Reactivity of Co-occurring Nano-particulate U(IV) and U(VI) phases in Abandoned Mine Wastes Under Oxidizing Conditions

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We investigated the reactivity of co-occurring nanoparticulate U(IV) and U(VI) phases in abandoned mine wastes collected from the Jackpile mine, Laguna Pueblo, NM, under oxidizing conditions by integrating laboratory experiments with various spectroscopy, microscopy and diffraction techniques. X-ray fluorescence analysis revealed U concentrations as high as 7000 mg/kg in the mine waste. X-ray photoelectron spectroscopy (XPS) of these mine waste samples suggested the co-occurrence of both U(IV) and U(VI). The co-occurring U-minerals were encapsulated by carbon based electron microprobe mapping data. Data from electron microprobe and X-ray diffraction also suggest the presence of coffinite (U(IV)SiO$_4$) and U-phosphates (U(VI)-P-K), similar to observations made in XPS. Furthermore, the scanning transmission electron microscopy (STEM) analysis on these nano-particulate U minerals suggested that the U-P-K phase was crystalline and the predominant U phase in the mine waste. Batch experiments were carried out in oxidizing conditions to assess the reactivity of these co-occurring U-bearing mineral phases using 18 MΩ water (DI), sodium bicarbonate (NaHCO$_3$), and sodium hypochlorite (NaOCl, bleach) over 2 weeks. DI exposure invoked U concentrations of ~ 300 µg/L, an order of magnitude greater than the U MCL (30 µg/L). Reagents consisting of sodium hypochlorite and sodium bicarbonate yielded U concentrations peaking at ~8000 µg/L, and 10 mM bicarbonate yielded the highest U concentrations of 24000 µg/L. These results suggest that the bulk of U release is attributed to desorption of U as a result of complexation with carbonate (CO$_3^{2-}$) to form uranyl-carbonates (U exposure to CO$_3^{2-}$ ions) and dissolution.

Keywords: Uranium, Nanoparticulates, Spectroscopy, coffinites, U-phosphates