Impact of heat, gamma- and alpha-radiation on the clay barrier in a geological disposal facility for radioactive waste

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There is an urgent need to implement engineered geological disposal for the safe isolation and containment of heat generating radioactive wastes. Clay is an integral component of all geological disposal facilities (GDFs) proposed worldwide, both in the engineered barrier and in potential host rocks. Due to its low hydraulic conductivity, high swelling properties, high ion exchange and sorption capacity for radionuclides, bentonite, consisting primarily of the dioctahedral smectite, montmorillonite, has been proposed as the engineered clay barrier (ECB). Montmorillonites have variations in chemical composition and structural site occupancy, affecting layer charge characteristics, which impact on the permeability and sorption properties of the ECB. Even after decades of interim storage, disposal of heat generating waste will lead to temperatures of 80-90°C in the ECB. The ECB will also be exposed to ionizing γ-radiation. Over time, the primary containment will corrode releasing corrosion products (Fe_3O_4, $Fe(OH)_2$, Fe(II), Cu(I)) which will interact with the ECB, and the ECB will additionally be exposed to α-irradiation from the longer lived radionuclides (actinides and fission products) in the waste. Given the complexity of this multi-component system and the potential for strongly coupled effects, the aim of this work is to provide definitive information on crucial knowledge gaps in the GDF safety case. Specifically, microfocus X-ray diffraction (μ-XRD), X-ray absorption spectroscopy (XAS), infra-red (IR) spectroscopy and electron paramagnetic resonance (EPR) is being used to determine how damage caused by temperature, γ- and α-radiation affects the ECB, including changes to the crystal structure resulting in defect formation and amorphisation; and subsequent impacts of these changes on sorption properties, as a function of clay composition. This research will ultimately yield a predictive understanding of the ECB safety functions of radionuclide containment and isolation within the GDF near field.