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# **Influence of polysulfide radicals on the mechanisms and intensity of corrosion of a carbon steel API 5L X65 in contact with cement grout in future nuclear waste disposal program**

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Cigeo is the future French deep Nuclear Waste Disposal (NWD) project for long-lived highly radioactive wastes. The clayey Callovo-Oxfordian formation (COx) was chosen as natural barrier to isolate the waste from people and environment. The waste will be placed in API 5L X65 steel linings, separated from the COx by a cemento-bentonitic material (FS10B at the time), whose formulation may still evolve, and which will, among other things, keep the steel in a passive state thanks to its basic pH (>12). For safety reasons and reversibility of storage, continuous monitoring of the Corrosion Current Densities (CCD) or corrosion rate of the carbon steel in contact with this cementitious material is therefore necessary. That is the framework of this work.

A specific electrochemical methodology, consisting of i) stationary techniques (measurement of polarization resistance by Linear Polarization Resistance (LRP) and by Chrono-Amperometry (CA), Voltammetry around Open Circuit (VaOCP) by fitting the kinetic parameters), and ii) transitory techniques (measurement of impedance by Electrochemical Impedance Spectroscopy (EIS)), was carried out at regular intervals, for more than eight months, into a metric corrosion pilot, in which the steel was immersed into the cementitious FS10B material (water/solid ratio of 3). The system used a solid AgCl/Ag reference electrode that showed its robustness over the period and a Platinum counter-electrode. In complement, potentiometric measurements (pH, temperature, conductivity, redox potential, permittivity, dissolved O<sub>2</sub>), and continuous analyzes of the level of sulfides were simultaneously made. Particular attention was given to the visual analysis of the evolution of the grout color, which acquired a green tint in the first days of the experiment, due to the anoxic conditions prevailing into the hydrated FS10B material and the presence of polysulfide radical anions (S<sub>n</sub><sup>-</sup>) coming from the blast furnace slag used for CEM III/A cement production.

Indeed, the loss of the green tint was observed after five months of experiment. It suggested  $S_n^-$  oxidation in agreement with FS10B partial dehydration and oxygen diffusion into the cement material. In addition to EIS, this specific behavior helped to understand, in particular, the reduction reaction involved during the API 5L X65 steel corrosion process and to choose the best kinetic model to apply during the first five month of experiment and beyond.

During the first five months of immersion into the cementitious FS10B material, the API 5L X65 steel was in an active state ( $2\text{-}5 \mu\text{A}/\text{cm}^2$ ) in the presence of  $S_n^-$ , despite the formation of a layer of corrosion products highlighted by EIS (Fig. 1A), and the corrosion current density was limited by the diffusion of  $S_n^-$ . Beyond five months, in the absence of  $S_n^-$ , the steel become passive and the corrosion was limited by charge transfer of water molecules. These changes occurred in a few days and the corrosion current dropped down to about  $0.02\text{-}0.07 \mu\text{A}/\text{cm}^2$  (Fig. 1B).

Whatever the experimental conditions, the corrosion kinetics of API 5L X65 immersed in FS10B is dominated by the cathodic reduction reaction of water molecules. The latter is catalyzed by the presence of  $S_n^-$ . The use of the specific electrochemical methodology developed for corrosion sensing of API 5L X65 immersed in FS10B provides redundant information: it clearly identifies the changes at the API 5L X65 steel/FS10B interface as well as both the corrosion mechanisms and the kinetics.

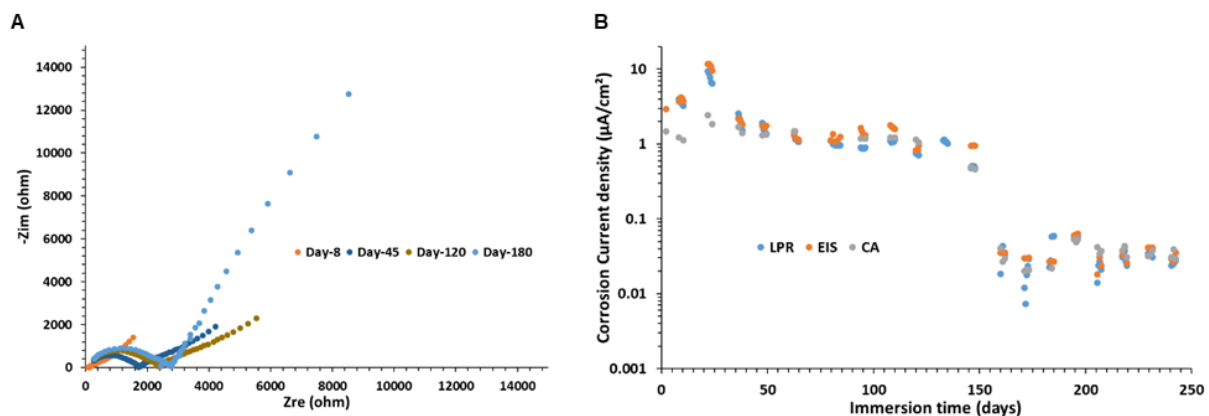


Figure 1. A) Evolution of impedance measurements in Nyquist mode derived from EIS measurements on API 5L X65 electrode immersed in a cement grout as a function of time. Evolution of API 5L X65-grout interface with time. B) Evolution of corrosion current densities (CCD) of an API 5L X65 carbon steel immersed in a cement grout as a function of time. LPR: CCD determined by Linear Polarization Resistance, EIS: CCD determined by EIS, CA: CCD determined by Chrono-Amperometry.