^{99}Tc Immobilization by Redox/Coprecipitation with Nickel-doped Iron Spinel from Off-gas Waste Stream at Hanford Site

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Generation of an aqueous off-gas waste stream with a high content of ^{99}Tc (~2.5×10^{-5} M) is expected through condensation and scrubbing of the melter off-gas during the vitrification operations in the Hanford Waste Treatment and Immobilization Plant (WTP) at Hanford Site (WA, USA). Instead of the proposed off-gas recycling back into melters, which will prolong operations and increase risk of equipment failure, an alternative treatment technology involving ^{99}Tc removal from off-gas and immobilization as separate waste forms is being investigated.

Through well-controlled laboratory batch experiments at varied geochemical conditions (pH, temperature, etc.), we explored transformation of Ni-doped $Fe(OH)_2(s)$ to spinel minerals as a redox/coprecipitation method for ^{99}Tc removal from off-gas simulants. The ^{99}Tc removal was calculated by mass balance based on the measured ^{99}Tc concentrations in both aqueous and solid phases, and solid characterizations (XRD, SEM/FIB-TEM-EDS, and XANES) were conducted on the final spinel products.

The results showed that the Ni-doped $Fe(OH)_2(s)$ - spinel mineral transformation process can simultaneously reduce $^{99}Tc(VII)$ to $^{99}Tc(IV)$ and incorporate reduced $^{99}Tc(IV)$ into spinel minerals. Solid phase characterizations confirmed the formation of iron spinel, and suggested incorporation of reduced $^{99}Tc(IV)$ in the spinel products. The mothod is effective for the WTP off-gas which contains both redox-sensitive contaminants Cr(VI) and $^{99}Tc(VII)$. The results indicate that nearly 100% Cr and over 80% ^{99}Tc can be simultaneously removed by adding $Fe(OH)_2(s)$ to off-gas simulants with a solid to solution ratio of 5 g/L under near neutral and alkaline conditions. The ^{99}Tc removal approach developd herein provides an alternative treatment method to eliminate the proposed recycle process of the off-gas waste stream, which ultimately can reduce WTP mission cost and operation time.