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Electrochemical corrosion monitoring of iron in mixture cement paste – bentonite

Julia Agullo¹, Christian Bataillon¹, Nicolas Michau²

¹CEA, DEN, DANS, DPC, SCCME, LECA, F-91191 Gif sur Yvette Cedex, France

²ANDRA - DRD/CM -Parc de la Croix Blanche, 1/7 rue Jean Monnet, F-92298 Châtenay-Malabry Cedex, France

ABSTRACT

In the framework of French radioactive waste repository, Andra is developing a 500-metre deep disposal concept for the high-level waste (HLW). It plans to inject a grout containing cement-bentonite mixture between the liner steel and the wall of HLW cell drilled in the Callovo-Oxfordian layer. This mixture has two purposes: neutralization of the acidity due to oxidation of the sulfurs in the claystone after drilling operation and limitation of oxygen diffusion into the claystone along the wall of cells during the exploitation phase. The pH in the genuine claystone at room temperature is about 7. The present of cement implies that the pH of the close-environment of the liner will be alkaline ($10 < \text{pH} < 13$). So at the beginning low corrosion rate of the liner would be expected. Then the pH would decrease and the corrosion rate should increase only slightly due to steel passivation at alkaline pH at shorter time.

The scope of this work is to check that the processes in the grout could be electrochemically monitoring. Electrochemical Impedance Spectroscopy (EIS) was carried out to follow the hydration of grout and the iron corrosion together. A methodology based on complex impedance analysis was developed to extract the different parameters at high and low frequencies of the spectra. At initial time the cement-bentonite mixture is pasty and becomes solidified with time. After a 100 days cure at room temperature (21 °C) the temperature of the system (grout + steel) was increased at 80 °C to simulate the introduction of nuclear waste overpack. The evaluation of the ionic conductance shows a continuously decreased at room temperature followed by an increase at 80 °C (cf. Figure 1). After an initial decrease the corrosion rate has increased to quasi-steady state value around 1.5 $\mu\text{m}/\text{year}$ (cf. Figure 2). Then the corrosion rate has decreased to about 0.8 $\mu\text{m}/\text{year}$. The scanning voltammetry performed in parallel on gold electrode has shown that this decrease corresponded to the completely consumption of oxygen initially dissolved in the grout. The increase of the temperature to 80 °C has induced to an increase of the corrosion rate about 30 $\mu\text{m}/\text{year}$. Then the passivation of iron seemed to occur since the corrosion rate has decreased continuously down to 0.1 $\mu\text{m}/\text{year}$. For longer time the corrosion monitoring based on Stern-Gary method could not be applied.

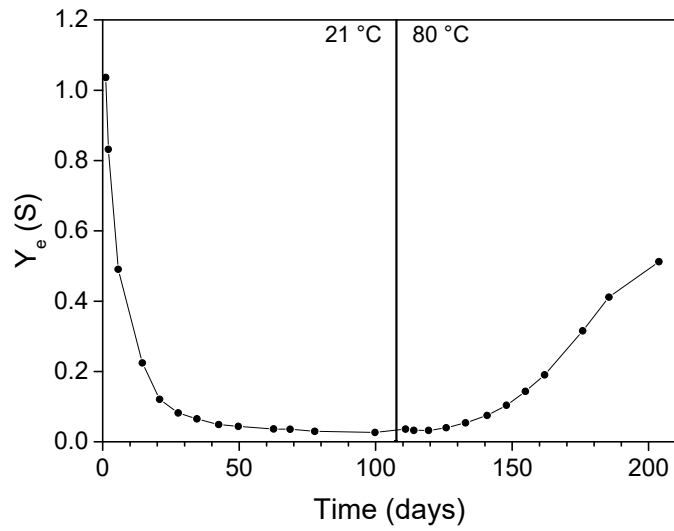


Figure 1: Time evolution of the ionic conductance Y_e of the cement-bentonite mixture.

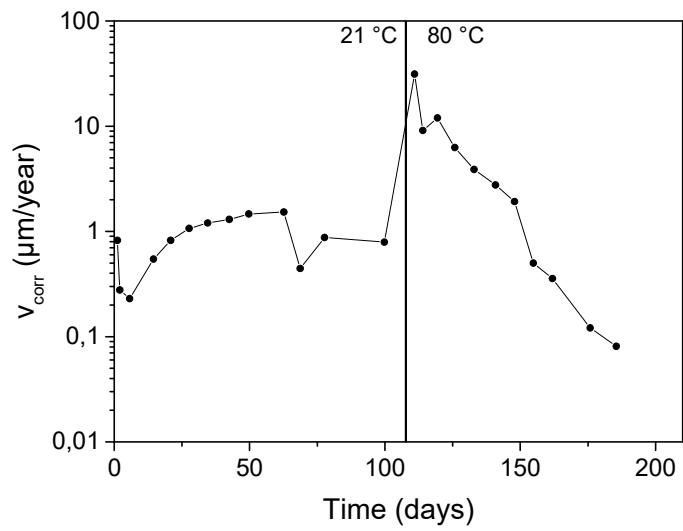


Figure 2: Time evolution of the iron corrosion rate in the cement-bentonite mixture.