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## The Effect of pH and Ionic Strength on the Dissolution of TcO<sub>2</sub> in Hyperalkaline Environments

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The UK has a relatively high inventory of <sup>99</sup>Tc in the wastes to manage. Under the planned cementitious Geological Disposal facility (GDF) conditions of high pH and low Eh most Tc should be immobile as amorphous technetium dioxide. Nevertheless, not much is known about the behaviour of this solid phase. The Loughborough University task within work-package 3 was to investigate the dissolution behaviour of amorphous Tc (IV) oxide containing the isotope Tc-99 as a function of various parameters, viz. pH (10.5, 12.5 and 13.3), ionic strength (I = 1 mol/L and I = 3 mol/L), and the age of the solid phase (1 day to 1 year).

To this aim, fully-reduced, high purity <sup>99</sup>-TcO<sub>2</sub>(s) was produced electrochemically from ammonium pertechnetate. The ammonium pertechnetate solution was adjusted to pH 10.5, 12.5 or 13.3 and ionic strength 1.0 mol L<sup>-1</sup> or 3.0 mol/L before reduction for 2 hours at 6 V in the presence of 0.01 mol/L sodium dithionite as a holding reductant. Five replicates of each system were set-up and kept in an anaerobic glove box for up to 12 months for sampling and counting by LSC.

### Results and Discussion

#### Effect of ionic strength at pH 10.5

The steady-state aqueous concentration of Tc changed from  $12.8 \pm 1.0$  nmol/L to  $17.0 \pm 2.1$  nmol/L as the ionic strength rose from 1.0 to 3.0 mol/L in the presence of NaCl as a background electrolyte. This change has been investigated using the ANOVA single factor test and is significant at the 99% confidence level. Therefore, it can be said that the change in ionic strength has affected the steady-state aqueous concentration of Tc. Until a new set of experiments has been set-up it is not possible to determine if this is due to changes in the solid phase of chloride complexation or some other factor(s).

#### Effect of ionic strength at pH 12.5

The data show that there is no effect of changing the ionic strength from 1.0 mol/L to 3.0 mol/L at pH 12.5

#### Effect of ionic strength at pH 13.3

The measured aqueous concentration of Tc changed from  $175 \pm 33$  nmol/L to  $356 \pm 59$  nmol/L as the ionic strength rose from 1.0 to 3.0 mol/L. As the pH 13.3, I = 3.0 mol/L system has not reached steady state, no firm conclusion can be drawn except to say that it appears as though the same ionic-strength-effect as at pH 10.5 is being observed. Table 1 summarises the final measured aqueous concentrations of Tc measured.

#### pH Ionic Strength Steady state [Tc]

(mol/L) (nmol/L)

10.5 1  $12.8 \pm 1.0$

10.5 3  $17.0 \pm 2.1$

12.5 1  $29.6 \pm 3.1$

12.5 3  $30.3 \pm 2.7$

13.3 1  $175 \pm 33$

13.3 3  $356 \pm 59^*$

Table 1: Steady-state supernatant concentration of <sup>99</sup>Tc from pH 10.5 to 13.3 at ionic strengths of 1.0 and 3.0 mol/L, 5 replicates of each, errors  $\pm 1$  s.d. \*Not at steady-state after 12 months

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