Layered Bi-based Materials for Remediation of Subsurface ^{129}I and ^{99}Tc Contaminants

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Removal of the long-lived radionuclides technetium-99 (^{99}Tc) and iodine-129 (^{129}I) from the contaminated subsurface plums found in the 200 Area of the U.S. Department of Energy Hanford site, WA state, is a critical piece of the environmental cleanup and protection of the Columbia River. The dissimilar nature of these contaminants and inter-conversion among their various chemical forms and oxidation states necessitates unique solutions for each contaminant often hindering design and successful implementation of the remediation strategies. In Hanford groundwater and vadose zone, ^{129}I exists in various chemical forms including iodate (IO_3^-), iodide (I^-), and organoiodine. Removal of ^{129}I by simple ion exchange is often impeded by its occurrence in multiple chemical forms, regional variations in pH and the presence of competing ions in high concentrations. These obstacles make iodine removal exceedingly difficult.

One possible approach is to develop innovative engineered materials that (i) can remove contaminants of concern from groundwater and stabilize them in a stable matix for long-term disposition and/or (ii) enable leave-in-place strategies to mitigate the flux of contaminants from the vadose zone to the groundwater. Our research has been focused on designing and testing of such materials. It was discovered that layered Bi oxide/hydroxides and ternary Bi-M-Al (where M is divalent transition metal) materials resembling layered double hydroxides exhibit high affinity toward IO_3^-, I^-, pertechnetate TcO_4^- and/or Tc(I) tricarbonyl $[fac\text{-}Tc(CO)_3]^+$ coordination compounds which were found in the Hanford tank waste containing elevated organics content.

A library of the Bi-based materials has been synthesized and evaluated for IO_3^-, I^-, TcO_4^- and $[fac\text{-}Tc(CO)_3]^+$ removal from various aqueous matrices including Hanford groundwater. Materials exhibiting rapid and efficient sorption of these dissimilar species were selected for the further studies. Molecular, structural and electronic properties of the selected composites were characterized with an effort to identify the contaminat-solid matrix interactions responsible for the selective sorption. Distribution and speciation of Tc and I within the solid

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matrices were also assessed using a range of physical characterization techniques such as FTIR and Raman spectroscopies, X-ray diffraction analysis, scanning and transmission electron microscopies, and X-ray photoelectron spectroscopy.

This research suggests that Bi-based materials can be a viable remediation strategy for subsurface ^{99}Tc and ^{129}I contamination. This may provide an alternative iodine removal methodology from the well-established silver functionalized materials as they are toxic not only to humans but also to aquatic organisms and microbiota and interfer with simultaneous application of bioremediation approaches for other contaminants.