Department of Chemistry Seminar Series
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Environmental Mobilization of Tc-99 by High Valent Manganese Species Under Anoxic Conditions

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Technetium-99 is considered a risk driving radionuclide at several US Department of Energy (DOE) sites across the United States. The half-life of Tc-99 is $2.13*10^5$ years, long enough that it will persist for the foreseeable future, but short enough to give it substantial contribution to expected dose at contaminated sites. However, under anoxic conditions, TcO_4^- can be attenuated by reduction to TcO_2, which is sparingly soluble and environmentally immobile. The potential for *in-situ* reduction of Tc-99 has been demonstrated experimentally, and it is commonly stated that Tc(IV) is expected to be stable in environments lacking reactive oxygen species. However, the current body of literature largely overlooks the fact certain oxidants are known to exist in anoxic environments. Manganese oxides and Mn(III)-ligand complexes are known to persist in anoxic environments, and have standard reduction potentials rivaling that of O_2. Manganese oxides are have been shown to oxidize As(III) to As(V), Cr(III) to Cr(VI), U(IV) to U(VI) and many other environmental contaminants, and at rates much faster than O_2. Mn(III)-ligand complexes are far less studied than their oxide counterparts, but they have been shown to rapidly oxidize U(IV) to U(VI). Despite this, redox interactions of manganese oxides and Mn(III)-ligand complexes with Tc(IV) have not previously been studied.

In the present study, the kinetics of the oxidative dissolution of TcO_2 by manganese oxides and Mn(III)-ligand complexes were examined. The effect of various parameters such as pH and oxidant concentration on the kinetics of the reaction were investigated. Three manganese oxides with different structures and manganese oxidation states chosen. Three different ligands were also investigated. The results of the study indicate that both manganese oxides and Mn(III)-ligand complexes are capable of mobilizing Tc-99 via oxidation and thus pose a threat to the remediation of Tc-99.