effect, in term of brittleness around aggregates during the leaching process, was demonstrated by Carde and François [17,22], which is in part due to the high Portlandite concentration within the ITZ.

2.1. Evolution of porosity

The main consequence of dissolution is a strong increase in material porosity [18,23], which amplifies the accessibility of external solutions to sound material and thus may amplify degradation phenomena. This porosity increase induced by leaching, reported in various works, is due to the loss of calcium: dissolution of Portlandite leads to a porosity growth corresponding to the initial volume of this constituent. Decalcification of C-S-H induces hydrate reorganization and modifies little their physical properties. Tognazzi [19] showed that the calcium decrease in the solid phase corresponds to the increase in total porosity of samples (cement paste and mortars with CPA-CEM I). This additional porosity is of comparable size to capillary porosity. C-S-H decalcification, which occurs after the Portlandite dissolution, only results in a small porosity modification compared to the variation caused by the Portlandite dissolution. Such results had also been observed by Revertegat et al. [20] or Gérard [21].