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Influence of iron corrosion on nuclear glass alteration processes: nanoscale investigations of the iron-containing phases

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Several strategies propose to store and confine High Level Radioactive Waste in a deep geological disposal. Andra (French National Radioactive Waste Management Agency) suggests a multi barrier system including the glass canister, a carbon steel overpack and a low permeability clay host rock to prevent borosilicate glass alteration and to limit migration of radionuclides released under the action of water. However after thousand years and resaturation of clay, water will corrode the carbon steel overpack causing the release iron ions and precipitation of iron carbonates as corrosion products. The glass matrix will alter through glass hydrolysis and release silicon in solution. Thus, neoformed Fe-Si-O phases can precipitate in the glass alteration layer (GAL) or at the outer part of the GAL and lower the concentration of Si in solution, increasing glass dissolution. Consequently identification and characterization of nanocrystallized Fe-Si-O phases is crucial for modelling the mechanism of glass alteration in contact with iron.

The results presented here are obtained on samples (mix of nuclear glass and iron powder) altered in the underground Laboratory of Bure (France). Micro and Nanoscale investigations (Transmission Electron Microscopy, Scanning Transmission X-Ray Microscopy, nanoAuger electron spectroscopy) show presence of neoformed nanocrystallized phases (iron silicates) inside GAL and in the iron corrosion products (ICP). Several families of structured Si-Fe-O phases are identified (e.g. smectite in ICP, chlorite and iron sulfide in GAL) according to the localization and the valence of iron in CP. Moreover study of the provenance of silicon and iron found in phyllosilicates was carried out in mass spectroscopy (TOF-SIMS). Thereby it is possible to know the proportion of silicon and iron arising from the glass, initially substituted for ²⁹-silicium and ⁵-iron, to form these silicates.

REFERENCE: Dillmann P. *et al*, (2016) Effect of natural and synthetic iron corrosion products on silicate glass alteration processes. *Geochimica et Cosmochimica Acta* 172, 287-305.