Examining radiation damage effects in phyllosilicates with applications to radioactive waste geodisposal

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The globally favoured solution for the safe disposal of radioactive waste is permanent isolation in a deep geological disposal facility (GDF). Phyllosilicates, such as mica and clays, will play a critical role within a GDF as highly reactive, physio-chemical barriers intended to limit radionuclide release. Investigating the mechanisms by which irradiation from α-emitting radionuclides causes structural and chemical changes across these mineral phases is an important and necessary consideration for any GDF safety case.

Preliminary radiation damage studies have focused on biotite mica, chlorite and montmorillonite, serving as model systems for the clay-based bentonite backfill material surrounding a canister, as well as reactive clay and mica phases in the host rock.

Using the newly commissioned (2013) 5MV tandem pelletron at the Dalton Cumbrian Facility (DCF), mineral samples have been bombarded with accelerated $^{4}He^{2+}$ ions (alpha particles) over a range of doses. Following irradiation, samples have been analysed using microfocus X-ray diffraction (μ-XRD) and X-ray absorption spectroscopy (XAS) on beamlines I18 and B18 at Diamond Light Source. Further analysis of damaged samples has been undertaken using Infra-Red (IR) spectroscopy, electronprobe microanalysis and high resolution transmission electron microscopy to quantify structural and chemical changes.

Across the damaged phases, μ-XRD has demonstrated radiation-induced interlayer collapse. Increased mosaicism occurs over a micrometer scale. XAS coupled with IR analysis has shown radiation induced reduction of Fe^{3+} to Fe^{2+}, hypothesized to result from increased electron density following OH^- group radiolysis.

Initial X-ray microprobe mapping experiments have also demonstrated that these radiation-induced changes do have an effect on sorption properties, with $Se(IV)O_3^{2-}$ preferentially sorbing to irradiated biotite.