Mineralogy and porosity of a 13 years *in situ* concrete/bentonite contact by image processing of quantitative elemental mapping and ^{14}C MethylMethAcrylate (MMA) impregnated samples

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INTRODUCTION

Deep geological repositories (DGR) are actually the available solution for safe long-term storage of high-level radioactive waste (HLRW). The waste would be isolated from the biosphere by a system of engineered and natural barriers. The engineered barrier system (EBS) usually consists of a metal sleeve surrounding the waste, forming a canister cylinder, followed by a compacted bentonite clay barrier. The host rock, in which access galleries are excavated, will need concrete vaults for supporting the walls (clay rocks) or concrete plugs (crystalline rocks) to seal and stop the swelling of the hydrated bentonite backfill inside gallery. DGR would gain confidence according to the detailed multi-scale analysis and characterization carried out after the dismantling of long-term (10-30 years) in-situ simulated experiments. The FEBEX project was one of these experiments implemented in the underground research laboratory (URL) located at the Grimsel test site in Switzerland. It was based on the Spanish disposal reference concept for disposal of HLRW in crystalline rock (Huertas et al., 2005). Two cylinder heaters were maintained at a constant temperature of 100ºC on their surface, facing an annulus of compacted bentonite that filled the gap up to the granitic wall. A concrete plug was applied to seal the drift end. The operational stage started in 1997. In 2002, after five years of operation, one of the heaters was switched off and dismantled, along with the surrounding bentonite. A concrete plug was made by a shotcrete technology to seal the remaining part of the experiment. The experiment was definitively dismantled in 2015. The 13 years (2002-2015) concrete-bentonite interface is the focus of this study. It is a small piece of a global and exhaustive characterization work (FEBEX-dp and part of CEBAMA UE projects) attempting to describe the microstructural and geochemical perturbation produced in the reaction of concrete and bentonite. The paper deals to show some microstructural and mineralogical aspects revealed using the methodologies described by Gaboreau et al. (2012, 2017) studying both clay and concrete materials.

MATERIALS

The FEBEX bentonite was extracted from the Cortijo de Archidona deposit (Almería, Spain). The montmorillonite content of the FEBEX bentonite is above 90 wt.% (92 ± 3 %) and contains variable quantities of quartz (2±1 wt.%), plagioclase (3 ± 1 wt.%), K-feldspar (traces), calcite (1 ± 0.5 wt.%), and cristobalite-trydimite (2 ± 1 wt.%) (Ramírez et al., 2002).

The composition of the shotcreted plug was: CEM II A-L 32.5R paste (430 kg/m^3), water (170 kg/m^3), nanosilica (30 kg/m^3), aggregates 0-8 mm 1700 kg/m^3, steel fibres (50 kg/m^3), polypropylene fibres (800 g/m^3) and superplastisizer, curing and accelerator compounds (< 10% in cement weight). The first layer of the shotcreting (up to 2cm thickness) rebounded, so additives were slightly re-adjusted (Bárcena el al, 2003). Bulk mineralogy consists of quartz (40-30 wt%), calcite (30-20 wt%), plagioclase (albite , 15-10 wt%), orthoclase (10-5 %), etringite (10-5 %) and portlandite, biotite, muscovite, and clinochlore (< 5%). The mineralogy is complex and it is not possible to ascertain the contents of anhydrous cement phases or calcium silicates hydrates C-S-H.

Several unaltered bentonite/concrete contacts were obtained by a special overcoring technique (Jenni et al., 2014). Small drillings (up to 3-4 m long) were practiced around the desired core section and filled with a polymer for induration. Then the core drilling was able to capture the undisturbed contact (Fig. 1).

METHODS OF ANALYSIS AND RESULTS

The samples were impregnated in ^{14}C MMA during more than 4 months in order to achieve fully access even to the smectite interlayer porosity. Then, quantitative porosity measurements can be done by autoradiography (Fig. 2). It came evident that there is a porosity increase in concrete affecting more than 2 cm thickness from the bentonite contact. This is presumably related to the initial quality of the shotcreting.

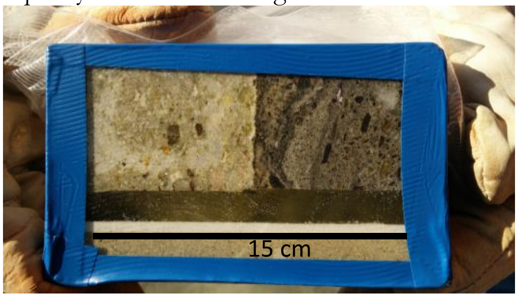

Fig 1*. Undisturbed 13 years bentonite (left)/concrete (right) contact.*

Quantitative X-ray intensity maps and BSE image were acquired with a Cameca SX *Five* EPMA equipped with five wavelength dispersive spectrometer (WDS) allowing totake 512x512 pixel elemental maps with a spatial resolution of 2µm per pixel. It is possible to compute mineral/phase maps based on procedures of chemical segmentation using ternary scatter plot projections.

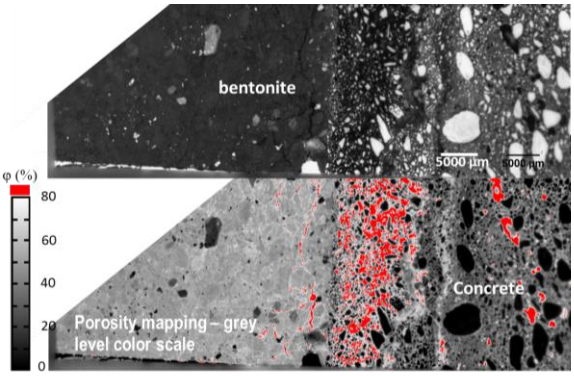

Fig 2*. Autoradiography of ^{14}C MMA impregnated concrete/bentonite contact. Porosity scale and measurements.*

In **Fig. 3** the pixels related to quartz (Si pole) have been separated from other anhydrous phases based on the sum of all their oxide components. If H or C are present, the sum of oxides cannot be 100 % for the corresponding pixel.

Actually we are processing complete mineralogical maps in order to detect the mineralogical evolution at microscale related to the geochemical perturbation of the complex mineralogy at the bentonite-concrete contact.

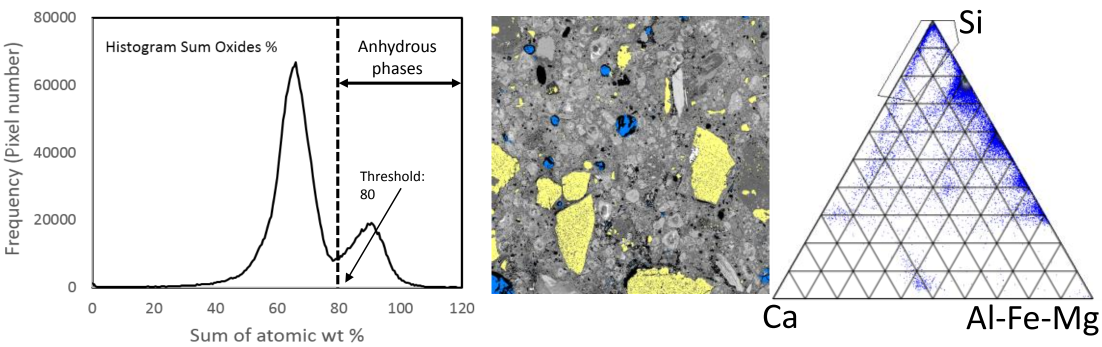

Fig 3*. Example of image segmentation process for anhydrous silica grains (quartz in yellow; (% oxide composition sums 100% if H or C is absent). Blue shades are etringite grains previously segmented using a S-Ca-Al diagram.*

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