Effect of ^{99}Tc(IV) coprecipitation with respect to Fe minerals on ^{99}Tc retention in glass

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Introduction

Vitrification of Hanford Site's radioactive tank waste into borosilicate glass has been considered as the baseline immobilization method because of the high stability of the final glass product. However, at the high temperatures (~1,000–1,200 °C) used in vitrification, a significant fraction of volatile radionuclide such as technetium (^{99}Tc) is expected to escape. Low retention of ^{99}Tc in a final glass is attributed to rapid volatilization of ^{99}Tc species during the high-temperature melt process. Iron minerals are stable hosts for Tc immobilization. ^{99}Tc(IV)-incorporation within Fe mineral structures such as spinel has also been proposed as a novel method to increase Tc retention even at the high temperatures used in vitrification.

Results

Reduced ^{99}Tc, ^{99}Tc(IV), substitutes for Fe(III) in the crystal structure by a process of ^{99}Tc reduction from ^{99}Tc(VII) to ^{99}Tc(IV) followed by co-precipitation of Fe oxide minerals. Two ^{99}Tc-incorporated Fe minerals (^{99}Tc-goethite and ^{99}Tc-magnetite/maghemite) were prepared and tested for ^{99}Tc retention in glass melt samples at temperatures between 600 – 1,000 °C. After being cooled, the solid glass specimens prepared at different temperatures were analyzed for ^{99}Tc oxidation state using ^{99}Tc K-edge XANES. In most samples, ^{99}Tc was partially oxidized from ^{99}Tc(IV) to ^{99}Tc(VII) as the melt temperature increased. However, ^{99}Tc retention in glass melt samples prepared using ^{99}Tc-incorporated Fe minerals were moderately higher than in glass prepared using $KTcO_4$ because of limited and delayed ^{99}Tc volatilization.

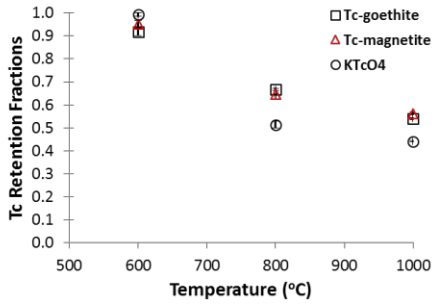

Fig.1. ^{99}Tc retention fractions in three different simulated glass melt samples prepared with ^{99}Tc-goethite, ^{99}Tc-magnetite, or $KTcO_4$ at three different temperatures (600 °C, 800 °C, and 1000 °C).