Governing factors of ^{99}Tc reductive removal by zero valent iron materials of a wide particle size range

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This study is developing a potential method to separate the radioactive contaminant, ^{99}Tc, from off-gas condensate to be recovered from the low activity waste vitrification facility at the U.S. Department of Energy's Hanford Site. Zero-valent iron (ZVI) was tested for the removal efficiency of ^{99}Tc from the liquid phase via reductive sorption of soluble Tc(VII), i.e. pertechnetate anion TcO_4^-, allowing immobilization of ^{99}Tc in the low valent state. Formation of immobile ^{99}Tc(0), incorporated into metal alloy, is a potential wasteform option which could encapsulate ZVI materials following reaction with ^{99}Tc [1].

Batch contact experiments were performed with a range (twelve) of ZVI materials using neutral pH NaCl solution to evaluate ^{99}Tc(VII) reduction kinetics and efficiency. ZVI materials were also monitored for formation of oxidation products and dissolution of Fe(II/III). Solid characterization of the ZVI materials and reduction kinetics variations were the important examination steps that allow us to proceed with the method development.

The four most promising ZVI materials were investigated further based on their reductive efficiency, purity, synthesis method, and particle size. Conditions were varied including potential washing procedures and initial solution pH.

Sorption results were correlated with the ZVI manufacturing method, particle size (from 100 nm to 2 mm), and surface area. Contrary to theory [2], ZVI reduction capability was not found to be dependent on specific surface area and particle size. Similar observations were obtained for ZVI with acid washing pretreatment. The potential for re-oxidation also varied with material. Our results imply that the manufacturing or synthesis methods of ZVI, presumably resulting in the dissimilar surface passivation and reactivity, constitute the most important factor affecting ^{99}Tc(VII) reduction, which should be accounted for in the selection of the most suitable and efficient ZVI.

[1] Serne *et al.* (2016) PNNL-25834. [2] Darab *et al.* (2007) *Chem. Mater* **19**, 5703-5713.