PREDICTIONS OF ACTINIDE SOLUBILITIES UNDER NEAR-FIELD CONDITIONS EXPECTED IN THE WIPP

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The order of importance from the standpoint of long-term performance assessment (PA) of the radioelements in the transuranic (TRU) waste to be emplaced in the U.S. Department of Energy's (DOE's) Waste Isolation Pilot Plant (WIPP) is Pu ≅ Am >> U > Th. However, the DOE has also included Np and other actinides in the WIPP Actinide Source Term Program (ASTP).

Anoxic corrosion of Fe- and Al-base metals and microbial consumption of cellulosic, plastic, and rubber materials will create strongly reducing conditions in the WIPP after closure. Under these conditions, Pu, Am, U, Th, and Np will speciate essentially entirely as Pu(III) or Pu(IV), Am(III), U(IV) or U(VI), Th(IV), and Np(IV) or Np(V). The use of MgO as an engineered barrier will result in low f_{CO_2} and mildly basic pH.

The WIPP ASTP developed thermodynamic speciation-and-solubility models for +III, +IV, and +V actinides in brines. Experimental data for Nd, Am, and Cm were used to parameterize the Pitzer activity-coefficient model for +III species; data for Th were used for the +IV model; and data for Np(V) were used for the +V model. These models include the effects of the organic ligands in TRU waste. The oxidation-state analogy was then used to extend the +III model to Pu(III), and the +IV model to Pu(IV), U(IV), and Np(IV). The solubility of U(VI) was estimated.

For the recent WIPP Compliance Recertification Application, we calculated +III, +IV, and +V actinide solubilities with f_{CO_2} buffered by the brucite-hydromagnesite ($Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$) carbonation reaction (vectors with microbial activity) or the brucite-calcite carbonation reaction (vectors without microbial activity), and with pH buffered by the brucite dissolution reaction. Calculated +III solubilities vary from 1.69×10^{-7} to 3.07×10^{-7} M, depending on brine type and whether or not microbial activity occurs; those for the +IV actinides vary from 5.84×10^{-9} to 2.47×10^{-8} M; the +V solubilities vary from 9.72×10^{-7} to 2.13×10^{-5} M. The estimated +VI solubilities are 8.7×10^{-6} or 8.8×10^{-6} M.

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