^{99}Tc Sequestration Using Nickel-Doped Iron Spinel

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Technetium-99 (Tc) incorporation within stable spinel phases is a method being considered for Tc removal and immobilization from waste streams during the radioactive waste disposal management. In this study, transformation of Ni-doped $Fe(OH)_2$(s) to spinel minerals, such as trevorite, is explored as a method for Tc sequestration.

The Ni-doped $Fe(OH)_2$(s) transformation to Fe spinel as well as the simultanoues reduction of Tc(VII) to Tc(IV) and incorporation into the produced spinel was explored using batch experiments at low temperatures under near neutral (pH 8) and alkaline conditions (pH 13.3). The spinel products were characterized by XRD and XANES for mineral phases and Tc speciation, while the Tc and Ni distribution in the solid products was examed by SEM/FIB-TEM-EDS. The Tc removal from solution was calculated based on the measured Tc concentrations in both aqueous and solid phases (acid digestion).

The XRD results showed that iron spinel is the dominant mineral product. The Ni molar content (x) in spinel ($Ni_xFe_{3-x}O_4$) was observed up to 0.49 at pH 8, whereas very limited Ni was incorporated at pH 13.3, indicating a nearly pure magnetite phase. The SEM/FIB-TEM-EDS analysis on the pH 13.3-sample illustrated that the Tc in the final solid was distributed into three domains: the octahedral-structured spinel particles; Tc enriched spinel nanocrystals; and especially, Tc-rich or likely metallic Tc. Instances of metallic Tc regions were found in spheroidal, Ni-rich and metallic nanoparticles in a core/shell microstructure, which suggests strong reduction and sequentital precipitation of Ni-Tc-Ni. The Tc mass balance analysis showed that nearly 100% Tc was removed from the solutions and resistant to release during water and weak acid washing. The results indicate that Tc sequestration using Ni-doped iron spinel provides a treatment method for Tc removal from waste streams, and could be combined into further disposal approaches.