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Partitioning and Speciation of Pu in the Sedimentary Rocks Aquifer from the Deep LNW Disposal

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The sedimentary rocks aquifers are used as the reservoir beds for the deep injection of liquid nuclear waste (LNW) in Russia. The reservoir beds are located at the depth of 290-370 m and are limited from above and from below by the impermeable clayey strata. LNW injections vary in composition including acidic technological wastes, which consist of sodium nitrate, acetic acid, corrosion products and radionuclides. Safety of the LNW disposal depends on the migration properties of radionuclides as well as on the ability of the bearing stratum to localize radionuclides within the nearest zone of the repository.

Sorption of Pu onto the components of the different types of sandy rocks of the LNW disposal under the hydrothermal (100-150°C) acidic (pH 1.8 - 2.5) conditions have been studied. It was established that the presence of corrosion products (Fe, Cr) in the LNW solutions causes formation of Fe(III)-Cr(III) oxides-hydroxides that stimulates sorption of Pu on the solid phase. In the presence of Fe, Cr, Mn, Ni in the LNW composition Pu was bound to the solid phase almost quantitatively while without corrosion products only 30-60% of Pu was sorbed onto the sands. Sequential extraction of Pu from sandy rocks has demonstrated the influence of corrosion products in the binding of Pu to the solid phase. In the presence of Fe, Cr, Mn, Ni about 70 % of Pu was fixed in the hardly-leachable phase (acid-soluble and residue fractions). The content of initial sandy rocks influenced the partitioning of Pu. Organic rich sands (with coaly minerals) contain higher fraction of Pu bound to organic phase while sands rich in Fe-containing dark minerals have larger Pu fraction bound to Fe-Mn oxides.

Mössbauer spectroscopy and XRD identified the Fe-Cr-containing phases as hematite and goethite. HR-TEM reveals the morphology of the Fe-Cr-containing colloids which have been responsible for Pu sorption under hydrothermal conditions in acid media (pH 1.8 - 2.5).

The Pourbaix diagrams for Pu in the presence and in the absence of hematite were calculated to describe the sorption equilibrium in the system. Equilibrium constants for Pu sorption reactions were calculated using linear free-energy relationship (LFER) that is based on the analogy of hydrolysis in solution and sorption onto surface hydroxyl groups of hematite. It was established using Pourbaix diagram that the mixture of Pu(3+), PuO₂(+) and some PuO₂(2+) could be expected in the absence of corrosion products under acid conditions of LNW deep injection. In the presence of hematite Pu in the sorbed form, mainly presents in tetravalent form as ≡FeOPu(3+).

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