Corrosion of Carbon Steel in Physically-Constrained Locations in HLNW Isolation

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Belgium proposes to dispose of high level nuclear waste (HLNW) by encapsulating the waste in "supercontainers", which would then be stored permanently (for 100,000 to 1,000,000 years) in clay deposits. It is assumed that the principal threat is the corrosion of the carbon steel overpack in contact with Portland cement concrete, which is situated between cylindrical overpack and stainless steel liner. A comprehensive, mixed potential model (MPM) that was developed for the corrosion of carbon steel in simulated porous environment (pH = 12-13.5) in the presence of radiation showed that within a few months, the annulus is predicted to become anoxic, particularly within the regions close to the metal surfaces, and that hydrogen evolution takes over as the principal cathodic reaction. Due to a redistribution of hydrogen within the annulus the corrosion potentials of the overpack and the liner are predicted to shift in the negative direction but ultimately to shift slightly in the positive direction, due to the falling temperature. The corrosion potential is found to be controlled primarily by the hydrogen equilibrium reaction, as described by the Nernst equation. The corresponding Point Defect Model for predicting rate of passive corrosion under these conditions has been developed and employed in the MPM.

Calculations show that, because the corrosion product (rust) occupies 2-6.5 times the volume of the iron from which it forms (the Pilling-Bedworth ratio, PBR), significant compression of the passive film, particularly of the precipitated outer layer, occurs, as the film on the overpack impacts the concrete within the annulus. In particular, the critical thickness, d_{cr}, of the corroded layer (loss of metal) that is required to induce a sufficiently large tensile stress in the concrete to induce cracking drops as the PBR of the corrosion products increases (see Figure 1). On the other hand, the compression of the outer layer (rust) leads to a corresponding decrease of its porosity. If the porosity decreases to a sufficiently low value the growth of the outer layer ceases and further corrosion is prevented (see Figure 2).

Moreover, it is possible that there insufficient water in the annulus, order to support corrosion via hydrogen evolution reaction (see Figure 3). For example, for a porosity of concrete of 10 %, and for saturation of water in the concrete pores of 50 %, it is predicted that the overpack (of thickness 3 cm) will not fail by general corrosion.

However, modeling shows that, in the presence of aggressive anions, such a chloride ion and at high redox potentials, passivity breakdown can occur and, accordingly, stable pits will develop upon the surface and grow into the overpack resulting in the accumulation of damage. Currently, we are generalizing the model, in order to assess the risk that localized corrosion, in the form of pitting, poses to the integrity of the overpack.
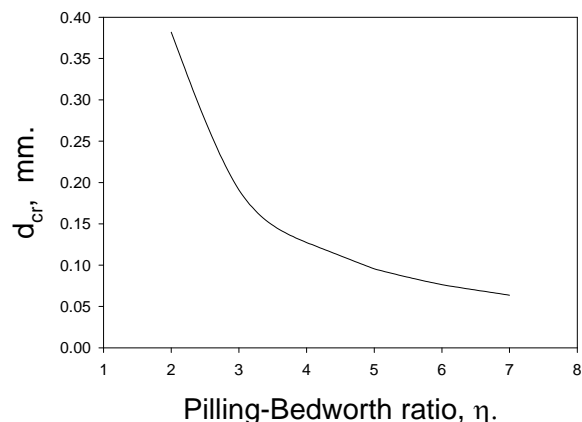
Figure 1. Critical thickness of corroded layer as a function of Pilling-Bedworth ratio.
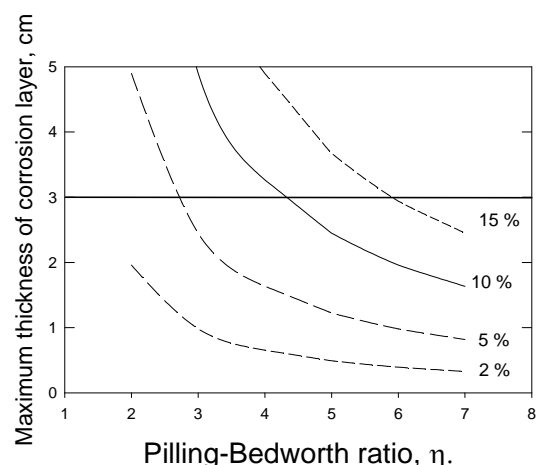

Figure 2. Maximum thickness of corrosion layer as a function of Pilling-Bedworth ratio for different values of initial porosity of the rust layer. Thickness of the overpack is 3 cm.

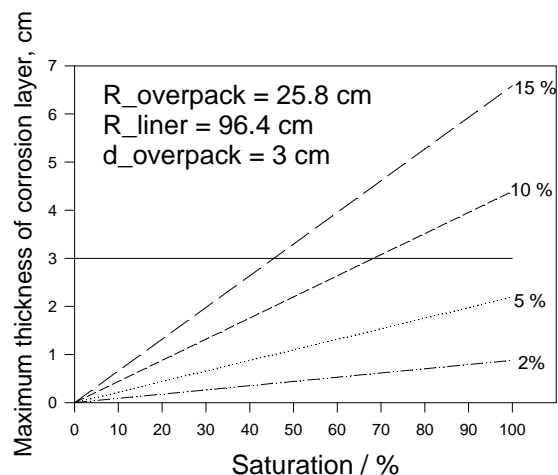**Figure 3.** Maximum thickness of corrosion layer as a function of saturation of water in concrete pores for different values of porosity of concrete.