Selenium fixation by adsorption and co-precipitation during the formation of hematite

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The long-lived and mobile radionuclide ⁷⁹Se is a fission product of ²³⁵U and ²³⁹Pu and contained in high-level nuclear waste (HLW). In the near field of an HLW repository, the interaction of dissolved Se(IV) or Se(VI) species with mineral phases is a main process for Se immobilization. In this study, the retention of Se by hematite was investigated, since iron oxides are known for their high adsorption capacity and occur in the host rock and barrier materials of HLW repositories as well as in corrosion products of steel canisters.

In contrast to the reversible adsorption of charged Se species on the hematite surface, incorporation processes may represent a possibility for a permanent Se immobilization. To investigate the Se fixation by hematite during the mineral formation, a published synthesis method for hematite [1] was adapted to reflect the expected hydrochemical conditions of an HLW disposal site. The synthesis took place in solution at temperatures of 50 °C and under neutral to slightly alkaline pH conditions.

The results of co-precipitation experiments show an uptake of more than 99 % for Se(IV) during the formation of hematite at initial concentrations of 10⁻⁵-10⁻³ mol/l, whereas in the case of Se(VI), the decrease of the initial Se content is only about 10-15 %. In order to find indications of a Se retention by incorporation processes, the results of co-precipitation experiments were compared with Se adsorption studies on hematite under the same hydrochemical conditions. The results indicate that for initial Se(IV) concentrations of more than 10-³ mol/l the uptake of Se(IV) by co-precipitation is significantly higher than by adsorption. In the same way, the data of desorption studies reveal that the amount of desorbed Se(IV) is much higher (~75-90 %) for hematite samples of a former Se(IV) adsorption than for samples of a former co-precipitation (~15-35 %). Therefore, Se co-precipitation seems to be the predominant immobilization process compared to adsorption.

[1] Schwertmann & Cornell (2000) *Iron Oxides in the Laboratory*. Wiley-VCH, Weinheim.