

Retention of selenite and selenate during the formation of magnetite

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The ^{79}Se isotope is a long-lived harmful radionuclide present in high-level nuclear waste (HLW). The expected predominant oxidation state of Se in vitrified HLW is Se^{4+} but oxidation processes induced by long-term irradiation could also lead to a transformation in Se^{6+} [1]. In an aqueous environment both Se^{4+} and Se^{6+} occur as oxyanions, selenite and selenate, which are well soluble and generally highly mobile due to the rather limited adsorption capacity of most geological materials, e.g. clays, for anionic species [2].

In this study, we investigate the retention of selenite and selenate by magnetite during its formation and growth in coprecipitation experiments, since magnetite is one of the most common corrosion products of the HLW steel canisters and interaction processes already during the mineral formation are very likely. In a first step a method for the synthesis of magnetite was developed to define the experimental setting and afterwards Se coprecipitation and comparative adsorption studies were carried out.

The ICP-MS results of the coprecipitation experiments showed that more than 99 % of both the selenite and selenate were removed from solution during the magnetite formation for all tested initial Se concentration in the range of 10^{-4} - $5 \cdot 10^{-3}$ mol/L. In the solid phase of the samples we detected the presence of elemental Se by using XRD and SEM. In the same way preliminary data of spectroscopic techniques, XPS and XAS, suggest that the magnetite formation lead to a reduction of selenite and selenate and thereby to the precipitation of elemental Se. In contrast to the Se coprecipitation, the adsorption studies showed that retention of selenite and selenate by pure adsorption is quite low under comparable hydrochemical conditions and there were no indications of a change of the Se oxidation state during the adsorption onto magnetite. Furthermore, a comparison of the stability of the retention indicated that coprecipitated Se^0 is more stable against a subsequent redissolution than adsorbed selenite and selenate species, even under oxidized conditions.

[1] Bingham *et al.* (2011) *J Non-Cryst Solids* **357**, 2726-2734

[2] Duc *et al.* (2003) *J Environ Radioactiv* **70**, 61-72