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Sorption of Tc(IV) to geologic materials associated with a geological disposal facility

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Technetium-99 is one of the most important isotopes likely to be disposed of in the proposed UK Geological Disposal Facility (GDF) for higher-activity radioactive wastes. This is due to its long half life (2.13 x 105 y), fission yield (6%), and its ability to migrate through soils and other environmental components when in its oxidised, pertechnetate, TcO4- form. However, much of the technetium in the GDF is likely to be in the lower oxidation state of Tc(IV) as TcO2(am) or TcO(OH)2(aq). Therefore, an important aspect of the behaviour of technetium in the near- and far-fields of a GDF is its sorption to geologic and cementitious materials in its reduced (Tc(IV)) oxidation state.

Batch sorption experiments on technetium in both oxidised and reduced forms have been conducted in the presence of various materials which are associated with a deep geological disposal facility. These solids can be placed in the following categories:

Single minerals;
Sedimentary rocks;
Crystalline rocks;
Carbonates;
Clays and clay minerals;
Concretes and cements.

Tc(IV) was produced by electrochemically reducing a solution of Tc-95m, (t1/2 = 60 days,) pertechnetate which was used as a spike added to a carrier solution of reduced Tc-99 pertechnetate solution. Tc(IV) solutions were used at concentrations from 10-9 to 10-11 mol dm-3.

The results for these studies show Rd values ranging from 0 –39 000 ml g-1 depending on pH and solid material. The results for both oxidised and reduced technetium in the presence of solids associated with a geological disposal facility are presented here. It is noticeable that the Rd is very dependent on the pH. For example, for a 10-9 mol dm-3 technetium solution in contact with bentonite at pH 7, Rd = 21 ml g-1, however, at pH 10 for the same concentration of technetium Rd = 2974 ml g-1.

Surface complexation modelling of the data generated has been performed, as well as characterisation of the solids. In general using bidentate constants give best agreement between model and data, e.g. for bentonite:

Bent-OH + H+ ↔ Bent-OH2+ (Log K = 4.5*)*
Bent-OH ↔ Bent-O- + H+ (Log K = -7.9)
Bent-OH + Na+ ↔ Bent-ONa + H+ (Log K = -0.1)
Bent-OH + TcO2+ + H2O ↔ (Bent-O)2TcO(OH)- + 3H+ (Log K = -0.75)
* Grambow et al. Radiochim. Acta, 94, 627

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