

Redox barriers in deep geological disposal sites

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The deep geological disposal concept is widely accepted for the storage of high activity level nuclear waste, using a multi-barrier system to isolate radioactive waste from the hydrosphere and biosphere. The multiple barriers include, from the waste to near- and far-field: metal (e.g., iron or copper) and/or concrete canisters/casks containing the radioactive waste, cement, clay (e.g., smectite) buffer and/or backfill materials, and naturally occurring host rocks (e.g., claystone and granite). All these phases can sorb redox-sensitive RNs and drive reductive immobilization processes, thus control the mobility of radionuclides (RN) [1]. The resulting reduced products are usually the most stable and least soluble phases, which can be identified by X-ray photoelectron spectroscopy (XPS) or X-ray absorption spectroscopy (XAS). The measure of the oxidized (soluble) species, combined to Nernst equation, has been used to compute the E_H potential within the concrete redox barrier [2]. As an example, we will discuss the long lived (0.6 million year) Se^{79} case, the reaction of $Se(IV)$ with a variety of $Fe(II)$ containing mineral and steel rust products, leading to little soluble Se^0 or $FeSe(s)$, and the consequent low E_H computed for cement pore waters. This E_H information combined with physical properties of the far field enhance our ability to predict rates of reactive diffusive transport processes for diverse dissolved and gaseous species in clay rock formations over periods up to thousands of years. [3].

[1] Ma et al. (2018) Applied Geochemistry: **100**, 414-431

[2] Ma et al. (2018) Environ. Sci. Technol., **52**, 1624–1632

[3] Charlet et al. (2017) Adv Water Res. **106**, 39-59