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Immobilization of Iodine and Uranium by Phosphate Minerals

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To study immobilization mechanisms for uranium and iodine (^{129}I, a mobile fission product), we investigate the uptake of these nuclides by phosphate minerals. Phosphate crystallization experiments were conducted at the temperatures similar to those expected at waste repository sites, where the thermal peak of the waste package is ~250°C [1]. Crystallization of apatite occurred through the transformation of brushite in the iodine or uranium-bearing aqueous solutions. Crystals and fluids were characterized with various spectroscopy and mass spectrometry techniques for evaluating the degree of uranium and iodine uptake by phosphates and the mode of their incorporation into mineral structures.

Uranium results showed: 1) Apatite can incorporate 0.35 wt.% of uranyl from the hydrothermal solution in the presence of solid uranium-VI compounds; 2) Uranyl substitutes Ca in apatite structure and U-O1 bond distance (2.32 Å) is close to those of Ca-O1 (2.40 Å) [2]; 3) Experimentally determined values of uranyl incorporation in apatite overlap with the values predicted by Lattice Strain Model [3]. Our data suggest that crystallization of 1.4-2.7 g of apatite is required for complete immobilization of 0.65-3.6 mg of uranium dissolved in 1 kg of hydrothermal aqueous solution at 150-250°C, assuming the presence of solid uranium-VI compounds (Fig. 1).

Iodine results showed: 1) Apatite can incorporate a few wt.% of iodine; 2) Iodate content in apatite, crystallized in hydrothermal solution, is higher than iodate concentration in solution by 2-3 orders of magnitude; 3) A degree of iodate uptake increases with decreasing of aqueous iodate concentration. Our data suggest: a) Iodate is more compatible in apatite compared to iodide; b) Iodate substitutes phosphate in apatite structure; c) Crystallization of 4 g of apatite is sufficient for immobilizing 30 mg of iodate dissolved in 1 kg of hydrothermal aqueous solution.

References:

[1] Greenberg and Wen . (2013) LLNL-TR639869-DRAFT, 38.

[2] Jiménez-Arroyo, Gabitov, et al. (2023), *Chemical Geology* 634, 121581.

[3] Brice (1975). *J. Cryst. Growth*. 28, 249-253.

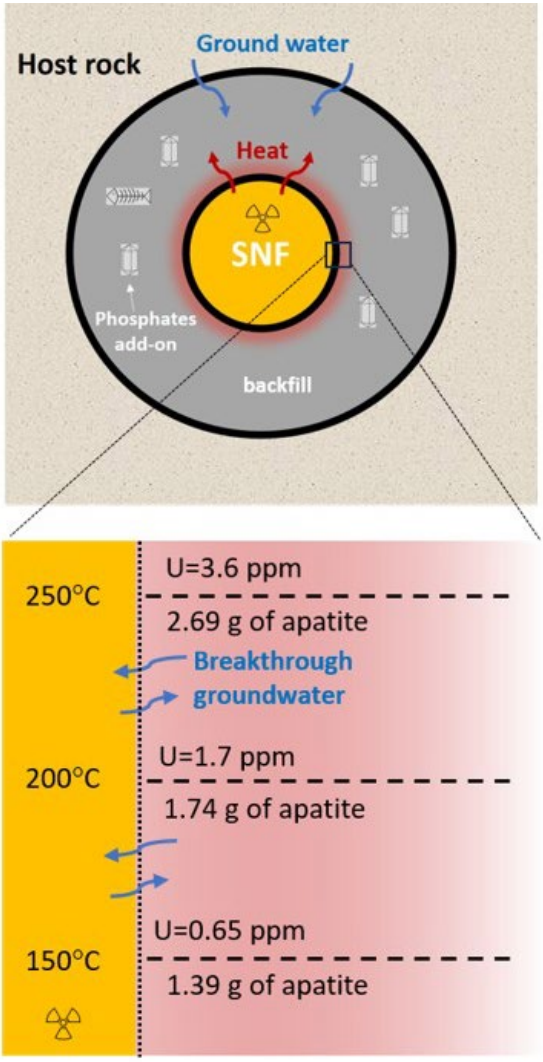

Figure 1. A Hypothetical scenario of uranium immobilization. Amount of apatite required for 100% removal of dissolved U-VI from 1 kg of solution for three waste package surface temperatures.