THE STUDY OF ALKALI-SILICA REACTION IN CEMENT BASED MATERIAL BY COMBINING SYNCHROTRON X-RAY MICROTOMOGRAPHY AND X-RAY DIFFRACTION TOMOGRAPHY

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Summary: In the present research the distribution of cement phases, aggregate phases and weathering products in mortar samples affected by Alkali-Silica Reaction (ASR), a weathering phenomenon affecting the performances of building materials was performed by combining synchrotron X-ray tomography and X-ray diffraction tomography.

1. INTRODUCTION

Recently, the investigations of cement based materials affected by Alkali-Silica Reaction (ASR), as a weathering phenomenon in building materials, has attracted much attention. The ASR consists of chemical reactions between silica aggregate affected by different degrees of structural disorder (*i.e.* opal, chert, etc) and the alkali and hydroxide ions in the pore water of concrete [1]. The ASR results in the precipitation of a gelatinous product (CKSH), nanometric in size, rich in silica, calcium-, sodium-, potassium-oxides and water that surrounds the aggregate. The expansion of CKSH, because of the osmotic pressure due to the imbibition of pore solution, promotes cracks and fractures through concretes [1]. The ASR mechanism is described by different theoretical models but the ASR growth within cement materials remains poorly understood. In particular, there has been debate as to which mechanism is dominant in the CKSH growth at the cement-aggregate interface. Hence, several authors assume that ASR products are gel whereas others report a "crystallised gel", but this is still an open debate.
Lots of studies have been performed to highlight concrete structures affected by ASR, using traditional analytical techniques such as optical and electron microscopy. We have performed the first synchrotron X-ray tomography study on cement materials weathered by ASR [2]. We have collected phase-contrast images, with a details detectability approaching that of optical microscopy (a few microns). The reconstructed volumes allow to characterize the main ASR weathering products (voids and cracks) within cement based materials [3,4].
Up to now, no studies have been performed to investigate what happens at the main mineralogical phases belonging to the aggregate and cement paste (i.e. quartz, hydratation products) in structures affected by ASR. In addition, it is still unknown the CKSH spatial distribution within cement as well as it is still matter of debate if CKSH products have an amorphous or crystalline structure.
In the present study several samples were prepared by mixing siliceous aggregate (previously checked as reactive towards ASR) with Portland cement; these specimens were demoulded in a solution of 1M NaOH to enhance the ASR products formation.
The aim of the present research is double-fold: on one side we would like to monitor the evolution of microstructure and contents of selected crystalline phases (i.e. quartz, cement phase) as the ASR proceeds and a map of their spatial distribution would be required. On the other side, we would map the evolution of the CKSH products in terms of their spatial distribution in ASR affected samples as well as we would characterise their microstructure with a non-invasive approach. Therefore X-ray diffraction tomography coupled with X-ray tomography seem to be the most viable, non-destructive methods for studying the cement based materials affected

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by ASR.

2. EXPERIMENTAL METHOD

Five cylindrical samples (with a maximum diameter of 2 mm) are prepared by mixing siliceous aggregate (previously checked as reactive towards ASR) with Portland cement. Note that the cement based samples are prepared by considering the fact that the crystals (in terms of grain size) in the samples should be an order of magnitude smaller than the X-ray beam size. The samples are aged for 1, 3, 7 and 14 days at 80°C in a solution 1M NaOH for promoting the ASR growth. The aged samples will be investigated by X-ray tomography and X-ray diffraction tomography using a synchrotron radiation. The experiments were performed on ID11 beamline of the European Synchrotron Radiation Facility (ESRF).

3. RESULTS

The rendering of the weathered samples allows to identify and quantify the main weathering products (voids, cracks, aggregate dissolution and ASR-gel growth) (Fig. 1a). From the XRD diffraction tomograms we obtain different information (Fig 1b): (i) as concerns the aggregate and cement paste, the difference of their microstructure as the ASR proceeds have been highlighted by analysing the line profile analysis of the peaks in the X-ray powder diffraction patterns. For instance, changes in crystallite size and microstrain of the aggregate crystals are present in the aggregate during the ASR-weathering. Indeed a map of the main phases spatial distribution and their content estimated by Rietveld analysis in the samples as the ASR attack proceeds have been performed, thus clarifying the ASR mechanism.
(ii) as regards the CKSH, new information about the new phases precipitated by ASR reaction are highlighted. These allow one to clarify the actual nature of the ASR reaction products.

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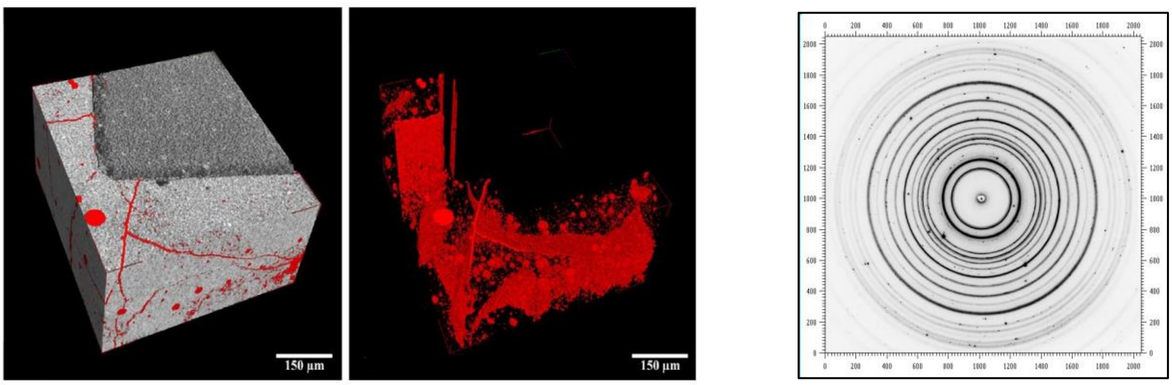

(a) (b)

Figure 1: Mortar bar affected by ASR: (a) rendering of the weathered samples with microcracks surrounding the aggregates (X-ray tomography); (b) diffraction image collected during X-ray diffraction tomography experiments.