Iron-bentonite interaction in a water-saturated low temperature environment: mineralogy and microstructure

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In the context of the NF-PRO project [1], an iron-FEBEX bentonite interaction experiment (FeMo) was initiated at CIEMAT in 2008 with the aim of evaluating iron mobility and bentonite alteration considering a conservative scenario: (i) saturated conditions, (ii) aerobic initial conditions, (iii) high corrosion rates (use of iron powder), and (iv) room temperature. FeMo was analyzed in the context of the European EURAD-ACED project after 15 years of operation (until 2023).

X-ray microtomography was performed on one of the six sections (referred to as section 3) into which the cell FeMo was divided during dismantling for analysis (Fig.1). The images are shown in Figure 2. Image A is a slightly rotated 3D reconstruction of the block. Image B is part of the apical view (cross section of the apex in Fig.1) and images C and D are different slices of the cross section. A brown colored halo was spread over the surface where hydration was maintained with a set of syringes containing distilled water. From the observation of stacks B through D, it can be inferred that the bentonite is apparently saturated, as there was no visible grain boundary, typical of unsaturated compacted samples. The iron-bentonite interface is sharp and there is no clear penetration of iron into the bentonite.

The environment of the cylindrical section containing steel inside and compacted iron powder in contact with the compacted bentonite is characterized by a black stripe (iron powder) followed by a brown zone of variable thickness (2-5 mm), which stains the bentonite, yellowish green (Fig 1). The black strip was analyzed by XRD preserved from exposure to atmospheric oxygen. The mineralogical composition of the iron powder shows a predominance of magnetite (Fe3O4 = 50%), accompanied by goethite (FeOOH = 25%) and siderite (FeCO3 = 5%), with metallic iron remaining in a proportion of around 20% by weight. The brown-colored bentonite contact contains bentonite minerals (montmorillonite, cristobalite, feldspar and quartz), magnetite, and the oxyhydroxides goethite and lepidocrocite, with minor siderite. The bentonite near this contact has the same composition as the original mineralogy. SEM-EDX elemental maps (Fig. 1) at iron powder-bentonite interface showed preferential zones of Fe, Mg and Ca concentration. The diffusion of the iron halo reaches up to 1 mm in the bentonite. This halo is segmented by a Mg concentration zone between 0.1 and 0.3 mm and a Ca concentration zone between 0.2 and 0.3 mm measured from the interface with the iron powder. There is a 0.1 mm band in the bentonite at the iron/bentonite contact where Ca and Fe are preferentially concentrated. These anomalies are very restricted to the 0.5 mm zone closest to the interface, except for iron, which penetrates up to 2-3 mm into the bentonite (presumably siderite).

The study of internal sections of one of the iron powder cylinders of the FeMo experiment shows that, after 15 years of interaction, a diffusion-stained zone less than 1 mm thick has developed in a practically preserved radial symmetry. The localized deposits (µm) of Mg- and Ca-rich phases suggests the existence of an alkaline contact produced by iron oxidation, the spatial extension of which is very limited (< 1mm). This experiment is unique in its temporal dimension, as there are no similar experiments of such long duration. The possibility of studying future experiments of longer duration (i.e. FEBEX mock-up > 25 years), with the same bentonite and including the effect of temperature, should be very positive.

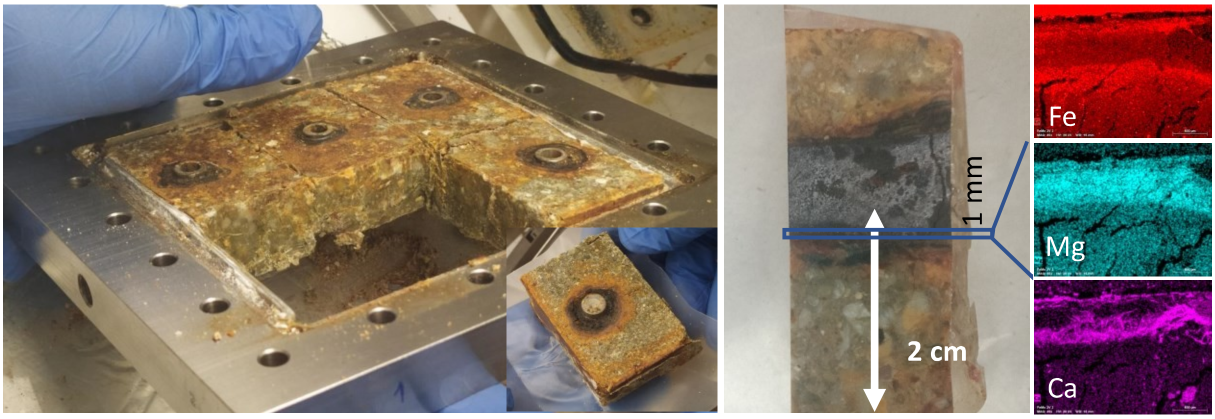

Figure 1: On the left is the dismantled FeMo experiment and a detail of section 3 showing the black-brown halo on the surface. On the right is a cross section of section 3 and the SEM-EDX maps showing the Fe, Mg and Ca chemical gradients at the iron powder-bentonite interface.

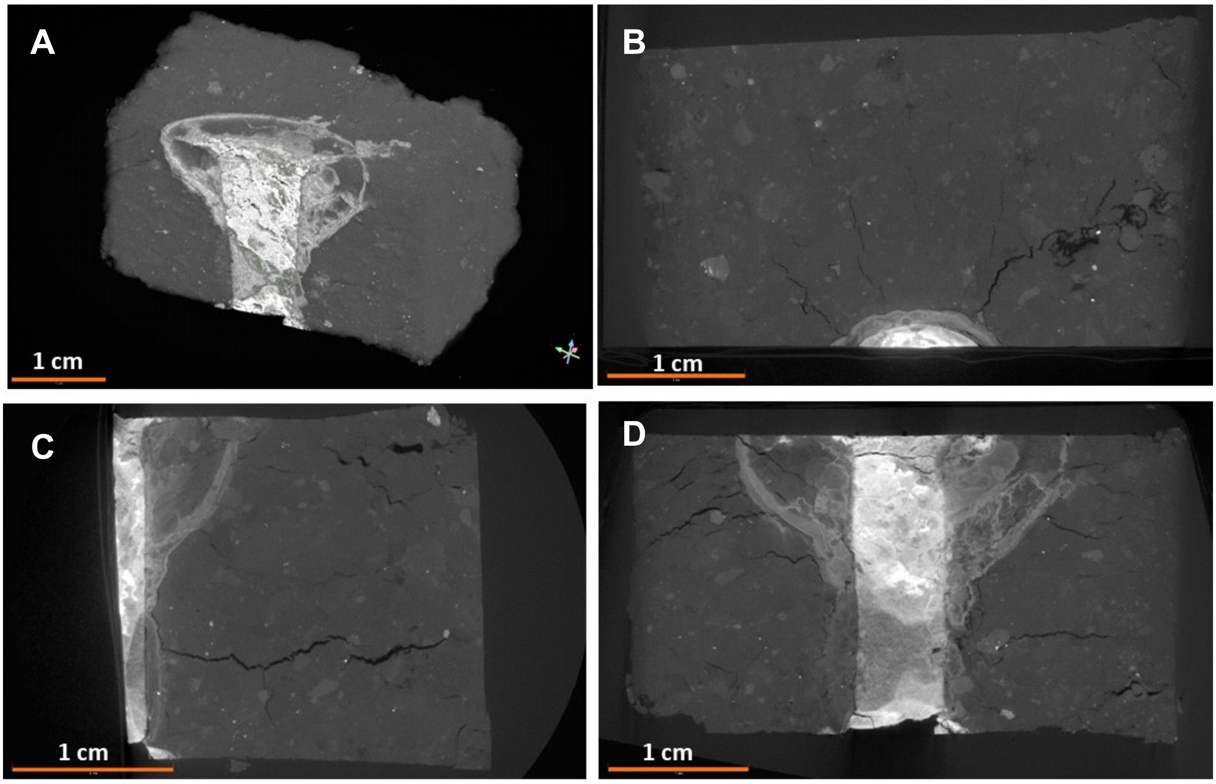

Figure 2: Various images of section 3 of the FeMo experiment (see Figure 1). A. 3D X-ray microtomography reconstruction. B. Stack of an apical view. C and D. Stacks showing a cross section.

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[1] A. Sneyers (coordinator) (2008). Understanding and Physical and Numerical Modelling of the Key Processes in the Near Field and their Coupling for Different Host Rocks and Repository Strategies (NF-PRO). EUROPEAN COMMISSION nuclear science and technology series. EUR 23730. 217p.