Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

TcO_2 oxidative dissolution by birnessite under anaerobic conditions: a solid-solid redox reaction impacting the environmental mobility of Tc-99

Jordan Stanberry, Ilana Szlamkowicz, Lauren R. Purdy Vasileios Anagnostopoulos*

Environmental Radiochemistry Group, Department of Chemistry, University of Central Florida, 4353 Scorpius Str, Orlando FL, 32816

*corresponding author, Vasileios.Anagnos@ucf.edu

Supplementary Information

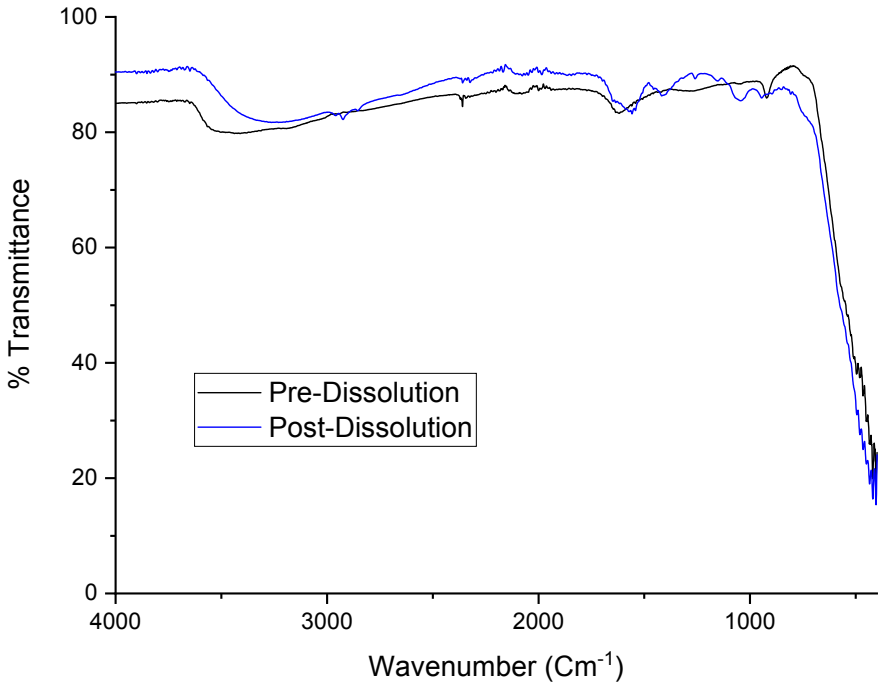

Figure 1: IR of Pre- and Post-Dissolution birnessite

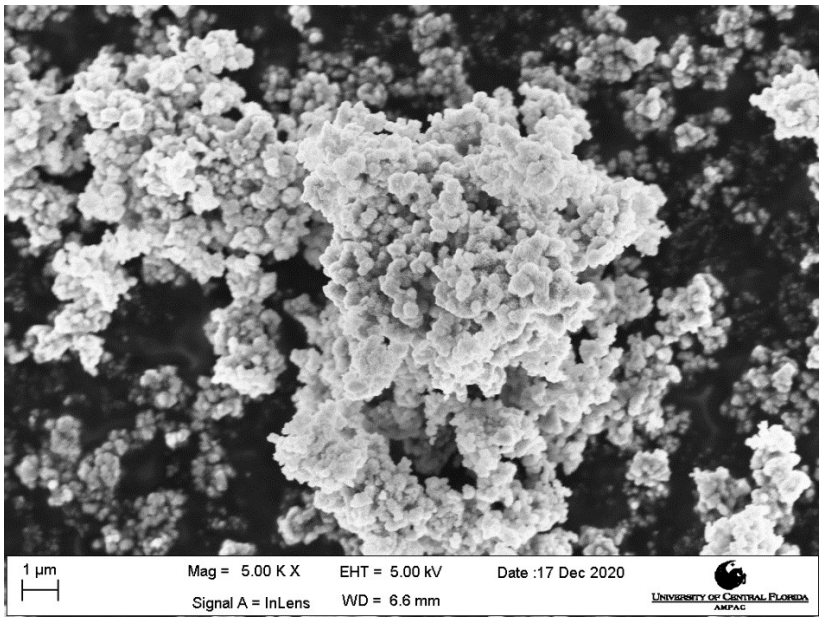

Figure 2a: SEM of pre-dissolution Birnessite (magnification = 5000x)

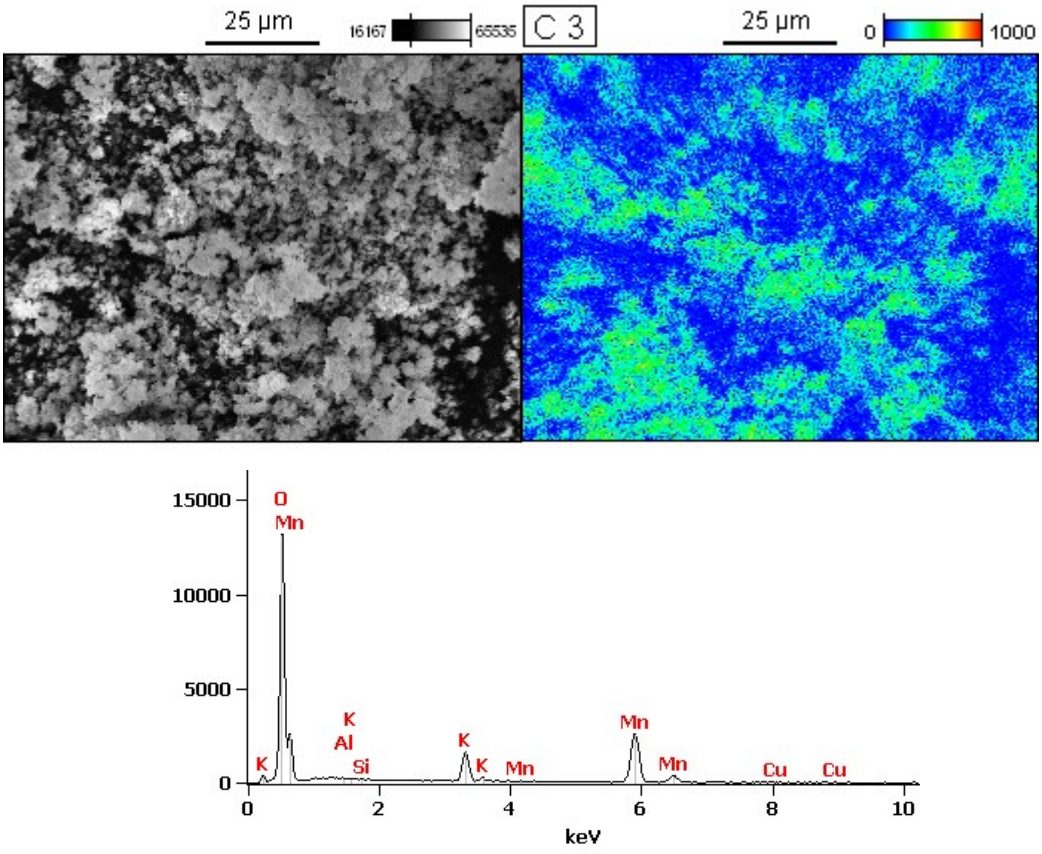

Figure 2b: SEM-EDS and Principal Component Mapping of pre-dissolution Birnessite (magnification = 1000x).

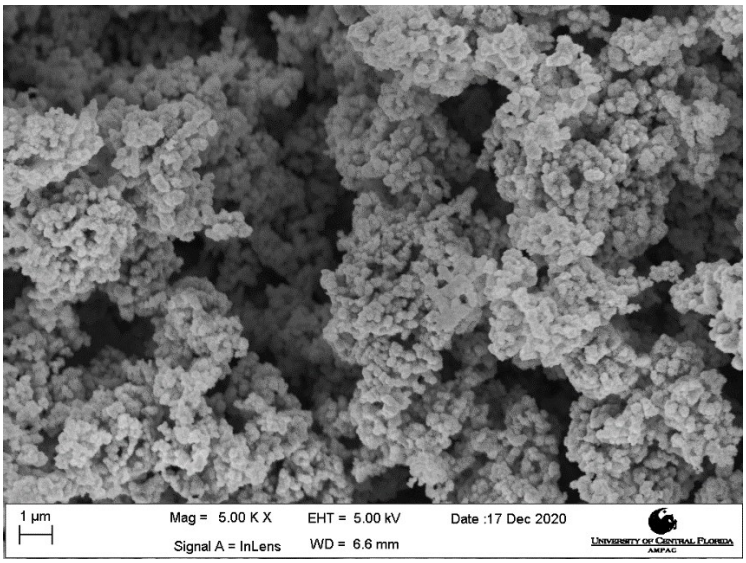

Figure 3: SEM of post-dissolution birnessite (magnification = 5000x)

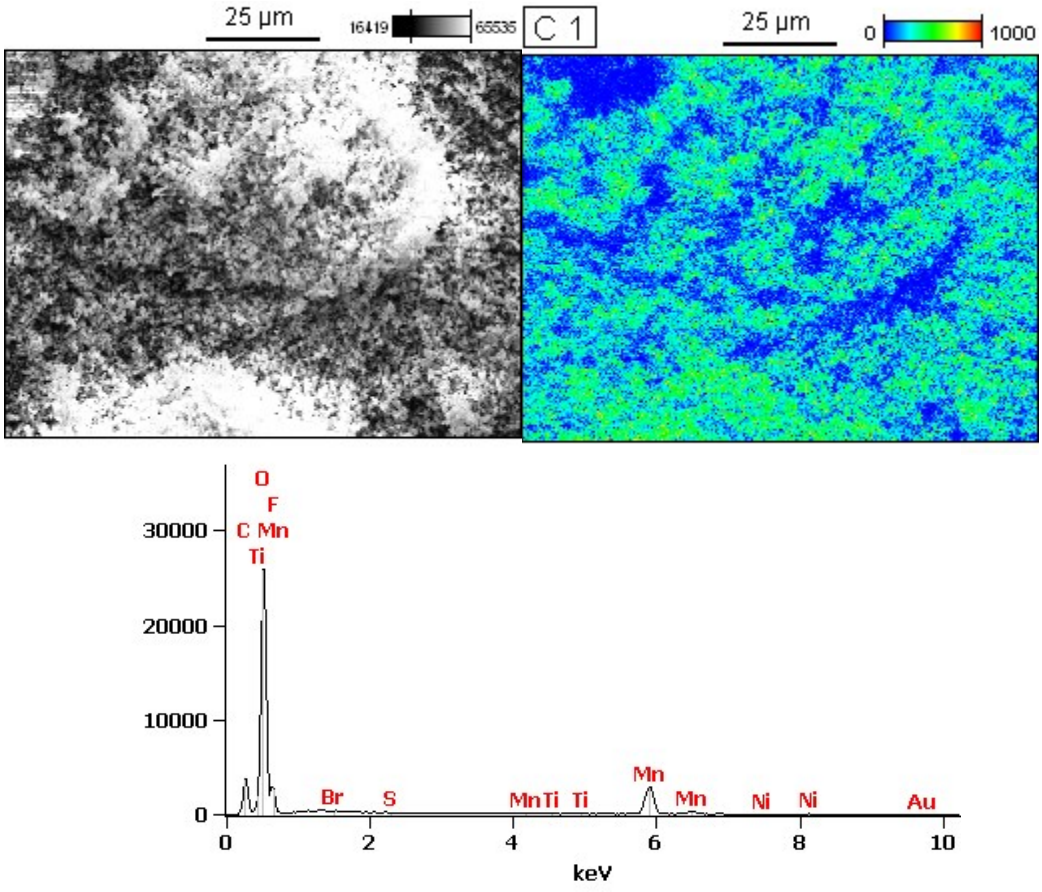

Figure 4: SEM-EDS and Principal Component Mapping of $MnCO_3$.

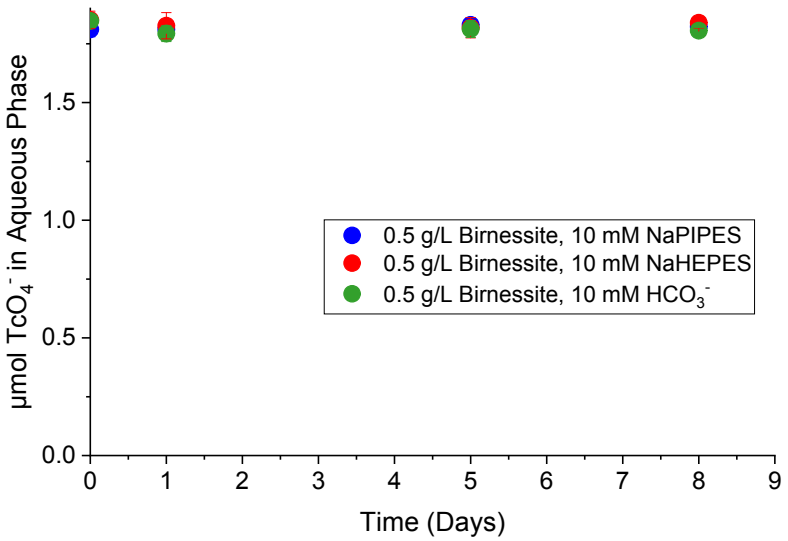

Figure 5: Sorption of TcO_4^- on birnessite. The blue set indicates samples buffered to pH 6.5 by 10 mM NaPIPES. The red set indicates samples buffered to pH 8.8 by 10 mM NaHEPES and the green set indicates samples buffered to pH 8.0 by 10 mM HCO_3^-. Error bars represent standard deviation of duplicate samples.

The sorption experiment depicted was performed in the anaerobic glovebox under the same conditions as the rest of the experiments. samples were spiked with 1.81 µmol TcO_4^- (the molar equivalent of Tc-99 in 0.25 mg TcO_2) and the concentration of TcO_4^- in the aqueous phase was monitored over time. The TcO_4^- concentration was measured by LSC at Time = 0 and then birnessite was added. Each sample contained 0.5 g/L birnessite and was buffered to pH 6.5 by 10 mM NaPIPES, or 8.0 by 10 mM NaHEPES or 10 mM $NaHCO_3$. The sorption of TcO_4^- on birnessite was determined to be 0.00±0.00%, 1.27±1.85%, and 0.02±0.01% for samples buffered by NaPIPES, NaHEPES and $NaHCO_3$ respectively and thus was considered negligible.