Savannah River Remediation

We do the right thing.

NRC Salt Waste Monitoring Technetium-99

April 26, 2011

SRR-CWDA-2011-00071

SRNL-STI-2010-00668

Long-term Technetium Interactions with Reducing Cementitious Materials

- Technetium Literature Review
  - Results and Conclusions
- Laboratory Study
  - Results and Conclusions

Literature Review

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- Purpose:
  - To compare relevant research on Tc interactions with slag-containing cementitious materials.

Literature Review Results

- Tc(IV) in a slag-cement converts to the more mobile Tc(VII) species when exposed to an oxidizing environment within a short time frame.
- SRS saltstone will reduce Tc(VII) in the absence of NaS or sodium dithionite in a reducing atmosphere but takes time to reach equilibrium as a function of slag content.
- Only trace concentrations of atmospheric oxygen at the high pH levels of cementitious systems is required to maintain Tc as Tc(VII).
- Experimental conditions must be responsible for wide variability of measured K_d values, such that they are either very low, ~1 mL/g, or they are very high ~1000 mL/g, suggesting that Tc(VII) or Tc(IV) dominate the systems.
  - Much of this variability appears to be the result of experimental conditions, especially direct controls of oxygen contact with the sample.

Literature Review Results

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- A field study conducted at SRS in the 1980s indicated that a slag-saltstone immobilized Tc for 2.5 years. Below background concentrations of Tc leached out of the slag-containing saltstone, whereas Tc leached out of the slag-free saltstone at the rate of nitrate loss.
  - One possible explanation for the immobilization of Tc in this study was that the slag-saltstone maintained reducing conditions within the core of the 55-gallon sample, whereas in the small-scale lab experiments, where samples were crushed to <1mm, oxygen diffused through the particles and reoxidize the slag during the contact period.
- Present site measured reduction capacity value of 820 µeq/g is in the realm of acceptable literature values that were either measured or theoretically estimated based on thermodynamic calculations.

Laboratory Study

- Purpose:
  - To measure Tc K_d values for saltstone under reducing conditions.

Laboratory Study Results

- Tc adsorption experiments were conducted under reducing conditions out to 56 days (< 0.5 ppm $O_{2(g)}$, -585 mV, 2% H_2, pH 11.66)
  - K_d values of ~1000 mL/g in a saltstone formulated with 45% slag (nominal concentration)
  - K_d of 10,000 mL/g when the saltstone contained 90% slag
  - K_d values logarithmically increased from 1 day to 56 days.
  - Steady state had not been achieved during the initial 56 days. However, the slag -free cement control samples also had K_d values near 1000 mL/g and extremely low redox conditions, due to the 2% H_2 atmosphere.

Laboratory Study Results

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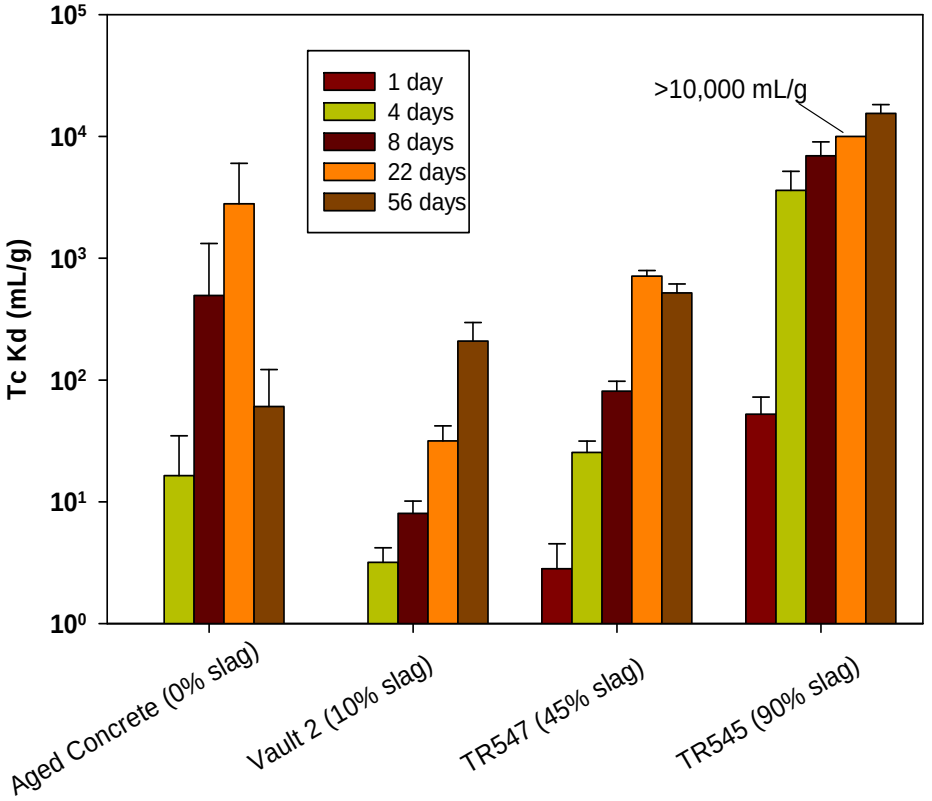
- Tc K_d Values (mL/g) under reducing conditions.
- Tc K_d values for various saltstone and concrete formulations measured after one, four, eight, twenty-two, and fifty-six day equilibration under reducing conditions.
- Total Tc concentrations in each system were 10 ppb and 5 ppb with each concentration set up in triplicate.
- The values expressed are averages of all six samples with the error bars representing the standard deviation.



Laboratory Study Results

- In a reducing environment, slag-containing cementitious material with the nominal composition of saltstone exhibits a K_d value of ~1000 mL/g.