'Geochemical' research: Key building block for radwaste disposal safety cases in clayrocks

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Disposal of high-level radioactive waste in deep underground repositories has been chosen as the solution by several countries. Because of the special status that this type waste has in the public mind, national implementation programs typically mobilize massive R&D efforts, last decades and are subject to extremely detailed and critical social-political scrutiny. The culminating argument of each program is a 'Safety Case' for a specific disposal concept containing, among other elements, the results of performance assessment (PA) simulations whose object is to model the release of radionuclides (Rn) to the biosphere. Public and political confidence in PA results (which generally show that Rn release will always be at acceptable levels) is based on their confidence in the quality of the scientific understanding in the processes included in the PA model, in particular those governing radionuclide speciation and mass transport in the geological host formation. Geochemistry (obviously) constitutes a core area of research in this regard.

Clay-mineral rich formations are the targets of radwaste programs in several countries (France, Belgium, Switzerland…), principally because of their demonstrated capacities to adsorb many radionuclides and to limit (diffusion-dominated) mass transport. The corresponding key processes in PA models are radioelement chemistry (redox state, speciation, reactions determining Rn solid-solution partitioning) and diffusion-driven transport. These processes take place in solution-filled pore spaces, most of which are bounded by the surfaces (basal, interlayer, edge) of permanently charged clay minerals. In addition, natural rocks always contain other solid phases potentially capable of influencing radionuclide speciation (pyrite, organic matter…) as well as being physically and chemically heterogeneous at space scales ranging from sub-millimetric to hectometric.

In order to gain the detailed scientific understanding needed for constructing those parts of the Safety Case supporting how Rn transfer is represented in the PA model, a wide range of coordinated (and internationally collaborated) research has been carried out. The purpose of this presentation is to illustrate how research results in fields ranging from molecular dynamic calculations of ion distributions and mobility near clay surfaces, to clay surface-catalyzed transformations of the redox state of certain elements (Se…), to the effects of electrical double layers on the composition and ion transport properties of clayrocks, to measurement and model representation of micro-scale mineral-porosity relationships and the effects on diffusion, to use of geochemical modeling to estimate porewater composition, to use of high resolution detection methods to quantify diffusion at sub-millimeter scales…, have contributed to building convincing Radwaste Safety Cases.

Depleted uranium in the environment: A biogeochemical study

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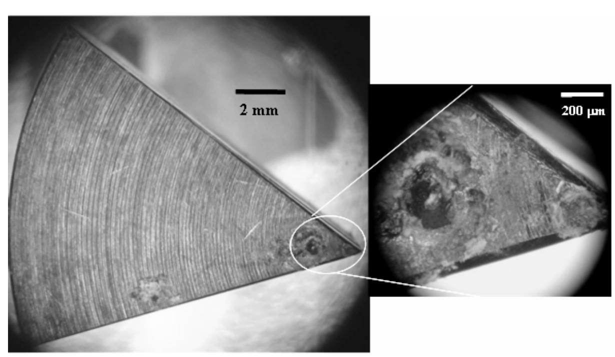
Geochemical and microbial processes affecting the decomposition and dispersion of military depleted uranium (DU)/titanium alloy penetrators are being investigated. Since the behaviour is unique to this form of uranium, laboratory model systems are being used to define the effects of the chemical and microbial processes involved in decomposition. The effects of DU on bacterial communities are of particular interest and are being studied using both culturing and molecular methods, together with quantification of dissolved, colloidal and macroparticulate fractions of uranium decomposition products.

DU coupons with a mass of ca 8 g have been exposed to aqueous media containing appropriate terminal electron acceptors (TEA: oxygen, nitrate, Fe(III) or sulphate), and indigenous bacteria, either those present in various selected soils, or an inoculum cultured from an estuarine sediment and a loamy clay soil. The concentrations of the added TEA groups were monitored over time. Microcosms were supplied with excess electron donor and sacrificed once TEA reduction was complete. Abiotic control samples were also prepared for comparison.

There was greater degradation of DU by cultured aerobes, including 'pitting' of the surface when compared to anaerobes. An example of the pitting is shown in the photograph below taken after only one day of incubation at 15 ^{o}C.



The extent of DU corrosion under different conditions can be related to changes in solution geochemistry and microbial community structure.