Incorporation of Radioactive Contaminants into Pyroaurite-Like Phases by Electrochemical Synthesis

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Abstract: During electrochemical remediation of radionuclide, ^{235}U, ^{238}U, and ^{99}Tc-contaminated aqueous solutions, pyroaurite-like phases, ideally $[M(II)M(III)(OH)_{16}CO_3 \cdot 4H_2O]$ where M = Fe, were synthesized following coprecipitation with iron from metal iron electrodes. The effect of radionuclides on the transformation of amorphous precipitates to crystalline pyroaurite-like phases was investigated using X-ray diffraction (XRD), scanning electron microscopy with energy dispersive X-ray analysis, Fourier-transform infrared (FTIR) spectroscopy, and fluorescence spectroscopy. The synthetic iron carbonate hydroxide phases showed primary XRD peaks at 0.7 and 0.35 nm and FTIR spectra that indicated the presence of a brucite-like sheet structure with carbonate anions occupying the interlayer. Divalent and trivalent iron, eroded from the electrode, occupies the octahedral sites of the brucite-like sheets. The carbonate anions in the interlayer balance the excess positive charge from isomorphous substitution of the Fe^{2+} or Fe^{3+} by reduced uranium (U^{4+}) and technetium (Tc^{4+}). Because of the lower solubility associated with crystalline phases than amorphous phases, incorporation of radioactive contaminants into pyroaurite-like phases by electrochemical syntheses represents a more effective approach for removing U and Tc from contaminated aqueous solutions than traditional technologies.

Key Words: Coprecipitation • Groundwater Remediation • Pyroaurite • Technetium • Uranium • Zero-Valent Iron

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