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Retention of selenium and cesium on crystalline rock: The effect of redox conditions and mineralogical composition of crystalline rock

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Cesium and selenium belong among long-lived fission products, being present in spent nuclear fuel (SNF) and high level waste (HLW) and contributes significantly to the risk associated with the storage of radioactive waste in deep geological repositories.

The presented work is focused on study of sorption behaviour of cesium and selenium species, selenite Se(IV) and selenate (VI), on two types of crystalline rock, granitic rock from Melechov Massif (Czech Republic) and diorite Äspö from the underground laboratory Äspö (Sweden). Used rock samples differed one from each other in mineralogical composition, sampling depth and redox conditions during the core drilling and sample preparation. The drill core samples from Melechov massif were obtained and completely prepared (sawed, crushed, sieved, washed) under aerobic conditions. On the other hand, drill core samples from Äspö underground laboratory were drilled and later prepared under anaerobic conditions in argon glove box.

The main aim of experiments was to compare sorption behaviour of cesium and selenium on different crystalline rock samples under aerobic and anaerobic conditions and determine the influence of redox conditions during sample preparation on their sorption properties. The other aim of experiments was to identify potential retention mechanisms of selenium on crystalline rocks and to observe correlation between sorbed selenium and iron present in rock samples.

The sorption behaviour of cesium and selenium was studied by static batch method and was described by values of sorption distribution coefficient K_d . The batch sorption experiments were conducted on two fractions of crushed rock material with different grain size and on intact samples (coupons) under aerobic and anaerobic conditions.

Results of batch sorption experiments demonstrated different sorption behaviour of studied elements. Selenate Se(VI) behaved as non-sorbing, conservative tracer without any retention on rock samples; however selenite Se(IV) and cesium showed weak sorption without significant influence of different grain size of rock samples or redox conditions (aerobic/anaerobic). No significant differences between Czech granite and Sweden Äspö diorite were observed concerning Se and Cs sorption.

The spectroscopic techniques of surface analysis were used for study of Se forms on the rock surface after the contact with selenite and selenate solution. The Electron Spectroscopy for Chemical Analysis (ESCA) and Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) were used. These methods can provide information about distribution of selenium on rock surface and information about oxidation state of sorbed selenium. The results of surface analysis of studied samples confirmed the initial assumption that retention mechanism of selenium species can be correlated to the presence of iron-containing minerals in the rock but the change of selenium oxidation state on samples surface was not observed.

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