

Part VIII

Experimental and Modeling Study of Sorption–Desorption Processes

Recent research findings strongly suggest that the subsurface radionuclide transport is often accompanied by nonideal (anomalous) phenomena caused by (a) adsorption hysteresis facilitated, in particular, by irreversible uptake of radionuclides by a specific group of minerals in the rock matrix or by secondary geochemical alteration of the minerals, (b) deterministic and stochastic heterogeneity of the geological strata resulting in preferential flow paths of the radioactive component, and (c) colloid-facilitated transport. In particular, nonideal radionuclide transport behavior was observed during investigations that have been conducted at several sites associated with near-surface and subsurface radioactive waste (RW) disposal and supervised by RosAtom, the State Nuclear Energy Corporation, Russian Federation.

Two such disposal sites are located within the Northwestern Center of Nuclear Energy (NWCNE), near St. Petersburg: (a) a solid RW disposal site (the so-called *Radon site*), which is used as the northwestern regional surface repository; and (b) an Engineered or Designed Underground Repository (*EUR site*) in the Vendian clay (a regional aquitard) for low- and intermediate-level RW storage and isolation. Two other sites are associated with deep-well injection repositories, operated by the Siberian Chemical Plant and the Siberian Mining-and-Chemical Plant, the *Tomsk-7 site* and *Krasnoyarsk-26 site*, located in Western Siberia. The latter site is associated with a surface reservoir, Lake Karachai, which has been used over a long period for the disposal of liquid RW by the Mayak Production Association (the *Lake Karachai site*), South Ural.

Radioactive wastes contain various long-lived decay products. The most dangerous radionuclides are fission products (such as Sr-90, Cs-137), activated corrosion products (such as Co-60), and actinides (such as Pu-239, Am-241). To evaluate radionuclide behavior in the subsurface environment, sorption and desorption kinetics and equilibrium were measured in batch, diffusion, and column (dynamic) experiments. Rock samples of both sedimentary (unconsolidated and consolidated) and crystalline/fractured types were selected from the radioactively contaminated site (see Table). Core samples of the sandy sediment (the Radon site) were also taken from boreholes to study the spatial variability of the adsorption and desorption constants based on variogram analysis.

Table Summary of the experimental setup: samples, solutions, temperature–pressure, and experimental conditions

RW disposal site	Type of rock	Radionuclides	Solutions	Temperature–pressure	Sample conditioning	Book's section
Radon	Sedimentary: weakly consolidated sandstone	^{90}Sr , ^{137}Cs , ^{60}Co	SGW ^a	Room	Conditioned	21.3–21.4
EUR	Sedimentary: clay	^{90}Sr , ^{36}Cl , ^{60}Co , ^{239}Pu	SGW, SrCl_2	Room	Conditioned	22.6–22.7
Tomsk-7	Sedimentary: clayey sand	^{90}Sr , ^{137}Cs	SGW, NaNO_3	Room, elevated T–P	Conditioned, unconditioned	23.1
Krasnoyarsk-26	Sedimentary: clayey sand	^{90}Sr , ^{137}Cs , ^{239}Pu , ^{241}Am	NaOH , $+\text{NaNO}_3$	Room, elevated T–P	Conditioned, unconditioned	23.2
Lake Karachai	Crystalline: tuffs, tuff-and-lava	^{90}Sr	SGW, NaNO_3	Room	Unconditioned	23.3

^a SGW synthesized groundwater

The purpose of this study is to quantitatively evaluate the adsorption-related reactions with respect to natural attenuation, sorption and desorption kinetics and equilibrium under different geochemical, temperature, and pressure conditions. Earlier developed analytical and numerical models, which take into account the microscopic heterogeneity of multimineral geosorbents and variations in the external chemical potential resulting from the rate-limited transformations of the mineral phase, will be applied (Part VII).