



Contribution ID: 117

Type: Poster

Redox behavior of the Tc(VII)/Tc(IV) couple in dilute to concentrated NaCl and MgCl₂ solutions

Wednesday, 19 September 2012 18:00 (1h 50m)

For the long-term performance assessment of nuclear waste repositories, reliable predictions of radionuclide mobility have utmost importance. ⁹⁹Tc is a β -emitting long-lived ($T_{1/2} \sim 211.000$ a) fission product highly relevant for nuclear waste disposal. Tc(VII) and Tc(IV) are the most stable and dominant redox states of technetium under environmental conditions. Heptavalent Tc exists under non-reducing conditions as the highly soluble TcO₄⁻ pertechnetate anion. Technetium present in the tetravalent oxidation state, however, forms hydrous oxides (TcO₂·xH₂O(s)) which drastically limit Tc solubility and mobility. Because of the significant impact on Tc chemistry, the redox behaviour of the Tc(VII)/Tc(IV) couple needs to be investigated in detail. In order to predict Tc behaviour for nuclear waste disposal in rock salt formations, dedicated studies under saline conditions are required.

In this study, the redox behaviour of the Tc(VII)/Tc(IV) couple was investigated in dilute to concentrated saline systems. Reduction experiments were performed under Ar atmosphere by using different homogenous and heterogeneous reducing systems in NaCl (0.5 M and 5.0 M) and MgCl₂ (0.25 M, 2.0 M and 4.5 M) solutions to provide for repository-relevant geochemical conditions. Eh and pH were measured after given aging times and the results systematised according to Pourbaix diagrams in order to assess the redox behaviour of technetium in highly saline systems. Technetium concentrations, redox state and aqueous speciation were analysed. The results show a clear effect of the specific reducing systems and ionic strength conditions on measured Eh values and Tc(VII)/Tc(IV) redox transformations. The experimental data are compared to thermodynamic calculations and geochemical model predictions.

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Session Classification: Poster Session

Track Classification: Radioactive elements in the environment, radiation archeometry and Health Physics