^{99}Tc incorporation and removal by iron mineral transformation

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Introduction

Technetium is generated in large quantities as a fission product in making of nuclear weapons and by irradiation of ^{235}U-enriched fuel during production of commercial power. Environmental concerns have been raised because of the long half-life and high mobility of ^{99}Tc in oxidizing subsurface environments. Even though reduced ^{99}Tc(IV) is more immobile, the reoxidation of ^{99}Tc(IV) by changing conditions is fast and results in a subsequent release of ^{99}Tc into the environment. Incorporation and retention of ^{99}Tc into Fe mineral is suggested to increase ^{99}Tc removal and limit ^{99}Tc reoxidation.

Results and Discussion

The results of mineral transformation tests using three synthesized Fe(oxy)hydroxide minerals [ferrihydrite, magnetite, and $Fe(OH)_2(s)$] showed that $Fe(OH)_2(s)$ transformed easily to a mixture of magnetite, maghemite, and goethite even at room temperature (RT) and circumneutral pH conditions. The transformation product from ferrihydrite with ^{99}Tc(VII) bearing alkaline solutions was solely goethite mineral in most of the conditions. Transformation product from reacting magnetite with ^{99}Tc(VII) bearing alkaline solutions was very limited and there were only small amounts of maghemite and goethite formed under high pH and temperature conditions. In addition, negligible ^{99}Tc removal from solutions was found in the resultant slurry when magnetite or ferrihydrite was used without aqueous Fe(II) addition.

Even though the most transformation products were found at high pH (~12) and temperature (75–80ºC) conditions, $Fe(OH)_2(s)$ can be used as an initial substrate for ^{99}Tc removal due to its high reactivity. Removal of ^{99}Tc from solution by $Fe(OH)_2(s)$ was fast and more than 95% of the initial ^{99}Tc (10^{-5} M) was removed from solution by the $Fe(OH)_2(s)$ transformation product, even without aqueous Fe(II) addition. Because $Fe(OH)_2(s)$ is oxidized to form magnetite (Fe_3O_4) by reacting with H_2O even in anoxic condition, the presence of $Fe(OH)_2(s)$ can provide aqueous Fe(II) to reduce ^{99}Tc(VII) to ^{99}Tc(IV). In addition, because the final pH of a slurry mixed with ^{99}Tc and $Fe(OH)_2(s)$ was fairly alkaline (pH = 9–11.5), ^{99}Tc removal was not considered to be from surface adsorption, but rather incorporation into transformed mineral product. Mineral transformation from Fe(OH)2(s) to a more stable Fe (oxy)hydroxide mineral can be used to effectively remove 99Tc(VII) in alkaline pH conditions germane to off-gas scrubber secondary waste and Hanford low-activity waste streams.