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Alkali technetium oxides and their relation to Tc loading in nuclear waste glasses

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Because of their radioactive nature, solid technetium-99 oxides have been rarely studied, and there is a dearth of modern spectra and diffraction patterns in the literature. This publication aims to address this by detailing a low-temperature synthesis method for pure, crystalline alkali pertechnetates, along with complete analysis by static and magic-angle-spinning nuclear magnetic resonance, Raman, neutron diffraction (ND), and X-ray absorption spectroscopy (XAS). Insight gained from these studies brings a deeper understanding of the periodic chemistry of alkali (Na,K,Rb,Cs) pertechnetates. Additionally, we report attempts to make 5- and 6-coordinate pertechnetate compounds of K, Na, and Li, i.e. TcO_5^- and TcO_6^-. It was found that higher coordinated species are very sensitive to water, and easily decompose into their respective pertechnetates. It was difficult to obtain pure compounds, but mixtures of the pertechnetate and other phases were frequently found, as evidenced by XAS, ND, and Raman spectroscopy. Additionally, we summarize other work on Tc loading in glasses, effect on glass structure, and salt formation in overloaded samples. For glasses, XAS chemometrics based on principal component analysis and linear combination fitting suggests that Tc is speciated first as isolated Tc^{4+}, as Tc content is increased, there is more Tc^{7+} which partitions first to K neighbors then to isolated sites. Preliminary work is reported assessing $HTcO_4$ and related compounds and their relationships to mechanisms of Tc volatility

Summary

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