Redox reactivity of actinides in the context of high level waste geological disposal: the role of hydrogen and iron

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We aim at describing the geochemical behavior (speciation and solubility) of actinides (U, Pu, Np) susceptible to be released by high-level waste (HLW) in the near field of deep geological repository. This behavior largely depends on the physicochemical conditions imposed by the environment which will evolve over time from an initial state perturbed by atmospheric conditions back to the reduced conditions prevailing in the deep geological environment. Moreover, hydrogen and iron will be produced in large amounts as a result of steel components corrosion. However, hydrogen is not considered to readily react with other species. We therefore investigated the effect of the reactivity of redox species (H_2, FeII/FeIII) and minerals on the redox state of radionuclides released in the HLW cell and in the claystone.

Batch simulations have been performed using thermodynamical and kinetics approaches with the geochemical codes Hytec and Crunchflow. The simulations were performed in different configurations:

- with magnetite and goethite (environment inherited from an oxidative phase), and, alternatively, with magnetite and siderite (natural conditions in deep geological environments),
- with a source of FeII and H_2 (simulating the active corrosion phase) in which actinides may react either with H_2 or FeII,
- with a source of FeIII and actinides in the oxidized state (VI) simulating the alteration of glass in the absence of H_2, i.e. post-corrosion phase where hydrogen has diffused away from the cell.

We discuss the results of the simulations in particular with regard to the effect on actinides.

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