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Comparison of thermal expansion of Tc and Re salts.

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99Tc is the most significant long-lived product of uranium fission, producing the largest fraction of the total long-lived radiation emissions of nuclear waste. Tc 7+ compounds are highly mobile in the environment. Relatively little is known regarding the solid state chemistry of Tc. Recently we studied the structural properties of (NH4)TcO4 (Tc7+) and confirmed that this is isostructural with (NH4)ReO4 adopting a tetragonal scheelite type structure in space group I41/a. The unit cell parameters of (NH4)TcO4 are strongly temperature dependent with the structure showing negative thermal expansion along both the a- and c-axis, albeit at different temperatures This behavior is significantly different to that previously reported for the isostructural oxide (NH4)ReO4, although we note that the data for (NH4)ReO4 was collected at much lower resolution. Nevertheless it is clear in the literature that the thermal expansion behavior of (NH4)ReO4 is highly anisotropic. Despite the difference in the thermal expansion between what we have observed for (NH4)TcO4 and that described by others for (NH4)ReO4 it is likely that the origin of the anomalous thermal expansion in is the same in both cases, namely it is a consequence of re-orientation of the ammonium ions in the surrounding cage of eight oxygen atoms. To verify this for (NH4)TcO4 requires we replace the ammonium cation with another small cation. Therefore we have compared the thermal behavior of AReO4 with ATcO4 to determine if Re oxides are suitable surrogates to predict the behavior of Tc oxides and how they may behave in the environment.

Summary

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