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Sorption of Cs (I), Eu (III) and U(VI) onto rock samples from Nizhnekansky massive (Russia)

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The repository for high level wastes (HLW) and spent nuclear fuel (SNF) in Russia is planned to be constructed in the deep underground crystalline rock formations near the Krasnoyarsk. Three areas of the Nizhnekansky Granite Massive, namely "Kamenny", "Itatsky"and especially "Eniseysky", are supposed as the most perspective locations for the future HLW and SNF disposal site. Physical and chemical properties of rock minerals (especially sorption towards radionuclides) are essential for the modeling of their long-term behavior and radionuclides migration through the granite body of the repository and, thus, for the Safety Assessment. Previously core materials from the specified areas have been studied in terms of petrographic and mineralogical characterization; definition of filtration, elastic, petro-physical and strength properties; estimation of hydrothermal-metasomatic transformation of rocks. In this work sorption of Cs(I), Eu(III) and U(VI) onto granite samples from aforementioned areas is studied.

Batch sorption experiments were performed both in glove box with inert atmosphere (N2) and at open-air conditions. Sodium perchlorate was used as a background non-complexing electrolyte (0.01 M). Initial concentration of each cation was 1E-7 mol/L and isotopes (U-233, Cs-137, Eu-152) were used for liquid-scintillation or gamma counting. The solid to liquid ratio was kept at 1:4. Rock samples were crushed and sieved with 1-2 mm grain size. Additionally, four undisturbed rock slices were used to reveal the minerals responsible for cation sorption.

The spatial distribution of cesium, europium and uranium(VI) onto surface of the undisturbed rock slices was investigated by digital radiography using imaging plate technique. Comparison of radiography and optical images of samples allowed us to reveal that dark minerals presented in the investigated rock samples (biotite, chlorite) are mostly responsible for Cs and Eu sorption. According to the results of digital radiography about 70% of radionuclides were sorbed onto these minerals whereas biotite and chlorite content is about 20%, means that dark minerals ca. ten times more effectively sorb Cs and Eu than other minerals. The distribution of U among different rock minerals is much more homogeneous.

The results of time-dependence study of radionuclide sorption showed that steady-state conditions of cations sorption onto investigated rock samples are reached within 15-20 days both in inert and air atmosphere. Thus, metal concentrations in solutions in other sorption studies were analyzed in three weeks after beginning of experiment. It was shown that in case of cesium sorption has no clear dependence on the pH values typical for ion-exchange mechanism. Thus, observed increase on Cs uptake with increasing of biotite content in rock samples is consistent. The pH-dependence of sorbed uranium fraction has typical hump-shape: increase of sorption percentage with increasing pH values to 6, plateau (90-98 % of uranium sorbed), decrease of sorption percentage with increasing pH values from 8 due to U(VI) hydrolysis and/or carbonate complex formation.

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