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U(VI) retention by potential host rocks: Comparison of clay and crystalline rock

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The long-term disposal of high-level nuclear waste in deep geological formations is discussed worldwide as main strategy for nuclear waste management. This approach requires the use of a multiple barrier system consisting of engineered, geo-engineered, and geological barriers to prevent any release of radionuclides into the geo- and biosphere. Sorption of radionuclides on the host rock of a repository is one important process for retarding their migration. Potential host rocks for nuclear waste repositories that are investigated internationally are salt domes, clay rock, and crystalline rock.
In the present work, the retention behavior of clay and crystalline rock towards U(VI) is compared. For this, sorption of U(VI) onto Opalinus Clay from the Mont Terri rock laboratory (Switzerland) was studied in the presence of Opalinus Clay pore water (pH = 7.6; I = 0.36 M) [1]. This is compared to U(VI) sorption onto anoxic diorite from Äspö Hard Rock Laboratory (Sweden) that was studied in the presence of Äspö groundwater (pH = 7.8; I = 0.18 M) [2]. The impact of various parameters, such as solid-to-liquid ratio, initial U(VI) concentration, temperature and atmosphere, on U(VI) sorption was studied.
Distribution coefficients, Kd values, determined for the U(VI) sorption onto Opalinus Clay and diorite at 25 ℃, amount to 22.2 ± 0.4 L/kg [1] and 3.8 ± 0.6 L/kg [2], respectively. This shows that U(VI) sorption onto Opalinus Clay is stronger than onto diorite, which can be attributed to its larger surface area. TRLFS and ATR FT-IR spectroscopic measurements showed that the U(VI) speciation in Opalinus Clay pore water as well as in diorite groundwater is predominated by the weakly sorbing Ca2UO2(CO3)3(aq) complex. Reduction processes of U play only a subordinate role. The U(VI) sorption increases with increasing temperature.
U(VI) diffusion experiments with intact Opalinus Clay bore cores [3] also showed that Opalinus Clay has a good retardation potential for U(VI) since the molecular diffusion process through Opalinus Clay retards the migration of Ca2UO2(CO3)3(aq).
Generally, it can be concluded that U(VI) retention by clay rock is stronger than that by crystalline rock. This supports decisions to use clay rock not only as host rock but also as backfill material. In case of crystalline rock the natural retention capacity for U is insufficient and has to be strengthened by additional geo-technical and technical barriers that preserve their enclosing capabilities over very long time scales.

[1] Joseph, C., Schmeide, K., Sachs, S., Brendler, V., Geipel, G., Bernhard, G.: Sorption of uranium(VI) onto Opalinus Clay in the absence and presence of humic acid in Opalinus Clay pore water. Chem. Geology 284, 240-250 (2011).
[2] Schmeide, K., Gürtler, S., Müller, K., Steudtner, R., Bok, F., Joseph, C., Brendler, V.: Interaction of U(VI) with Äspö diorite: A batch and in situ ATR FT-IR sorption study. Appl. Geochem., under review (2014).
[3] Joseph, C., Van Loon, L.R., Jakob, A., Steudtner, R., Schmeide, K., Sachs, S., Bernhard, G.: Diffusion of U(VI) in Opalinus Clay: Influence of temperature and humic acid. Geochim. Cosmochim. Acta 109, 74-89 (2013).

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