Assessing Technetium Immobilization in Cementitious Materials

J.C. SEAMAN[1], S.P. SIMNER[2], H.S. CHANG[3], AND F. COUTELOT[4]

[1]Savannah River Ecology Lab, The University of Georgia, Aiken, SC, USA, seaman@srel.uga.edu
[2]Savannah River Remediation, Steven.Simner@srs.gov
[3]North American Höganäs, Hyunshik.Chang@nah.com
[4]Savannah River Ecology Lab, coutelot@uga.edu

Technetium (Tc), a nuclear fission product with no stable isotopes, represents much of the long-term radioactivity associated with nuclear waste. Under oxidizing conditions, Tc persist in the environment as an anion in the +7 oxidation state, i.e., pertechnetate (TcO_4^-). In soil partitioning studies, the oxidized species displays limited retention generally associated with amphoteric soil oxides. However, Tc(VII) is subject to chemical reduction to the +4 oxidation state, which is less soluble and mobile in the environment. One disposition strategy for immobilizing ^{99}Tc containing waste is the production of cementitious materials that contain reducing agents, such as blast furnace slag (BFS), to enhance the retention of redox sensitive contaminants. At the Saltstone Disposal Facility (SDF) on the Department of Energy's Savannah River Site (SRS), low-level radioactive saltwaste is mixed with BFS, fly ash and portland cement to produce a solidified grout material known as Saltstone, which is deposited in a series of vaults for long-term disposal.

In the current study, contaminant mass tranfer rates for ^{99}Tc and other contaminants from saltstone simulants spiked with ^{99}Tc and rhenium (Re), often used as surrogate for ^{99}Tc, and actual SDF saltstone samples were assessed using EPA Method 1315, a recently adopted method for evaluating contaminant leaching from intact monolithic materials. Results from Method 1315 were compared to a novel Dynamic Leaching Method (DLM) in which a flexible-wall permeameter was used to achieve saturated leaching under an elevated hydraulic gradient in an effort to evaluate the persistence of reductive capacity and subsequent changes in contaminant partitioning within intact cementitious monoliths. Under similar conditions, Re leaching rates (and other poorly sorbing contaminants like NO_3^- and iodine) were much higher than ^{99}Tc, which was attributed to poor initial reduction or more rapid oxidation of Re, indicating that Re is not a suitable chemical analog for evaluating Tc partitioning under reducing conditions. In DLM tests ^{99}Tc leaching rates appear to be controlled by the solubility of a reduced Tc (i.e., Tc(IV)) solid phase.