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Mitigating the environmental impact of high salt liquid radwaste concentrates incorporated into cement packages

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The use of sulfuric acid instead of nitric acid for regeneration of cation-exchange resins arising from NPP operations produces sulfates which can form crystalline hydrates. Based on this capability, cement instead of bitumen can be used as a matrix for binding liquid radwaste (LRW), and high salt concentrates can be incorporated into Portland cement. Experiments simulating inundation of waste storage containers were performed. The experiments used a model salt solution representing an average composition of LRW from the VVER plants and containing 45% Na₂SO₄ of the total mineral content. Test solutions with a salt concentration from 200 g/l to 800 g/l and temperature of 80 °C were immobilized into cement matrix with water-to-cement ratio of 0.60 and addition of Cambrian clay as a sorbing agent. The percentage of added clay was about 10% of the total cement weight. The produced cement packages included from 6.7 to 25.1 wt. % of salts. Radionuclides were released from the cement surfaces into water with a rate of 10⁻¹ g/cm²×day for the first 24 hours of the experiment and after 90 days the rate of radiocesium leaching by diffusion was below 0.5×10⁻³ g/cm²×day meeting Russia's regulatory requirements (1×10⁻³ g/cm²×day) for standard concrete storage facilities. An increase in the test solution salt concentration from 200 g/l to 800 g/l resulted in an increase of the diffusion coefficient (De) from 0.5×10⁻⁵ to 2.8×10⁻⁵ cm²/day. However, this value is much below the regulatory requirements (≤ 8.6×10⁻⁴ cm²/day) on concrete disposal containers for radioactive waste. Since calcium sulfates are higher soluble than calcium aluminosilicate cement matrix, the maximum release of Cs-137 (2.2-3.0 % of the original activity), the most mobile radionuclide, from 200 L cement packages is mainly determined by surface leaching and occurs during the first three months of storage. This period is considered as the time of inundation scenario. The maximum diffusion leaching of Cs-137 taking its half-life into account will be observed during the first storage 20 years and the estimated release will be 10 times lower (below 0.2% of the original value) than that by surface leaching even for cemented concentrates with a salt concentration of 800 g/l. After 300 storage years the predicted radioactivity release will be maximum 0.004% of the original activity. Therefore, waste concentrates produced from LRW with a significant percentage of sulfate salts can be immobilized into Portland cement forms modified with sorbent clay which securely fix radionuclides even in case of high salt concentrates (up to 800 g/l).

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