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Studies on the stability of studtite in the presence of ligands commonly found in nuclear waste

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Nuclear waste in the United States has historically been stored in large underground storage tanks, which have degraded and leaked radionuclides into the surrounding environment. Studtite, a uranyl peroxide mineral, is known to form in nuclear waste as a result of the reaction between uranium and hydrogen peroxide, which is produced from the alpha radiolysis of water. This uranium phase is typically preferable over other U(VI) species due to its insolubility. Nuclear waste is also known to include organic ligands such as EDTA, HEDTA and NTA, which are known form metal-ligand complexes. The impact of these ligands on studtite stability is critical to understand the mobility of uranium post-leakage. With this in mind, this study aimed to determine the impact of EDTA, HEDTA and NTA on the stability of studtite using batch dissolution studies.

Results from this study show that at pH 7 and 9, studtite is stable in water and releases negligible uranium into the aqueous phase. However, when 15 mM EDTA, HEDTA or NTA are present, aqueous uranium concentrations are increased. At pH 3 and 11, EDTA had no quantifiable impact on studtite stability, but HEDTA and NTA increased aqueous uranium concentrations. NMR results confirmed the presence of U-ligand complex with EDTA at pH 7. The solid studtite was characterized with SEM and XRD before and after dissolution studies. Further batch dissolution studies were conducted in the presence of bicarbonate (a known uranium complexant), in a synthetic nuclear waste sludge, and with increased ionic strength of the solution. Sorption studies at pH 3 showed no significant variation in sorption of free or complexed uranium to quartz, bentonite, silica, or calcite. All in all, these results show that the ligands EDTA, HEDTA and NTA in nuclear waste can play a key role in uranium solubility and studtite stability.