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Dr. R. J. Starmer
Geotechnical Branch
Office of Nuclear Material Safety and Safeguards
U.S. Nuclear Regulatory Commission
623-SS
Washington, D.C. 20555

Dear John:

Enclosed is a copy of letter report L-290-2, "Studies of the Surface Character of Basalt and Its Ability to Absorb Technetium," for your review and comments. This is the work Ron did last summer at ORNL; his more recent work will be reported on at a later date.

Sincerely,

Susan K. Whatley, Manager
Engineering Analysis and Planning

SKW:kk

Enclosure

cc: W. D. Arnold
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1049

LETTER REPORT

TITLE: Studies of the Surface Character of Basalt and Its Ability to Adsorb Technetium

AUTHOR: Ronald J. Clark
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Florida State University
Tallahassee, Florida 32306

PROJECT TITLE: Laboratory Evaluation of DOE Radionuclide Solubility Data and Selected Retardation Parameters, Experimental Strategies, Laboratory Techniques, and Procedures

PROJECT MANAGER: A. G. Croff

ACTIVITY NUMBER: ORNL No. 41 37 54 92 4 (189 No.B0290)/NPC No. 50 19 03 01

HIGHLIGHTS

- * Batch contact sorption experiments were employed to study surface effects on the sorption of technetium (present in solution as the pertechnetate anion) by McCoy Canyon basalt from the BWIP site. Crushed basalt was prepared both by grinding in agate apparatus in air, and also by grinding and storing the basalt under an argon atmosphere containing one to two ppm oxygen.
- * Basalt that was ground in air had little or no sorption capability for technetium, whereas basalt ground and stored under the inert atmosphere adsorbed significant amounts of technetium in tests lasting a few days.
- * It was found that oxygen must be rigorously excluded from the technetium-containing solutions in order for significant sorption to occur with argon-ground basalt. Vacuum freeze-pump-thaw cycles were found to be effective in lowering the oxygen content of the solutions to the ppb range.
- * High surface area basalt (< 325 mesh size) that had been ground in air showed some ability to adsorb technetium in experiments which extended for months.
- * Several types of heat treatment at 500°C of basalt that has been ground in air produced a material that was effective for technetium sorption. Hydrogen treatment was the most effective, followed by argon, and then vacuum.
- * These preliminary results suggest that McCoy Canyon basalt *in situ* which has not been exposed to air may be effective for the sorption of technetium from migrating groundwater. They further suggest that basalt that had been crushed and stored in air may be ineffective for technetium sorption. Thus, crushed basalt backfill which has been mined and stored on the surface might not retain technetium in the engineered facility.

1. INTRODUCTION

Oak Ridge National Laboratory (ORNL) is supporting the NRC staff analysis of geochemical information related to potential DOE high-level waste (HLW) repository sites with a project to evaluate key radionuclide values and techniques which may be employed by DOE in performance assessment calculations to show compliance with regulatory requirements. This project is focused on parameters which are relevant to the mobilization and migration of radionuclides, radionuclide sorption and apparent concentration limits under anticipated repository conditions. The ORNL work will parallel the schedule of the DOE repository projects, resulting in initial emphasis on the Basalt Waste Isolation Project (BWIP) candidate site on the Hanford reservation in Richland, Washington. Surveys of existing BWIP literature lead to the pursuit of the following tasks during the first year of the project:

1. Experimental measurement of the sorption and limiting radionuclide concentration of technetium under a variety of conditions.
2. Experimental measurement of the sorption and limiting radionuclide concentration of neptunium under a variety of conditions.
3. Geochemical modeling with computer codes designed to calculate groundwater compositions and radionuclide speciation and solubility.

As a result of past experimental work on measuring geochemical parameter values for basalt, questions have arisen concerning the extent to which the methods used to prepare the basalt alter it, and thus affect subsequent experimental results. Inconsistent results found among various investigators (1) suggested that the experimental samples of basalt may have been altered during preparation. Basalt is a very hard material and the necessary grinding operations are not done easily. If basalt minerals were in a reduced state in the natural deep ground situation, then grinding in air could result in surface oxidation of species such as Fe(II). Since it appears likely that a surface phenomenon is involved in sorption, then it will not take too much oxidation to change totally the ability of the basalt to effect redox reactions. Thus, a small task was undertaken to evaluate

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whether the methods employed to prepare the basalt samples for studies to determine technetium sorption values and concentration limits have an effect on the values subsequently obtained.

2. EXPERIMENTAL

The basic procedure in this work was the batch adsorption technique. Specially made 15 x 70 mm Pyrex tubes which had 7 mm tubing extension on the top for loading and sealing were used. The basalt (generally 1.00 g) was loaded in the tube and the tube evacuated to 0.01 Torr and brought to a pressure of about one atmosphere with a gas such as hydrogen or argon. The tube was heated in a furnace for 30 min and then cooled while the tube was connected to the gas supply. If the heating was done in vacuum, the tube was subsequently brought to one atmosphere with argon after it returned to room temperature. All of the remaining operations such as the addition of tracers and groundwater were done quickly to avoid loss of the basalt surface condition. (Subsequent discussion will show that the surface can stand only mild exposure to the atmosphere.) Technetium-95m and/or technetium-99 (in the pertechnetate form in solution) and then 8.00 mL of groundwater were added. If oxygen was to be removed, the water was briefly sparged with argon to speed the process and finally the tube was connected to the vacuum system. The pressure was lowered to about 40 Torr before isolating the vacuum. At that point, degassing was visually obvious and the solutions appeared to be boiling. However, the pressure was still above the vapor pressure of the water. The tube was lowered by stages into liquid nitrogen. This procedure was necessary to avoid tube breakage through water expansion. Once frozen, the tubes were evacuated to about 0.01 Torr pressure and then isolated from the vacuum. The ice was melted in warm water. This process was repeated through three or four complete cycles. After the last cycle, the tube was sealed by a torch.

An electrochemical method (2) was used to estimate the oxygen content of a 0.1 M NaCl-0.01 M NaHCO₃ solution deoxygenated by this freeze-thaw technique. The results indicated a level of about 10 ppb as compared to an air saturation value of 8 ppm.

In most of these experiments a synthetic groundwater, GR-2, (3) was used. In a few experiments other solutions were used to check the effects of other constituents of the solution. The pH of the groundwater was 9.52 initially but often decreased somewhat after contact with the basalt to about 9 and occasionally to values as low as 8.3.

The basalt used in these experiments was McCoy Canyon basalt ground carefully without any contact with metallic iron. In some cases the basalt was ground in air and in others in argon. The air-ground basalt was then screened to 70-325 mesh and washed ultrasonically to remove the fines. The basalt ground in argon was screened to the same mesh size but was not washed.

In most of the experiments, the surface treatment was heating the basalt to 500°C in hydrogen. The tube was evacuated at room temperature, and hydrogen admitted to a pressure of nearly an atmosphere. The tube was then placed in a preheated furnace for 30 min. During that time, the gas was pumped off every 10 minutes and renewed. This was done in an effort to make sure that all of the basalt came in contact with fresh gas since neither agitation nor gas flow was possible. For instance, visible quantities of water were driven off the basalt during the initial heating so that the water vapor could possibly prevent good gas-surface contact. However, it is not known whether these precautions were actually necessary. The tubes were cooled in gas at a pressure slightly above ambient.

These conditions were altered by variations in the temperature, by substituting argon for hydrogen, etc. When samples were heated in a hard vacuum, the vacuum was broken by argon after the tube was cooled to room temperature.

The loaded tubes were agitated on their sides on a shaker table heated to 60°C. They were centrifuged to settle the basalt and opened by the use of a file scratch-mark and hot glass cane. Technetium-95m was counted for the 0.204 Mev gamma in plastic counting tubes using a NaI well counter. The tubes in which the equilibrations were done were often checked for technetium activity. Generally they absorbed only a very small fraction of the total Tc-95m.

The reproducibility of the data was fairly good. The R_s value will be reported along with the percent technetium removed. In a typical set of three identical samples which varied only in the time of shaking, the quantities of technetium left in solution were 3.6, 2.1 and 2.9% or 2.9 ± 0.6 . Since there is no significant trend in the values, these data can simply be reported as having a R_s value of 292 ± 67 mL/g.

3. RESULTS

Comparison of Basalt Ground in Argon and in Air

In Table 1, results are shown for four sets of experiments in which the basalt was ground and treated in different ways. In the first three sets, the basalt was ground in argon and either protected from air or deliberately exposed to air by some method. Those samples that were freshly ground in argon adsorbed substantially more technetium than those that were exposed to oxygen by some method.

Long Term Experiments - Comparison of Fines and 7-325 Mesh Basalt

During the initial phases of the work, it appeared that there was no adsorption of technetium on the air ground basalt unless it was contaminated (with iron) or subsequently heat treated. However, some experiments started late in the summer and completed during the fall show that this is not the case. In Table 2, data are shown for 19-day and 99-day runs. The material was basalt ground in air to < 325 mesh. The samples generally contained Tc-99 in addition to the Tc-95 m tracer. After rejecting one of the six data points on the 99-day run, the average technetium remaining after equilibration at about 60° was 22.4% with an average R_s value of 30.2. Most of the fine basalt did not remain in suspension, rather it tended to "clump" up in piles which could not even be manually shaken loose. This may have been responsible for the scatter in the data.

A corresponding set of three samples using the 70-325 mesh ultrasonically washed basalt adsorbed no measurable quantity of technetium in the same 99-day run.

It is not clear whether the major difference between the two types of basalt during the 99 days is the varying particle size (and surface area) or the use of ultrasonic washing with air saturated water. It is clear that

Table 1
Freshly Ground Basalt Tests

Description of Samples	%Tc Remaining	%Tc Removed	Rs(mL/g)
Set 1: 10^{-8} M Tc			
Argon Ground and Protected	75.4%	24.6%	2.60mL/g
Argon Ground and Protected	74.7	25.3	2.77
Argon Ground and Sealed in Air	99.5	0.5	0.04
Set 2: 10^{-8} M Tc			
Argon Ground and Protected	83.4	16.6	1.72
Argon Ground and Protected	84.8	15.2	1.55
Argon Ground and Air Oxidized	93.9	6.1	0.52
Argon Ground and Sealed in Air	96.8	3.2	0.27
Set 3:			
Argon Ground and Protected	46.0	54.0	11.1
10^{-10} M Tc			
Argon Ground and Protected	70.2	29.8	3.9
10^{-8} M Tc			
Argon Ground and Protected	63.1	39.9	5.7
10^{-6} M Tc			
Argon Ground and Air Oxidized	89.7	10.3	0.95
10^{-8} M Tc			
Argon Ground and Sealed in Air	99.9	0.1	0.00
10^{-8} M Tc			
Set 4:			
Air Ground but Protected	96.8	3.2	0.26
Air Ground and Sealed in Air	97.3	2.7	0.22
Air Ground and Sealed in Air	98.2	1.8	0.15

Conditions: All samples except set 4 were ground in an argon atmosphere; protected samples were loaded in the atmosphere box and given a freeze-pump-thaw degassing (except set 4 which were loaded in air); The other samples were either allowed to oxidize in air for 2-3 hours or sealed in air. No heat treatment was given to any of these samples. Equilibration periods were one to two days. GR-2.

Table 2
Long Term Adsorption Studies with Air-Ground Basalt

Days of Experiment	%Tc Remaining	%Tc Removed	Rs (mL/g)
19	85.8	14.2	1.2
19	89.1	10.9	0.90
99	16.7	83.3	42.0
99	23.2	76.8	27.9
99	20.4	79.6	32.7
99	25.8	74.2	24.3
99	25.7	74.3	24.3
99	47.7	52.3	9.2

Conditions: 1.00 g < 325 mesh basalt, 8.00 mL GR-2, Tc-95m tracer. In the 99 day samples, 200 μ L of either 10^{-6} or 10^{-8} Tc-99 was added. The labels rubbed off during the shaking and the results did not permit the samples to be sorted out. In the long runs, the basalt "clumped" up to various degrees making consistent results quite unlikely. In the 19 day samples, the 14.2% removal sample contained Tc-95m only and in the 10.9% sample, 100 μ L of 10^{-6} Tc-99 was added. Equilibration at 60°C.

Table 3
Hydrogen Treatment at Various Temperatures

Temperature	% Tc Remaining	% Tc Removal	Rs (mL/g)
500°	3.0	97.0	262
500°	7.4	92.6	101
450°	5.7	94.3	134
400°	10.7	89.3	67
350°	57.3	42.7	6.0
300°	83.7	16.3	1.6
175°	99.7	0.3	0

Conditions: 1.00 g 70-325 ultrasonically washed basalt, 1.00 ml of distilled water, 7.00 mL GR-2, Tc-95 m tracer only, about 42 hours of equilibration at 60°C. Various orders of addition of tracer and water were tried but it seemed not to matter in the results.

under the proper conditions, even air ground basalt can adsorb technetium if the surface conditions are right and if enough time is allowed.

Heat Treated Basalt

In Table 3, results are shown for basalt which had been heat-treated in hydrogen at temperatures from 175 to 500°C. Significant adsorption of technetium is not seen until the temperature is raised to nearly 300°C. At 500°C, almost all of the technetium was removed after about two days equilibration.

In Table 4, results are shown for basalt which had been heat-treated in argon. Again, substantial removal of technetium is indicated, but the removal did not approach 100% until the temperature was raised to 700°C.

The specific reason for the effect of the various forms of heat treatment is not known. The hydrogen treatment is capable of reducing Fe(III) to either Fe(II) or Fe(0). However, the vacuum and argon heat treatments are unlikely to produce the zero oxidation state. One likely possibility concerns the formation of a defect structure in iron oxides by the heat. The traditional oxides of compositions implied by formulas such as FeO, Fe₃O₄, and Fe₂O₃ do not generally exist with those precise atomic ratios in high temperature phases. Defects, structures and non-stoichiometric materials are the rule, not the exception. Oxygen loss by high temperature treatment could produce room temperature reducing surfaces. Probably, the oxygen partial pressure, considering both the presence and absence of reducing gases will be an important part of the final answer.

One condition that had to be controlled carefully was exposure of the basalt to oxygen. On perhaps six separate occasions, heat treated basalt was deliberately exposed to air or the samples were sealed in air after heat treatment. This was done to determine its effect on the amount of technetium adsorbed. When the heat treated samples were sealed in air, the amount of adsorption was invariably reduced to nearly zero. This was true even for the hydrogen-treated basalt that otherwise would have adsorbed at least 95% of the technetium. Several experiments were run to see how much abuse the treated basalt could really take. In these tests, the basalt was

Table 4
Argon Heat Treated Basalt

Temperature	Tc Initial (M)	% Tc Remaining	% Tc Adsorbed	Rs (mL/g)
First Set:				
500°	Tracer	27.9	72.1	20.8
500°	10 ⁻⁷	40.3	57.9	12.1
Second Set:				
400°	10 ⁻⁸	52.3	47.7	7.5
500°	10 ⁻⁸	38.7	61.3	13.0
600°	10 ⁻⁸	47.5	52.5	9.1
600°	10 ⁻⁸	51.0	49.0	7.9
700°	10 ⁻⁸	4.5	95.5	183

Conditions: 1.00 g Basalt, 8.00 mL GR-2, Tc-95m tracer, initial concentration of Tc-99 added, about 42 hours equilibration at 60°. All treatments in Pyrex except for 600 and 700° runs which were in quartz.

Table 5
Oxygen Treatment of H₂-Reduced Basalt

Sample Description	% Tc Removed	Rs (ml/g)
Normal 500° H ₂ Treatment	97 - 93	262 - 100
Shake sample in air with 1 mL GR-2 for 7 minutes followed by freeze-pump-thaw cycles	82.4	17.6
Seal sample in air	0.3	0

Conditions: 1.00 g 70-325 ultrasonic cleaned basalt, 500° H₂-treated basalt, cooled in H₂, then commence air test. Tc-95m tracer only, 60° equilibration, 42 hours.

subjected to both partial oxidation (7 minutes shaking in air-saturated water before subsequent vacuum degassing) and total oxidation (sample sealed in air). As expected, the latter material adsorbed no significant quantities of technetium and the former adsorbed only 83%. These data are shown in Table 5. The conclusion is that the basalt surface was moderately fragile. This was also true for the inert gas ground material discussed earlier.

The kinetics of technetium removal by the basalt is obviously of interest. The data from Table 6 shows adsorption at times of 5, 17, 42 and 117 hours. It can be seen that there is little difference between the various times. The adsorption must be fairly fast, and it is logical to assume that the initial quantities go on the surface at the fastest rate. The typical run time of about 42 hours chosen for most of our experiments was selected to provide a safe margin in case there was some wide variation in the rates. As discussed earlier, the adsorption of technetium on the untreated basalt is much slower.

The amount of technetium adsorbed as a function of the initial technetium concentration is obviously important. The Tc-95m tracer is about 10^{-12} M. A series of Tc-99 solutions were made having concentrations of 4×10^{-3} , 10^{-4} , 10^{-5} , 10^{-6} , 10^{-8} , and 10^{-10} M. Typical data are shown in Table 7. It can be seen that 95% of the technetium is removed by the normal 70-235 ultrasonically washed basalt until the 10^{-4} M solution concentration is reached. Beyond that point, the amount removed is much less. The most plausible explanation is that the reduction capacity of the surface had been reached. However, when a sample of the "fines" (less than 325 mesh) was used, a considerably greater amount of technetium was adsorbed. In this case, there was more surface and thus more reduction capacity.

Over the broad range of concentrations used, it appears that if the surface has any reduction capacity, it will adsorb technetium even at the lowest tracer levels. These data appear not to tell much about the mechanism or the oxidation state of the technetium either in the solution or on the solid.

Table 6
Rate of Technetium Uptake with Time

Time	% Remaining	% Removed	Rs (mL/g)
5.2 hrs.	3.6	96.4	222
14.7 hrs.	2.1	97.9	382
42 - 46 hrs.*	3-7	97-93	262 - 100
117	2.9	97.1	272

*Typical data, not run as a part of this set of experiments.
Conditions: 1.00 g 70-325 ultrasonically washed basalt, 8.00 mL GR-2, Tc-95m tracer, 10^{-8} Tc-99 initially, 500°C H₂-reduced, equilibrated at 60°.

Table 7
Adsorption of Technetium with Variable Quantities of Added Tc-99

Tc-99 Conc. Before Adsorption (m)	% Tc Remaining	% Tc Removed	Rs (mL/g)
Tracer	3-7%	97-93	101-262
10 ⁻¹⁰	4.0	96.0	195
10 ⁻⁸	8.3	91.7	90
10 ⁻⁶	2.5	97.5	312
10 ⁻⁵	9.6	90.4	77
10 ⁻⁴	35.2	64.8	15.0
10 ⁻⁴	20.7	79.3	31.5
4 x 10 ⁻³	88.2	11.8	1.10
None*	2.6	97.4	301
10 ⁻⁴ *	0.8	99.2	996
4 x 10 ⁻³ *	74.2	25.8	2.85

*Basalt is < 325 mesh.

Conditions: 1.000 g 70-325 ultrasonically washed basalt except for the last three samples in table, Tc-95m tracer and variable quantities of Tc-99; 500°C H₂-treatment; equilibration for about 42 hours at 60°C. GR-2.

Desorption

The desorption of technetium from the solid is a subject of considerable concern to adsorption studies. Time did not permit a systematic investigation, but some qualitative observations were made. Desorption experiments were attempted with GR-2, NaCl solutions, 1 M HCl, and H₂SO₄, all in air. There appeared to be a fairly rapid desorption of a small fraction of the technetium which was estimated to be about a quarter of the material. After that, the desorption rate became very slow. However, the addition of strong oxidizing agents resulted in rapid removal of Tc. In these studies, KMnO₄ in sulfuric acid and H₂O₂ in 0.1 M NaCl were tried.

Technetium Speciation

The nature of the technetium that remains in solution is an interesting but unresolved mystery. At these tracer levels, there are not too many methods available to determine the oxidation state of the element. It has been shown that soluble TcO₄⁻ can be extracted almost quantitatively into a chloroform solution of tetraphenylarsonium chloride. This has been reconfirmed by a number of us working on this project. It is also possible that reduced technetium species will not extract in this system. An extensive but not yet exhaustive search has not turned up literature confirmation of the latter assumption. The arsonium cation forms an ion pair with large anions like ReO₄⁻, TcO₄⁻ and ClO₄⁻. These can either precipitate from aqueous solution or be extracted into chloroform. The question is, can any lower valent technetium anionic species be extracted as an arsonium salt? If one includes the realm of organic complexes, the answer is absolutely yes. However, if one restricts the consideration to simple ground water anions such as chloride, fluoride, carbonate, sulfate, silicates, and more complex species, then the answer may well be no. It is particularly likely that such species may not exist at the neutral or slightly basic pH conditions of ground water. There is simply not much known about the solution species present with these anions. Species like TcCl₆²⁻ which exist at high HCl concentration would presumably hydrolyze in such media but it is not known how fast.

Tetraphenylarsonium extractions were done for many of the runs. A pattern was very quickly established. If more than half of the technetium remained in solution, it was quantitatively extracted into the chloroform layer presumably as TcO_4^- . However, when only 5 to 30% of the technetium was left after equilibration, a considerably lower fraction of the technetium was extracted into the chloroform. An unusual observation was that the counts not found in chloroform were also not found in the aqueous layer. When the residue was counted (which contained a small amount of both aqueous and organic material plus the interfacial boundary), the missing technetium was found, and there was a fairly good material balance. There appeared to be a scum or precipitate in the interfacial zone. An example of the data is shown in Table 8. In Table 9 is given an example of percent extraction as a function of time for a representative sample.

It can be concluded from the discussion above, that something present in either the basalt or the ground water stabilized a solution species of technetium in some lower oxidation state. Alternately, it is possible that the material in solution was a colloid. On the assumption that one of the synthetic groundwater anions was responsible, a series of tests were run using the 500° hydrogen-reduced basalt and various 0.1 M salt solutions. The data in Table 10 show almost complete removal of technetium. It may be that the basalt contained the complexing species.

4. RESULTS AND CONCLUSIONS

Basalt can be altered by a number of means to yield a material that has a good adsorption capacity for technetium. Heat treatment is one. If the basalt is heated to temperatures of about 500° under a number of conditions, a material is created that can remove up to 98% of the pertechnetate ion from solution. An atmosphere of hydrogen gas is the most effective in producing this material, argon is next, followed by vacuum. Heat treatment in air is ineffective. In addition, the basalt that had been made active by any technique becomes inactive after exposure to air at room temperature. Grinding basalt in an inert gas glove box having an oxygen content of perhaps < 10 ppm will also yield a material that adsorbs technetium. Clearly, rigorous oxygen exclusion is essential for this work.

Table 8
Typical Chloroform Extraction

	<u>Counts</u>
Initial Sample	10088
Chloroform Layer 0.4 ml of 0.5 mL	4018
Aqueous Layer 0.8 ml of 1.0 mL	1374
Interfacial Residue	<u>4686</u> 10078

$$\% \text{ extraction } = \frac{4018 \times 1.25}{10088} \times 100 = 49.8$$

Residue count should be
 $4018 \times .25 + 1374 \times .25 = 1348$

This particular solution was in the
air for 48 hours before extraction.

Table 9
Technetium Extraction Versus Time

Date	% in CHCl_3
6/22/83	25.6%
6/24/83	49.8
6/27/83	59.5
7/18/83	60.0

Conditions: 1 mL fractions of sample 42 that originally had 91.7% of the technetium removed from solution. These fractions were taken from the basalt 6/22/83 and allowed to stand in contact with air. Sample originally had 10^{-8} Tc-99 and Tc-95 m tracer.

Table 10
Technetium Adsorption with Various Salt Solutions

Solute	% Remaining	% Removed	Rs (mL/g)
KCl	1.2	98.8	633
K_2SO_4	2.4	97.6	322
NaHCO_3	3.6	96.4	217
NaF	2.2	97.8	367

Conditions: 1.00 g 70-325 ultrasonic washed basalt, 500°H_2 treatment, Tc-95m tracer only, 0.1 M solutions, about 42 hours equilibration at 60°C . Of the technetium remaining in solution between 32 and 60% could be extracted in chloroform solutions of tetraphenylarsonium chloride. The extraction data was of low accuracy because there was little technetium in solution.

Air-ground basalt, even a material having an extremely high surface area, does not adsorb significant quantities of technetium in the 24 to 48 hour time periods used for the experiments discussed above. However, that same high surface area material absorbs 10 to 15% of the technetium in 19-day experiments and samples that were shaken for about three months adsorbed 60 to 80% of the technetium. This adsorption occurs at orders of magnitude slower rates than the previously discussed results.

There is no specific information available from these experiments that gives any details about the mechanism of the removal of the soluble pertechnetate species. However, everything is consistent with the general hypothesis that reduction to insoluble technetium dioxide is responsible.

Acknowledgements

The assistance of J. S. Johnson, Jr., C. G. Westmoreland, W. D. Arnold, F. I. Case, and R. E. Meyer is gratefully acknowledged.

References

1. For instance, E. A. Bondietti and C. W. Frances, Science **203**, 1337 (1979), claim extensive absorption of technetium on basalt whereas separate investigations by J. S. Johnson, Jr., and R. E. Meyer (in ORNL progress reports) show limited adsorption in equilibration periods of a few days.
2. R. E. Meyer, R. A. Posey and P. M. Lantz, Desalination **11**, 329 (1972).
3. P. F. Salter, L. L. Ames and J. E. McGarrah, RHO-BWI-LD-48 (1981).