

Reduction and Sequestration of Pertechnetate to Technetium Dioxide and Protection from Reoxidation - 13012

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Contractor for the U.S. Department of Energy
Office of River Protection under Contract DE-AC27-08RV14800



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Reduction and Sequestration of Pertechnetate to Technetium Dioxide and Protection from Reoxidation – 13012

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ABSTRACT

This effort is part of the technetium management initiative and provides data for the handling and disposition of technetium. To that end, the objective of this effort was to challenge tin(II)apatite (Sn(II)apatite) against double-shell tank 241-AN-105 simulant spiked with pertechnetate (TcO_4^-). The Sn(II)apatite used in this effort was synthesized on site using a recipe developed at and provided by Sandia National Laboratories; the synthesis provides a high quality product while requiring minimal laboratory effort.

The Sn(II)apatite reduces pertechnetate from the mobile +7 oxidation state to the non-mobile +4 oxidation state. It also sequesters the technetium and does not allow for re-oxidation to the mobile +7 state under acidic or oxygenated conditions within the tested period of time (6 weeks). Previous work indicated that the Sn(II)apatite can achieve an ANSI leachability index in Cast Stone of 12.8.

The technetium distribution coefficient for Sn(II)apatite exhibited a direct correlation with the pH of the technetium-spiked simulant media.

INTRODUCTION

Technetium (Tc-99) is a major fission product from nuclear reactors, and because it has few applications outside of scientific research, most of the technetium will ultimately be disposed of as nuclear waste. The radioactive decay of Tc-99 to ruthenium 99 (Ru-99) produces a low energy β^- particle (0.1 MeV max). However, due to its fairly long half-life ($t_{1/2} = 2.13E05$ years), Tc-99 is a major source of radiation in low-activity waste [1].

Technetium forms the soluble oxy anion pertechnetate (TcO_4^-) under aerobic conditions. This anion, which is very mobile in groundwater, poses a health risk [2]. Several reports have been published on the mobility of the pertechnetate ion in the environment. For example, Tc-99 has been shown to increase in the western Irish Sea as a direct result of the increases in discharge of Tc-99 from the Sellafield reprocessing site. During the period of 1996 to 1998, the mean committed effective doses to Irish typical and heavy seafood consumers due to Tc-99 were 0.061 μ Sv (0.0061 mrem) to 0.24 μ Sv (0.024 mrem) respectively. Technetium-99 emanates from the Enhanced Actinide Removal Plant at Sellafield which removes actinides but is ineffective at removing Tc-99 [3]. In a study by the International Atomic Energy Agency, it was reported that very mobile radionuclides such as technetium have a soil to plant transfer factor of 100 or greater.

The soil to plant transfer factor (dimensionless) is defined as the ratio of the activity concentration of the radionuclide in the plant (Bq/kg) to that in the soil (Bq/kg). Furthermore, it was reported that technetium is very mobile in the soil and may be subject to considerable migration to deeper soil layers [4].

Researchers have identified that redox conditions play an important part in the retardation of mobility of the pertechnetate species [5, 6]. Capitalizing upon the redox conditions and using Sn(II)apatite, further work has demonstrated that pertechnetate is reduced to the non-mobile TcO_2 species. It was also demonstrated that technetium-loaded Sn(II)apatite achieves a leachability index in Cast Stone of 12.8 [7, 8]. The mechanism appears to be an exchange with the phosphate group in the apatite molecule. It has been reported that the apatite lattice is very tolerant to substitutions [9, 10].

This study is part of the technetium management initiative to provide data for the safe handling and disposition of technetium in Hanford tank waste treatment. To that end, the objective of this study was to challenge Sn(II)apatite against double-shell tank 241-AN-105 (AN-105) simulant spiked with pertechnetate (TcO_4^-).

MATERIALS AND METHODS

Simulant

Envelope A (also known as Double-Shell Slurry Feed and Double-Shell Slurry) was chosen because it is an alkaline ($[OH^-] > 1 M$), high sodium ($>8 M$) supernate. The AN-105 supernate recipe [11] was formulated and spiked with 15 mg/L of Tc-99. Table 1 shows the simulant recipe.

The simulant was formulated following the directions of Dr. Dan Herting, Process Chemistry, Washington River Protection Solutions LLC. First, all of the acid nitrate salts were mixed in a small portion of de-ionized (DI) water and set aside for later addition. Next, the aluminum source was dissolved; sodium aluminate was used as it dissolves quickly in 60 to 70 °C NaOH. After the sodium aluminate had dissolved, the temperature was maintained at 40 to 60 °C, and the remaining salts were added beginning with those which dissolve more slowly. Sodium nitrate and sodium nitrite were added last. If the simulant were to be formulated with the organic component, then acidic organics such as oxalic acid would be added with the acid salts, while sodium oxalate would be added with sodium nitrate. The simulant presented a very small amount of solids after 24 hours and was filtered using a 0.45 μm PVDF filter.

**Table I. Tank 241-AN-105 Double-Shell Tank Simulant
Chemical Makeup (1 L Batch). (2 pages)**

Reagent	Formula	g/L
Boric Acid	H_3BO_3	0.292
Cadmium Nitrate	$Cd(NO_3)_2 \cdot 4H_2O$	0.009
Calcium Nitrate	$Ca(NO_3)_2 \cdot 4H_2O$	0.236
Cesium Nitrate	$CsNO_3$	0.024
Lead Nitrate	$Pb(NO_3)_2$	0.085
Magnesium Nitrate	$Mg(NO_3)_2 \cdot 6H_2O$	0.057

**Table I. Tank 241-AN-105 Double-Shell Tank Simulant
Chemical Makeup (1 L Batch). (2 pages)**

Reagent	Formula	g/L
Potassium Nitrate	KNO ₃	19.221
Silver Nitrate	AgNO ₃	0.026
Zinc Nitrate	Zn(NO ₃) ₂ •6H ₂ O	0.046
Glycolic Acid	HOCH ₂ COOH, 70 WT%	1.665
Sodium Chloride	NaCl	14.984
Sodium Fluoride	NaF	0.420
Sodium Chromate	Na ₂ CrO ₄	4.205
Sodium Sulfate	Na ₂ SO ₄	1.140
Potassium Molybdate	K ₂ MoO ₄	0.204
Ammonium Acetate	CH ₃ COONH ₄	0.513
Aluminum Trihydroxide	Al(OH) ₃	114.77
Sodium Hydroxide	NaOH	196.68
Selenium Dioxide	SeO ₂	0.001
Sodium Metasilicate	Na ₂ SiO ₃ •9H ₂ O	2.135
Sodium Acetate	NaCH ₃ COO•3H ₂ O	3.865
Sodium Formate	HCOONa	4.351
Sodium Oxalate	Na ₂ C ₂ O ₄	0.929
Sodium Phosphate	Na ₃ PO ₄ •12H ₂ O	2.281
Sodium Carbonate	Na ₂ CO ₃	22.149
Sodium Nitrate	NaNO ₃	209.70
Sodium Nitrite	NaNO ₂	166.48
Water	H ₂ O	669.37

Pertechnetate

Pertechnetate was obtained as ammonium pertechnetate from the Oak Ridge High Flux Isotope Reactor. The material was solubilized in DI water, and appropriate dilutions were carried out and confirmed using a PerkinElmer^{®1} Elan DRC-e model inductively coupled plasma/mass spectrometer (ICP/MS).

For testing the concentration of pertechnetate remaining in the liquid after reduction by the Sn(II)apatite, a standard curve was generated using a Ludlum Model 2929 dual scaler, Figure 1. The standard curve was generated by comparing the 5-minute averaged and background-corrected counts to the analyses obtained using the ICP.

For the resin test, a sample of 200 µL of resin bed effluent at progressive bed volumes (BV) was pipetted onto a smear pad (Hi-Q Environmental Products Company, part number FP1441-20), allowed to dry, and subjected to a 5-minute count. The average count over the 5 minutes was background corrected and compared to the standard curve in Figure 1 to determine the concentration of pertechnetate.

¹ PerkinElmer[®] is a registered trademark of PerkinElmer, Inc., Wellesley, Massachusetts.

cpm vs 99Tc in simulant

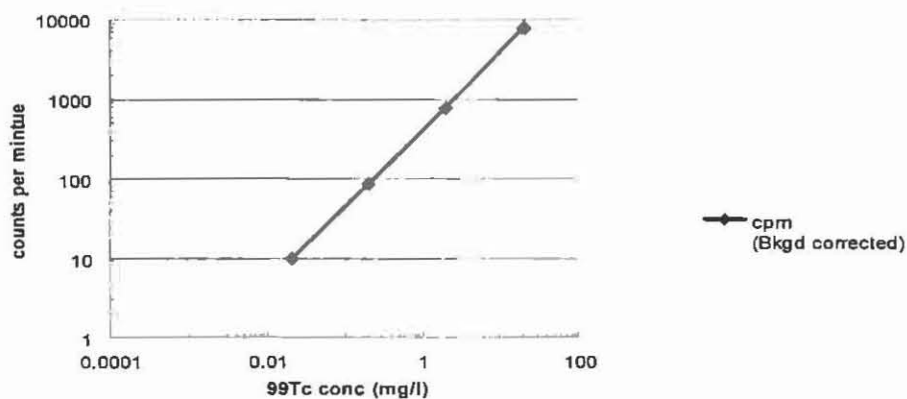


Figure 1. Calibration Curve using Ludlum Model 2929.

Tin(II)Apatite Synthesis

Sn(II)apatite was synthesized using a ChemGlass^{®2} 2-L reactor, as shown in Figure 2. One liter of DI water was added to the reactor and heated to boiling to eliminate as much carbon dioxide as possible. According to McConnell and Foreman (10) "carbon dioxide substitution for phosphate will take place unless great care is exercised to exclude carbon dioxide from all reagents including the water" during the preparation of apatite. After several minutes of boiling, the heat was reduced to 37 °C and high purity nitrogen gas was sparged into the water. Three milliliters of hydrazine was added after the temperature reached 37 °C for reducing conditions.

² ChemGlass is a registered trademark of ChemGlass, Inc., Vineland, New Jersey.

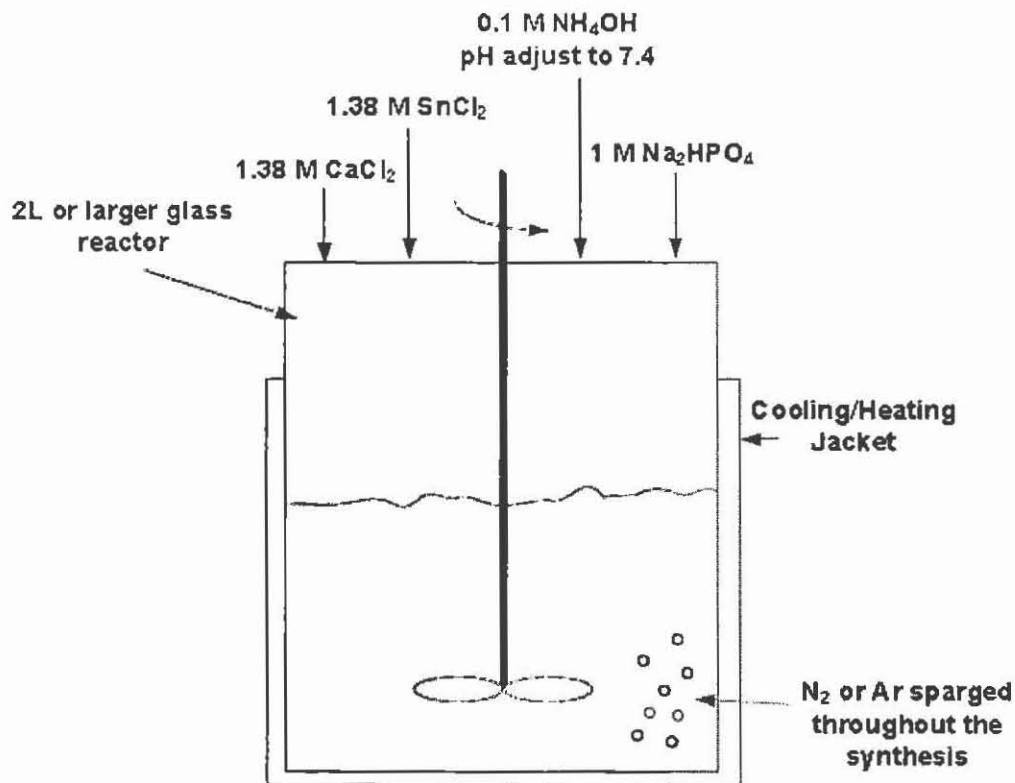


Figure 2. Bench Set-up for Synthesis of Sn(II)apatite.

The procedure to synthesize Sn(II)apatite is as follows:

Note: The Sn(II) must be kept from oxidizing during the synthesis. This is accomplished by keeping all oxygen out of the system and/or adding a reducing agent (3 mL of hydrazine) to the SnCl_2 reagent solution and the initial DI H_2O in the reactor vessel.

1. Bring the water (1 L) in the reactor to a boil and cool to 37 °C while sparging with N_2 or Ar gas. Add the reducing agent.
2. Prepare 250 mL of 1.0M Na_2HPO_4 solution and adjust the pH to 7.4 with NH_4OH . Purge any air from the flask.
3. Prepare 250 mL of 1.38 M CaCl_2 solution and adjust the pH to 7.4 with NH_4OH . Purge any air from the flask.

4. Prepare the SnCl₂ solution by adding 1 mL of hydrazine to 250 mL of DI, degassed water, and then add the SnCl₂ to make a 0.4-M solution. Purge any air space in the vessel with N₂ or Ar and seal the vessel. Do not adjust the pH.
5. While maintaining the reactor at 37 °C, slowly add the reagents at the flowrate of approximately 1 mL/min., all the while vigorously stirring and bubbling N₂ or Ar through the solution.,
6. The pH of the solution must be maintained at approximately 7.4 ±0.2. The entire first phase of the procedure takes about 6 hours to complete.
7. After all reagents have been added, keep stirring the mixture and bubbling in N₂ or Ar overnight, maintaining the temperature at 37 °C and the pH at 7.4 at all times. For a more crystalline material, let it set for additional time, 2-3 days to allow Ostwald ripening.
8. Filter the Sn(II)apatite using a Buchner funnel and 0.65-µm filter paper. Repulp the Sn(II)apatite and filter it two more times. Place the product apatite into a Pyrex^{®3} vial and dry for 24 hours at 50 °C under a constant flow blanket of dry, high purity nitrogen. [If the material is placed in an oven to dry, the Sn(II) will oxidize to Sn(IV).]

Upon completion of this procedure, the Sn(II)apatite formed large white aggregates as shown in Figure 3. In order to achieve more surface area for use in the contact experiments, the Sn(II)apatite was placed in liquid nitrogen to minimize oxidation of the tin and ground to a fine powder using a mortar and pestle.

Challenges to Tin(II)Apatite

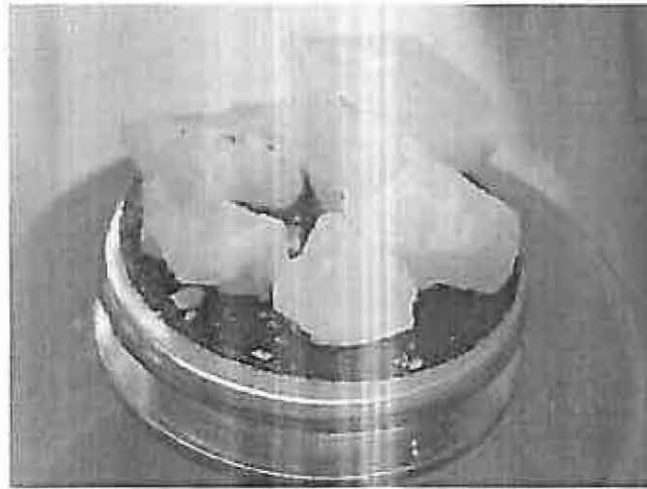
Initially, the main interest in the Sn(II)apatite was to reduce and sequester pertechnetate from a tank waste simulant of high pH and high ionic strength. However, the Sn(II)apatite may also have value in the reduction and sequestering of pertechnetate in other less aggressive media. To that end, the Sn(II)apatite was challenged across the pH range with varying molarities of sodium in the AN-105 simulant for the high pH range, as well as with DI water and nitric acid spiked with pertechnetate for the lower pH ranges.

RESULTS AND DISCUSSION

Tin(II)Apatite

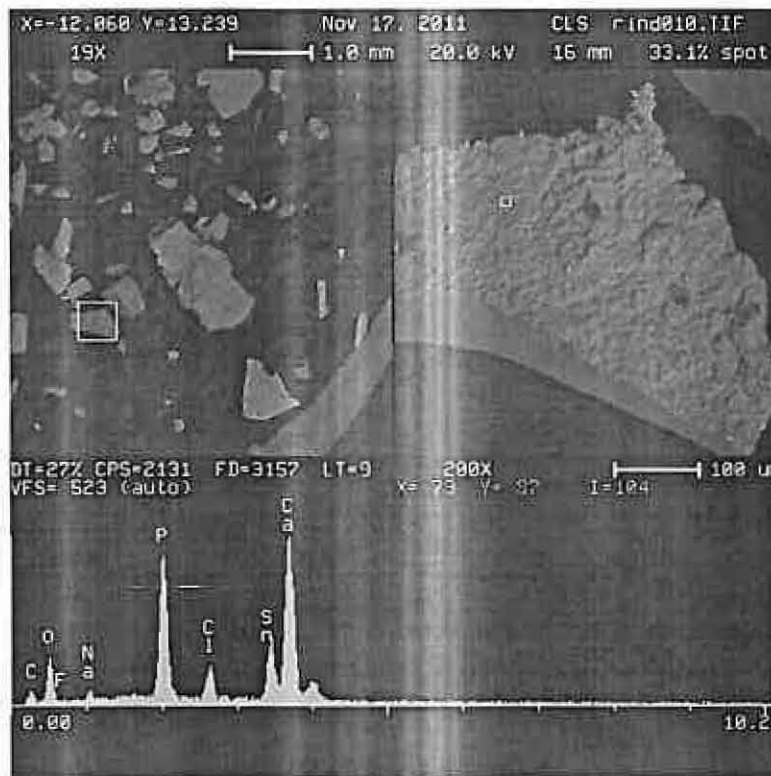
Figure 3 shows the synthetic Sn(II)apatite product.

³ Pyrex[®] is a registered trademark of Corning Glass Works Corporation, Corning, New York.



**Figure 3. Sn(II)apatite on a Scanning Electron Microscope Mount.
(Mount is 1 cm diameter.)**

Figure 4 shows a secondary electron image with electron dispersive spectrometry (EDS) spectrum indicating the relative intensities of the identified elements.



**Figure 4. Secondary Electron Image with Electron Dispersive Spectrometry of
Sn(II)apatite.**

An X-ray diffraction (XRD) scan is presented in Figure 5. The XRD pattern indicates the presence of a poorly crystallized hydroxyapatite, but does not indicate the presence of either a crystalline Sn(II) or Sn(IV) compound. It is known that poorly crystallized apatite has a higher solubility and is much more reactive than highly crystalline apatite. These results are consistent with the rapid reduction and immobilization of technetium by this material.

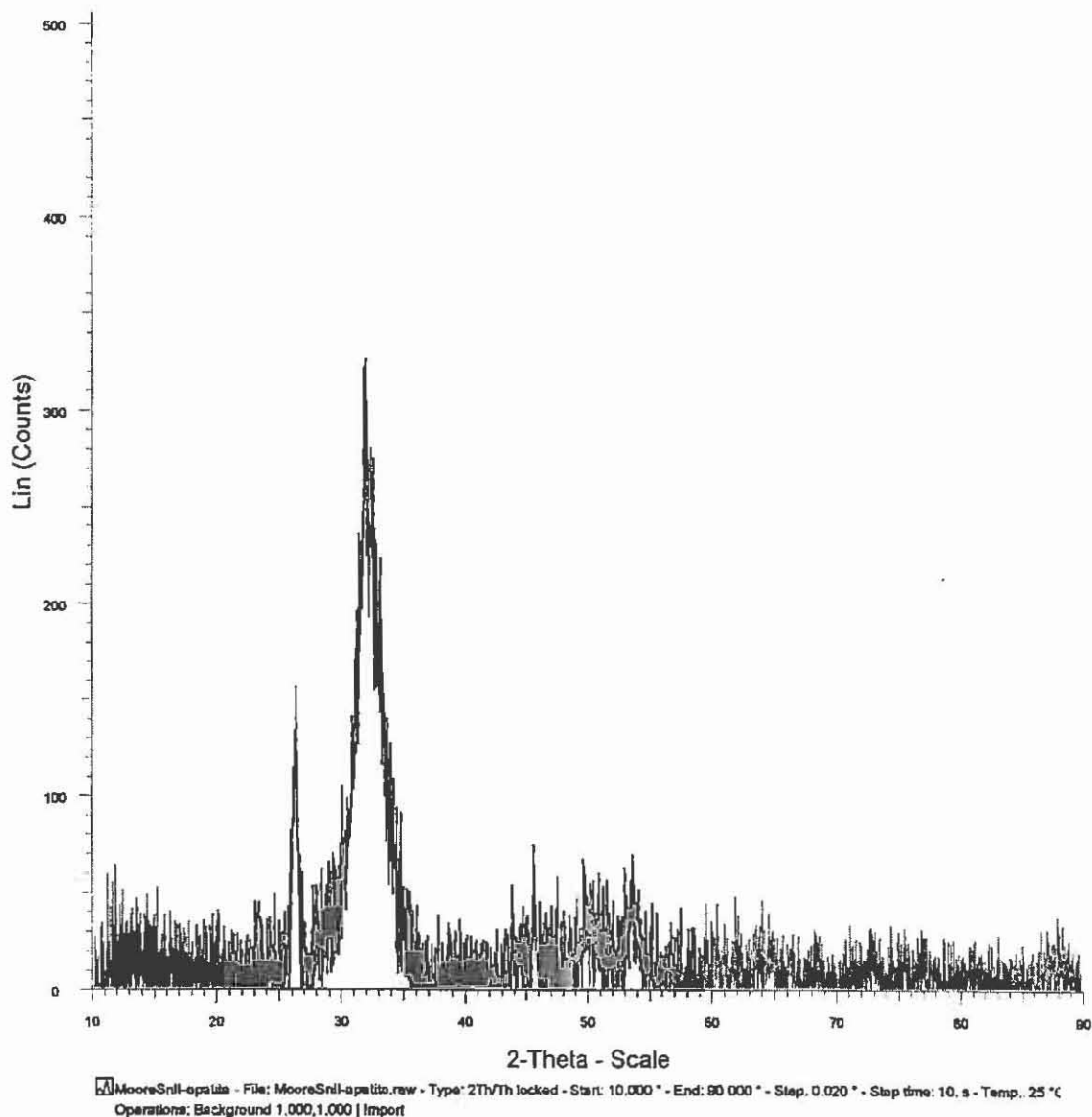


Figure 5. X-ray Diffraction Pattern of the Sn(II)apatite Formulation.

Distribution Coefficients

The Sn(II)apatite was initially challenged with the AN-105 simulant. In order to determine the range of usability in the field, the Sn(II)apatite was challenged against a range of pH and sodium molarities. Included in the challenge was DI water (spiked with pertechnetate). Table II shows the results of the distribution coefficients (K_d) as a function of pH. The challenge media from AN-105 simulant was made from dilutions of the initial 10-M Na simulant. The dilutions were carried out using Milli-Q^{®4} water with no visible precipitate formed. The pH values reported in Table II are measured values, and Appendix A presents the data from which the distribution coefficients were derived.

The K_d was determined using Equation 1 [12]:

$$K_d = [\text{mass in the solid phase}]/[\text{mass in liquid phase}] * V_s/m_e \quad (\text{Eq. 1})$$

Where:

- K_d = distribution coefficient
- V_s = volume of the solution
- m_e = mass of the sorbent (dry weight)

Table II. Distribution Coefficients as a Function of Sodium Molarity and pH.

Challenge Media	Initial Tc-99 Concentration (mg/L)	Distribution Coefficient, K_d mL/g	pH
10 M Na	15.2	36	>14
1.25 M Na	17.1	68	13.3
0.125 M Na	17.1	703	12.5
0.0125 M Na	17.1	65,669	11.4
0.00125 M Na	17.1	170,900 ^a	10.2
0.000125 M Na	17.1	170,900 ^a	8.5
DI Water (spiked)	17.1	170,900 ^a	2.5
0.05 M HNO ₃	17.1	6,059	1.3
0.1 M HNO ₃	17.1	237	1
1.0 M HNO ₃	17.1	18	0.1

^a The results were below detection of the instrument. Therefore, the lower detection limit was used to calculate the K_d .

As indicated in the above table, the Sn(II)apatite imparts an impressive distribution coefficient against pertechnetate within a pH range of 1 < pH < 12. However, when compared to ion exchange sorbents such as Purolite^{®5} A530E which achieves a distribution coefficient of ~900 in a 10-M sodium AN-105 simulant [13], the Sn(II)apatite does not perform as well in the higher alkalinity ranges.

Nitric Acid Leach Tests

In order to ascertain how well the Sn(II)apatite protected the TcO₂ from re-oxidation to the more stable and water-soluble pertechnetate species, a series of nitric acid washes was carried out. A mass of 200 mg of

⁴ Milli-Q[®] is a registered trademark of the Millipore Corporation, Billerica, Massachusetts.

⁵ Purolite[®] is a registered trademark of Brotech Corporation, Bala Cynwyd, Pennsylvania.

Sn(II)apatite was loaded with 0.311 mg of Tc-99. The Sn(II)apatite containing TcO₂ was mixed with the molarities of nitric acid (as indicated in Table II), gently shaken for 1 minute, and filtered using a syringe filter; the filtrate was then analyzed for technetium, using an ICP-MS. The process was repeated for a total of two washes. Table III presents the results of the test.

Table III. Weight Percent Technetium Lost in Nitric Acid Washes.

Solution	Tc-99 Leached into Filtrate (wt%)	
	First Wash	Second Wash
0.01 M HNO ₃	Non Detect	Non Detect
0.05 M HNO ₃	0.52	0.33
0.1 M HNO ₃	0.24	0.19
1 M HNO ₃	0.31	50.0 (after a 72-hour soak)

As can be seen from the above table, the apatite-technetium complex is stable against a robust oxidation attack. Only after leaving the loaded Sn(II)apatite sample in contact with 1 M nitric acid for 3 days was a considerable amount of Tc-99 (50 weight per cent) released. Due to budget and time constraints, the lower nitric acid molarities were not tested at longer times, and therefore a release rate cannot be determined. The test does indicate that Tc-99 is chemisorbed rather than physisorbed.

Water Column Leach Tests

The next test carried out was to ascertain whether or not the apatite-technetium complex would be subjected to releasing technetium if exposed to a more "environmentally friendly" oxidation. For the water leach test, 0.342 mg of technetium was captured by 200 mg of Sn(II)apatite, placed in a 200-mL graduated cylinder, and continually sparged with air. The results are presented in Table IV.

Table IV. Water Leach Test.

Exposure Time	Dissolved Oxygen (mg/L)	Technetium Concentration (mg/L) ^a
2 hours	8.7	<0.01
24 hours	8.6	<0.01
1 week	8.6	<0.01
2 weeks	8.4	<0.01
3 weeks	8.6	<0.01
4 weeks	8.4	<0.01
5 weeks	8.4	<0.01
6 weeks	8.6	<0.01

^a Activity determined by Ludlum and recalculated to mg/L.

The data from Table IV indicates the sequestered technetium is protected from re-oxidation from the technetium dioxide to the pertechnetate species in an oxygenated water column.

Proposed Sequestration Mechanism

In order to gain insight into the reactions associated with the technetium retention by Sn(II)apatite, pertechnetate was reacted with Sn(II)apatite in DI water. After the reaction, the water was analyzed for phosphorous and calcium. The results indicate that there were 0.173 mmol of technetium taken up by the apatite, while 0.177 mmol of phosphorous and 0.308 mmol of calcium were released. The results are presented in Table V.

Table V. Analyses of De-ionized Water Medium.

Amount used to Synthesize An(II)apatite M/L	Analyte in Solution	Filtrate from Milli-Q Water Leach of Sn(II) Apatite (mg/L)	Milli-Q Water mg/L by itself	Tc-99 Bearing Solution	Filtrate after Reaction of Sn(II)apatite with 17.1 mg/L Tc-99 Solution (mg/L)	Difference of Filtrates (mg/L)	Difference of Filtrates (mmol/L)
1	P	0.216	0.042	0	5.754	5.496	0.177
1.38	Ca	30.756	0.103	0	43.194	12.335	0.308
1.38	Sn	0.447 (ug/L)	ND ^b	0	0.465 (ug/L)	0.018 (ug/L)	1.52E-7
NA ^a	Tc-99	NA	NA	17.1	ND	17.1	0.173

^a NA = Not Applicable

^b ND = Not Detected

The calcium may be in excess in the control due to substitution of tin for calcium [10]. The calcium that appeared in the Sn(II)apatite-loaded Tc-99 water analysis may well be a calcium phosphate compound, which cannot be ruled out, and it is beyond the present scope to ascertain the exact composition of the leaving analytes.

The more probable explanation is that the technetium dioxide substituted for phosphate. The apatite lattice is very tolerant of substitutions, vacancies, and solid solutions [9]; the phosphate can be replaced by arsenate (AsO_4^{3-}), orthovanadate (VO_4^{3-}), silicate (SiO_4^{4-}), or carbonate (CO_3^{2-}).

The protection of reduced technetium in the Sn(II)apatite complex from re-oxidation by nitric acid and less robust oxygenated water indicates that there is actual incorporation into the crystal structure rather than a mechanism such as surface physisorption.

Truly answering the question of what mechanism occurs to sequester technetium dioxide and protect from reoxidation would involve *ab initio* quantum calculations involving van der Waals radii, electronegativity values (Pauling), and lattice forces, to name a few parameters. A possible place to begin would be the use of the Born-Oppenheimer Schrödinger equation. The Born-Oppenheimer Hamiltonian, giving total energy of the system, would allow calculations of the electron kinetic energy, nuclear attraction energy, electrical repulsion energy, and the nuclear repulsion energy. It is highly recommended that a mechanism be delineated to aid in the

understanding of the protection that Sn(II)apatite will afford to the reoxidation of technetium dioxide to pertechnetate.

CONCLUSIONS

From the results presented and discussed above, the Sn(II)apatite is not applicable to being used on direct Hanford tank waste at the high pH that it would encounter.

However, for diluted waste streams (pH between 2 and 11), it appears to be a robust candidate for sequestering Tc-99 from a liquid stream and immobilizing it in a solid crystal form.

The indications are that the Sn(II)apatite will lend itself to sequestering and inhibiting the re-oxidation to the mobile pertechnetate species, thus keeping the radionuclide out of the environment. The Sn(II)apatite would be efficacious in the reduction and sequestration of pertechnetate in the following scenarios, for example:

- water elution from pertechnetate resins such as SuperLig^{®6} 639
- treatment of pertechnetate-containing waste streams from the Effluent Treatment Facility
- treatment of the off gas from the Waste Treatment Plant to capture technetium
- groundwater barrier to prevent pertechnetate mobility.

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⁶ SuperLig[®] is a registered trademark of IBC Advanced Technologies, Inc., American Fork, Utah.

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