

Water Rock Interaction [WRI 14]**Groundwater geochemistry near the storage sites of low-level radioactive waste: Implications for uranium migration**

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Abstract

This paper presents results of detailed sampling of groundwater and surface water near the storage sites of radioactive waste from the Electrochemical Plant ECP (Zelenogorsk, Krasnoyarsk region, Russia) and the Angarsk Electrolysis Chemical Complex AEC (Angarsk, Irkutsk region, Russia), both of which have produced enriched uranium since 1960s. The liquid (*LRW*) and solid (*SRW*) radioactive wastes belong to the category of low-level activity waste. The main result is that the uranium is below the recommended MPC for drinking waters in all types of groundwater around the sludge of ECP and AEC. But alkaline nitrate solutions have been penetrating and spreading into the aquifers under the *LRW* sludge pits. According to our calculations, redox conditions in the groundwater influenced by discharge are controlled by the couple $\text{NO}_3^-/\text{NO}_2^-$ that facilitates U(VI) migration. The groundwater under *SRW* repositories is distinguished by its low mineralization and neutral pH. Co-contaminants, such as Mo, V, and Zr may serve as markers of technogenous contamination in storage sites of the *LRW* sludge.

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1. Introduction

The Electrochemical Plant **ECP** (Zelenogorsk, Krasnoyarsk region, Russia) and the Angarsk Electrolysis Chemical Complex **AEC** (Angarsk, Irkutsk region, Russia) have produced enriched uranium since 1960s. The activities of these plants produce solid and liquid radioactive wastes; all of them belong to the category of low-level waste. Liquid radioactive wastes (*LRW*) are formed during neutralization of nitric acid slurry after the uranium extraction by hydrated lime elevating pH to above 8. Some nitric acid is taken up as calcium nitrate $\text{Ca}(\text{NO}_3)_2$, however NO_3^- residual content in the solution can reach several

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g/l. A significant part of the residual UO_2^{2+} precipitates as poorly soluble calcium urinates or double oxides of calcium and uranium. Uranium content should be less than 0.5 mg/l. The neutralized slurry is transported to the *LRW* long-term storage site, i.e. the slurry tanks. The insoluble minerals precipitate from the slurry, which then drains through the walls and floor of tanks to groundwater. Solid radioactive waste (*SRW*) is stored in monolithic concrete structures under a mound of soil. The *SRW* composition is very diverse – sublimate production waste of chemical plant (sludge after the hydrometallurgical treatment, ash from the incineration of combustible RW, sludge after the pyrometallurgical decontamination of ferrous metal), a significant amount of iron oxide. Average uranium content in the *SRW* is from 1 to 2%; uranium oxide is a predominant U form in the solid waste.

Previous studies (the first in 1960, and the second in 2007) have shown that the chemical composition of groundwater within the industrial site conforms to the maximum permissible concentrations (MPC) standards in Russia. The goal of this work is to reveal chemical changes, which could occur over the years; calculating a number of thermodynamic equilibriums to reveal the factors effecting the redistribution of radioactive elements (uranium first of all) from permanent repositories to the biosphere. For the first time focus was made on the behavior of associated trace elements leached from the host rocks under the action of nitrate technogenic drainage waters.

2. Study area and methods

The slurry tanks are in the immediate vicinity of the industrial zones. Geological cross-section of the **ECP** slurry pond is made up of Quaternary diluvia deposits represented by sandy loams and loams up to 15 m thick. Jurassic argillic clays underlie the diluvium. The water table at the headwaters of the Khan and Syrgil Rivers occurs at the depth of 9-14m. Total annual and seasonal rainfall affects the water table of the aquifer. Near the Syrgil River, the water table approaches the land surface, and groundwater discharges into the bed of Syrgil bog at a depth of 0.5 m. The shallow aquifer is unconfined and recharged by infiltrating precipitation. Anthropogenic components contribute much to groundwater recharge in the area near the slurry storage sites. By their chemistry, natural waters are hydrocarbonate calcium–magnesium with neutral pH (7.45-7.8) and salinity of 0.35 g/L. Geological cross-section of the **AEC** is made up of Jurassic strata, covered with Quaternary alluvial sands and sandy loams. The Jurassic stratum is represented by eroded and broken sandstones. The chemistry of water in the Quaternary deposits is hydrocarbonate with $\text{Ca} > \text{Mg} > \text{Na}$. Salinity ranges from 0.15 to 0.3 g/l. Depending on the topography, the shallow aquifer ranges from 0.5 to 7 m deep. Seepage of solutions from the *LRW* sludge tanks creates a hydrologic cone of water under the tanks, as documented by drill holes.

The area under study is characterized by acutely continental climate with short summer, long winter and temperature variations from -50 to +35° C. The average annual rainfall is near 450 mm. Ground-, river- swamp waters near the ECP and AEC storage facilities were collected during 2 different seasons: later autumn/winter and spring. This double sampling was performed to show possible seasonal differences in dissolved salts. The total dissolved solids (TDS), electrical conductivity and pH were measured at the site. Anion species in solution were determined by the ion chromatography. Metals were analyzed by the ICP-MS and ICP-AES.

Thermodynamic simulation at 25°C and 1 bar total pressure was performed by the “HCh” computer code and the UNITHERM database using a Gibbs free energy minimization algorithm [1]. The equilibrium in a heterophase, 22-component system H-O-C-Cl-N-S-Al-Si-Na-Ca-Mg-Mn-Fe-U-Co-Ni-Mo-Zr-Cu-Zn-Ba-Sr was modeled. The U(IV) and U(VI) species were incorporated into the model using the reference data [2].

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