Pacific Northwest
NATIONAL LABORATORY

PNNL-29445

FY2019 Radionuclide Migration Tests

November 2019

EA Cordova
EC Golovich

U.S. DEPARTMENT OF ENERGY

Prepared for the U.S. Department of Energy
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Summary

The Waste Management Project provides safe, compliant, and cost-effective waste management services for the Hanford Site and the U.S. Department of Energy (DOE) complex. Part of these services includes safe disposal of low-level waste and mixed low-level waste at the Hanford Low-Level Waste Burial Grounds in accordance with the requirements of DOE Order 435.1, Radioactive Waste Management. To partially satisfy these requirements, performance assessment analyses were completed and approved. DOE Order 435.1 also requires continuing data collection to increase confidence in the critical assumptions used in these analyses to characterize the operational features of the disposal facility that are relied on to satisfy the performance objectives identified in the order.

Cement-based solidification and stabilization is considered for hazardous waste disposal because it is easily done and cost-efficient. One critical assumption is that concrete will be used as a waste form or container material at the Hanford Site to control and minimize the release of radionuclide constituents in waste into the surrounding environment. Concrete encasement would contain and isolate the waste packages from the hydrologic environment and act as an intrusion barrier. Any failure of concrete encasement may result in water intrusion and consequent mobilization of radionuclides from the waste packages. The radionuclides iodine-129, selenium-79, technetium-99 (Tc-99), and uranium-238 have been identified as long-term dose contributors.[1,2] Because of their anionic nature in aqueous solutions, these constituents of potential concern may be released from the encased concrete by mass flow and/or diffusion and migrate into the surrounding subsurface environment.[3,4,5,6,7] Therefore, it is necessary to assess the performance of the concrete encasement structure and the ability of the surrounding soil to retard radionuclide migration.

This report presents results from a set of sorption experiments completed in fiscal year (FY) 2019 to evaluate partition coefficients for iodine (I) and Tc-99 using intact concrete monoliths. Surface complexation, ion exchange, and potential precipitation processes are all included in the partition coefficient parameter. There were several changes to the experimental conditions from the set completed in FY 2018. These changes included 1) using a solution composition more representative of the Hanford site groundwater instead of a saturated calcium hydroxide [$Ca(OH)_2$] solution, 2) increase of I and Tc-99 concentrations, and 3) reduction of the number of test durations to two (one month and three months) due to time constraints.

Partition coefficients (K_d) calculated for iodine range from 19.51 mL/g for large monoliths with a one-month test duration to 52.70 mL/g for small monoliths with a three-month test duration. The partition coefficients were approximately three times larger than those calculated as part of the FY 2018 testing. The increase in partition coefficients is most likely a result of the change in

[1] Mann, FM, RJ Puigh II, SH Finfrock, J Freeman, E.J., R Khaleel, DH Bacon, MP Bergeron, PB McGrail, and SK Wurstner. 2001. *Hanford Immobilized Low-Activity Waste Performance Assessment: 2001 Version*. Pacific Northwest National Laboratory, Richland, WA.
[2] Wood, MI, R Khaleel, PD Rittman, AH Lu, S Finfrock, RJ Serne, and KJ Cantrell. 1995. *Performance Assessment for the Disposal of Low-Level Waste in the 218-W-5 Burial Ground*. Westinghouse Hanford Company, Richland, WA.
[3] Serne, RJ, JL Conca, VL LeGore, KJ Cantrell, CW Lindenmeier, JA Campbell, JE Amonette, MI Wood. 1993. *Solid-Waste Leach Characterization and Contaminant-Sediment Interactions*. Pacific Northwest Laboratory, Richland, WA.
[4] Serne, RJ, LL Ames, PFC Martin, VL LeGore, CW Lindenmeier, and SJ Phillips. 1993. *Leach Testing of in Situ Stabilization Grouts Containing Additives to Sequester Contaminants*. Pacific Northwest Laboratory, Richland, WA.
[5] Serne, RJ, RO Lokken, and LJ Criscenti. 1992. “Characterization of Grouted LLW to Support Performance Assessment.” *Waste Management* 12:271-87.
[6] Serne, RJ, WJ Martin, and VL LeGore. 1995. *Leach Test of Cladding Removal Waste Grout Using Hanford Groundwater*. Pacific Northwest Laboratory, Richland, WA.
[7] Serne, RJ, WJ Martin, VL LeGore, CW Lindenmeier, SB McLaurine, PFC Martin, and RO Lokken. 1989. *Leach Tests on Grouts Made with Actual and Trace Metal-Spiked Synthetic Phosphate/Sulfate Waste*. Pacific Northwest Laboratory, Richland, WA.

composition used for the starting solutions. Studies about the Hanford Site have shown that iodine species co-precipitate with calcium carbonate.[1] The alkalinity values measured within the starting solutions are large and could help to preferentially co-precipitate the iodine within the solutions and cause the observed increased K_d values.

Additional analysis is needed to confirm the cause for the observed increase in K_d values for iodine and to obtain reliable data for Tc-99 for calculations. A set of experiments is planned for FY 2020 to repeat the studies using the $Ca(OH)_2$ and the modified groundwater solutions. A more detailed surface analysis (i.e., scanning electron microscopy) will be completed to better understand the changes in surface interactions in addition to a more complete analysis to evaluate pH and alkalinity throughout the test durations.

[1] Truex M.J., B.D. Lee, C.D. Johnson, N. Qafoku, J.E. Szecsody, J.E. Kyle, and M.M. Tfaily, et al. 2017. *Conceptual Model of Iodine Behavior in the Subsurface at the Hanford Site*. PNNL-24709 Rev. 2. Richland, WA: Pacific Northwest National Laboratory.

Acknowledgments

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Acronyms and Abbreviations

ASTM	American Society for Testing and Materials
COPC	constituents of potential concern
DDI	distilled deionized
DOE	U.S. Department of Energy
FY	fiscal year
IC	ion chromatography
ICP-MS	inductively coupled plasma-mass spectrometry
ICP-OES	inductively coupled plasma-optical emission spectrometry
LLBG	Hanford Low-Level Waste Burial Grounds
PVC	polyvinylchloride

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1.0 Introduction

The Waste Management Project provides safe, compliant, and cost-effective waste management services for the Hanford Site and the U.S. Department of Energy (DOE) complex. Part of these services includes safe disposal of low-level waste and mixed low-level waste at the Hanford Low-Level Waste Burial Grounds (LLBG) in accordance with the requirements of DOE Order 435.1, Radioactive Waste Management. To partially satisfy these requirements, performance assessment analyses were completed and approved. DOE Order 435.1 also requires continuing data collection to increase confidence in the critical assumptions used in these analyses to characterize the operational features of the disposal facility that are relied on to satisfy the performance objectives identified in the order.

Cement-based solidification and stabilization is considered for hazardous waste disposal because it is easily done and cost-efficient. One critical assumption is that concrete will be used as a waste form or container material at the Hanford Site to control and minimize the release of radionuclide constituents in waste into the surrounding environment. Concrete encasement would contain and isolate the waste packages from the hydrologic environment and act as an intrusion barrier. Any failure of concrete encasement may result in water intrusion and consequent mobilization of radionuclides from the waste packages. The radionuclides iodine-129 (I-129), selenium-79 (Se-79), technetium-99 (Tc-99), and uranium-238 (U-238) have been identified as long-term dose contributors (Mann et al. 2001; Wood et al. 1995). Because of their anionic nature in aqueous solutions, these constituents of potential concern (COPCs) may be released from the encased concrete by mass flow and/or diffusion and migrate into the surrounding subsurface environment (Serne et al. 1989; 1992; 1993a, b; 1995). Therefore, it is necessary to assess the performance of the concrete encasement structure and the ability of the surrounding soil to retard radionuclide migration. A critical component of this is to provide (1) quantitative estimates of radionuclide retention within concrete waste form materials (source term) similar to those used to encapsulate waste in the LLBG, and (2) provide a measure of the effect of concrete waste form properties on radionuclide release and migration within the near-field environment.

This report presents results from a set of sorption experiments completed in fiscal year (FY) 2019 to evaluate partition coefficients for iodine (I) and Tc-99 using intact concrete monoliths. Surface complexation, ion exchange, and potential precipitation processes are all included in the partition coefficient parameter. There were several changes to the experimental conditions from the experiments completed in FY 2018. These changes include 1) using a solution composition more representative of the Hanford Site groundwater instead of a saturated calcium hydroxide solution, 2) an increase of the I and Tc-99 starting concentrations, and 3) reduction of the number of test durations to two (one month and three months) due to time constraints.

2.0 Materials and Methods

2.1 Concrete Composition

The concrete composition for the burial encasement was specified in *Specification for Concrete Encasement for Contact-Handled Category 3 Waste* (HNF-1981). This specification was used as the basis to prepare a concrete for fabrication of test specimens. The composition includes sulfate-resistant Portland Type I or Type II cement, a pozzolanic material (Class F fly ash), fine and coarse aggregates, and steel fiber. Additional specifications include a water-to-cement ratio of 0.4 and an air content of 6.0 ± 1.5%. The nominal proportions and material specifications based on this initial design are listed in Table 2.1.

Table 2.1. Concrete material specifications and composition

Material	Specifications	Specified Field Mix	Normalized Specification Design
Cement	Portland Type I or Type I/II sulfate-resistant cement	381 kg/m^3	0.27
Fly Ash	Class F fly ash; nominal 15% of cement by volume	54 kg/m^3	0.04
Coarse Aggregate	No. 676 or equivalent (3/4 in. nominal size)	55% by volume	0.04
Fine Aggregate	Sand	45% by volume	0.51
Water	Nominal water-to-cement ratio: 0.4	399 kg/m^3	0.10
Steel Fiber	Deformed Type I, nominal length 2.5 to 3.8 cm (1 to1.5 in.)	59 kg/m^3	0.04
Air Content		6.0±1.5%	

2.2 Materials and Laboratory-Scale Mixture Design

The laboratory-scale concrete mixtures (Table 2.2) were prepared based on specifications shown in Table 2.1. Due to the required small dimensions of the laboratory test specimens, the coarse aggregate was omitted, and 40 to 60 mesh sized sand was used instead. Based on these modifications, a concrete mix was prepared that consisted of Portland cement (Type I/II sulfate resistant, ASTM C-150 compliant cement); Class F fly ash, sand, and a water-entraining agent (PolyHeed 997). A water-entraining agent was included in the mix to facilitate the workability of the concrete. The volumes of the PolyHeed 997 were not included in the normalization calculations because of their negligible contribution to the overall mix volume. The material specification and composition for the laboratory-scale concrete mixture is given in Table 2.2.

Table 2.2. Laboratory-scale material specification and composition

Material	Material Specifications for Field Mix	Normalized Laboratory Design	Material Specifications Used in Revised Laboratory Mix Comparison
Cement	Portland Type I or Type I/II sulfate-resistant cement	0.27	Portland Type I/II
Fly Ash	Class F fly ash nominal 15% of cement by volume	0.04	Class F fly ash; nominal 20% of cement by volume
Fine Aggregate	Sand	0.51	Industrial quartz Accusand 40 to 60 mesh (0.420 to 0.250 mm)
Water	Nominal water-to-cement ratio: 0.4	0.10	Water-to-cement ratio: 0.5
Steel Fiber	Deformed Type I nominal length 2.5 to 3.8 cm (1 to 1.5 in.)	0.04	Iron powder 40 to 60 mesh [(a)] (0.149 to 0.177 mm)
PolyHeed 997	--	0.00375	Water-entraining agent

(a) Historically, iron was included in the monolith materials, so it remains included within this table. No iron was included in monoliths used for the current set of sorption experiments.

2.3 Concrete Mix and Specimen Preparation

Concrete monoliths were prepared by mixing the dry ingredients (sand, fly ash, and cement), adding the PolyHeed 997 and water, and mixing. The concrete was mixed with a whisk in a steel bowl for 3 to 5 minutes prior to pouring into molds.

The molds for casting concrete specimens were fabricated from Fisher brand poly sample vials with a hinged cap. After filling, the molds were lightly tapped on the laboratory bench and vibrated using a handheld vibrator until a significant decrease in the release of air bubbles was observed. The forms were stored in a humidity chamber for 28 days while the concrete set. Following the curing period, monoliths were set to soak in a saturated calcium hydroxide [$Ca(OH)_2$] solution (pH = 12.33), representing a simple cement pore water composition, for 30 days.

A series of batch sorption tests were conducted to determine the partition coefficient (K_d) for concrete monoliths using iodine (I) and Tc-99 solutions. The test matrix included three sizes of monoliths with different surface areas, three concentrations for I and Tc-99, and two test durations as shown in Table 2.3. The test solution concentrations were prepared using a saturated $Ca(OH)_2$ solution to which several groundwater constituents representative of groundwater from the Hanford site were introduced. Although the solution was significantly changed, additional carbonate input was avoided. Table 2.4 lists the groundwater constituents used for the test solutions prior to addition of I or Tc-99. A total of 54 different test conditions, shown in Table 2.3, were run in duplicate. The 108 individual tests were initiated on the same day, so that each set of test conditions was only sampled once and maintained a constant solution volume throughout the duration of the experiment.

Table 2.3. Batch sorption test variables

Variable	Conditions
Species	Tc-99 or I
Concentrations (μg/mL)	$1x10^{-3}$, $5x10^{-3}$, and $1x10^{-2}$
Diameter [(a)] (cm)	2.2 (small), 3.3 (medium), and 4.3 (large)
Length (cm)	4.0 (small), 4.2 (medium), and 4.3 (large)
Duration (months)	1 and 3

(a) Diameter is an average calculated from measurements at the top and bottom of the cylinder.

Table 2.4. Modified groundwater composition

Constituent [(a)]	Concentration (g/L)
$Ca(OH)_2$	1.038
H_4SiO_4	0.0153
KCl	0.0082
NaCl	0.015
$CaSO_4$	0.067
pH	12.34

(a) The recipe was modified from artificial groundwater recipe described as reagent 1 in Truex et al., 2017a.

3.0 Sorption Experiments

A set of sorption experiments was initiated in FY2019 to evaluate partition coefficients (K_d) for I and Tc-99 using intact concrete monoliths. The test matrix included three sizes of monoliths resulting in three different surface areas, three solution concentrations (1, 5, and 10 μg/L), and two test durations (1 month and 3 months). Monolith composition is described in Section 2.1. Test variables are described in Table 2.3.

3.1 Iodine Sorption

Specific test conditions including monolith characteristics, starting and final concentrations, and calculations of iodine sorbed are provided in Appendix Section A.1. Separate tables contain details for tests using small monoliths (Table A.1), medium monoliths (Table A.2), and large monoliths (Table A.3). Figure 3.1 shows the iodine sorbed to the monoliths as a function of the final iodine concentrations for all test conditions. The remaining figures in this section show the data based on the various test conditions, such as monolith size (Figure 3.2) or test duration (Figure 3.3). Figure 3.4 and Figure 3.5 fit the data based on both monolith size and test duration.

A summary of the partition coefficients calculated for iodine is given in Table 3.1. Partition coefficients calculated for iodine range from 19.51 mL/g for large monoliths with a one-month test duration to 52.70 mL/g for small monoliths with a three-month test duration. The partition coefficients were approximately three times larger than those calculated as part of the FY 2018 testing.

The increase in partition coefficients is most likely a result of the change in composition used for the starting solutions. In FY 2018, the starting solution was a saturated $Ca(OH)_2$ solution spiked with iodine. This year, we included additional chemical species into the starting solution as described in Table 2.4. Additional analyses were completed on the test solutions and potential effects are discussed in Section 3.3.

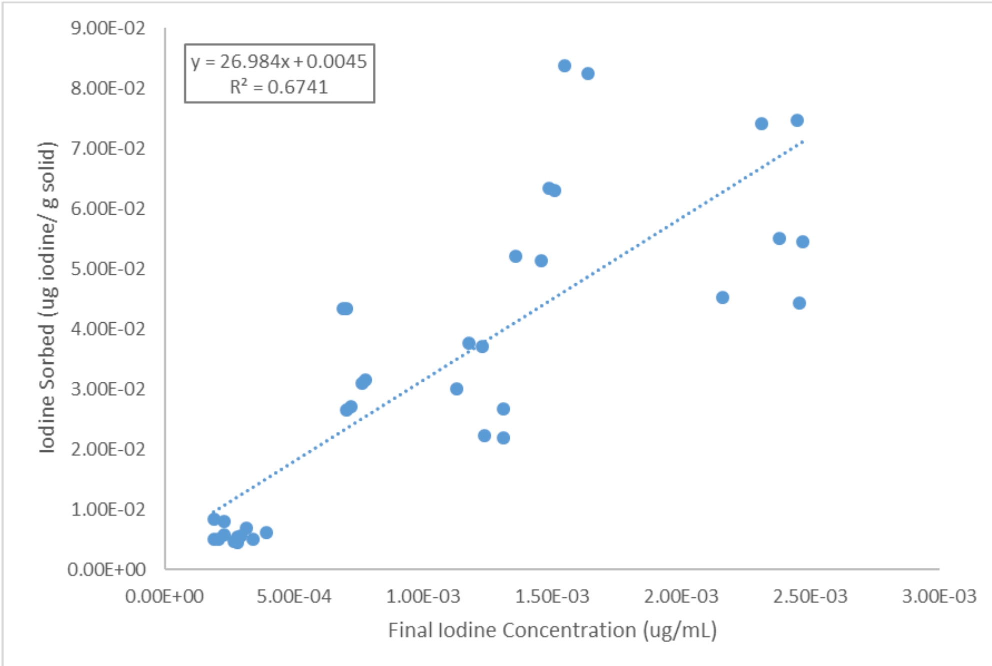

Figure 3.1. Calculated sorbed iodine as a function of final iodine concentrations for all test conditions.

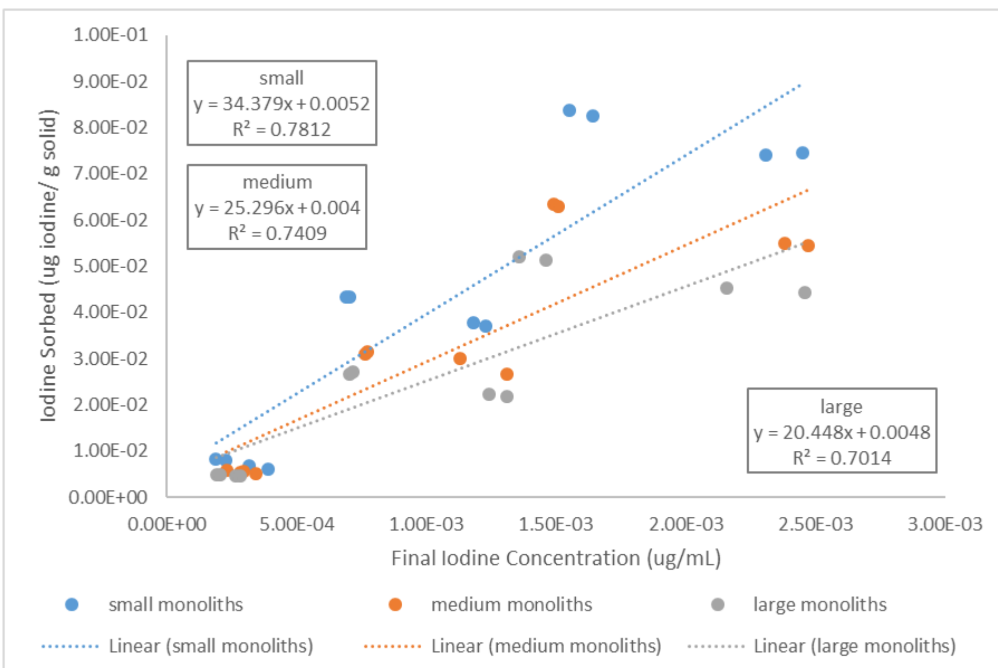

Figure 3.2. Calculated sorbed iodine as a function of final iodine concentration with both test durations shown with linear fits for small, medium, and large monoliths.

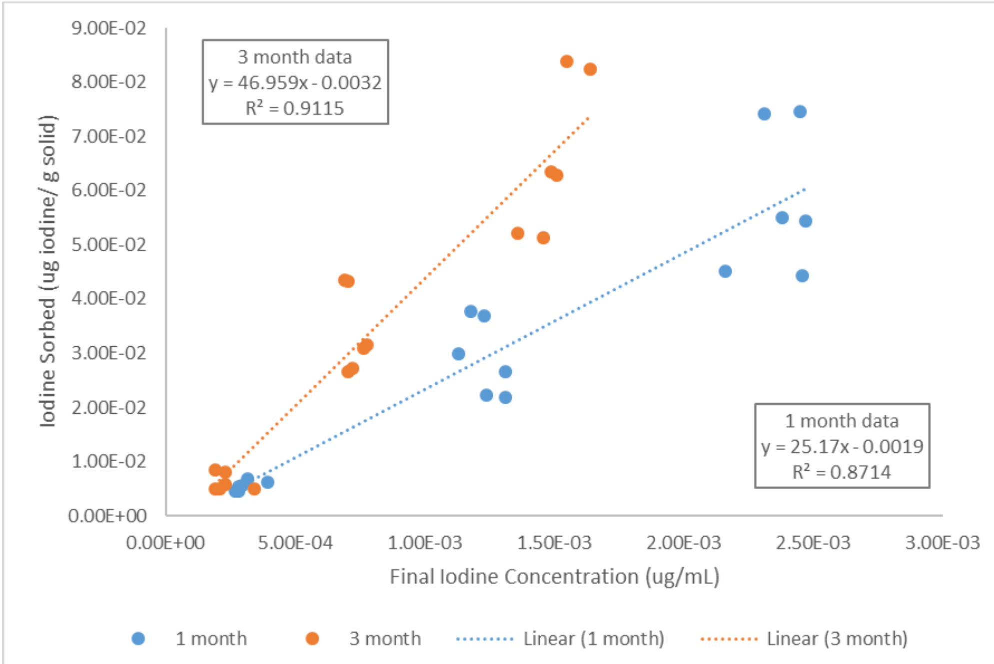

Figure 3.3. Calculated sorbed iodine as a function of final iodine concentration with all monolith sizes shown with linear fits for 1-month, and 3-month test durations.

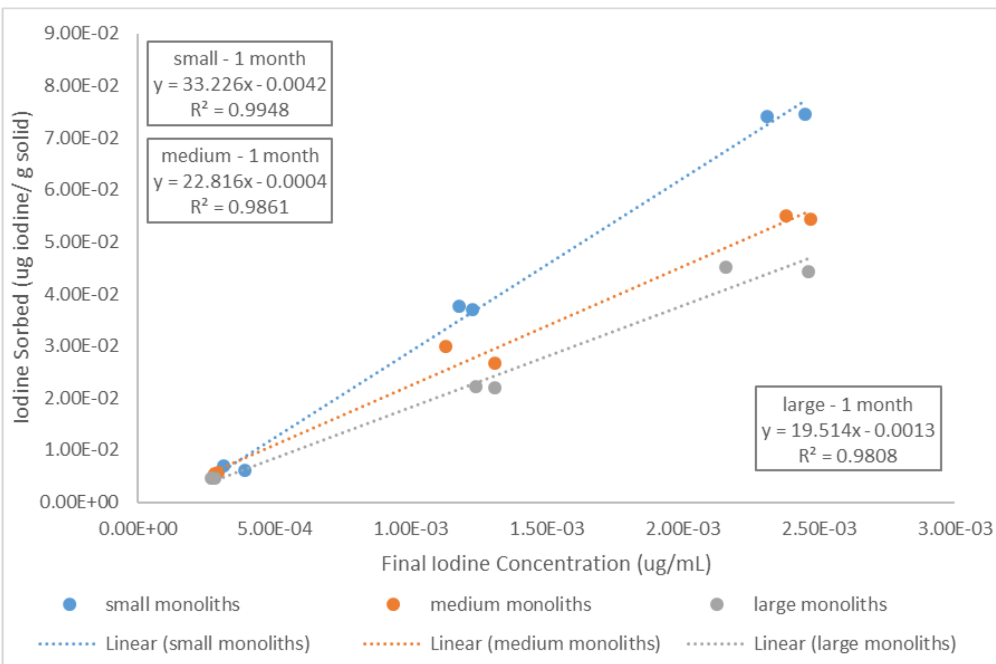

Figure 3.4. Calculated sorbed iodine as a function of final iodine concentration with 1-month test durations shown with linear fits for small, medium, and large monoliths.

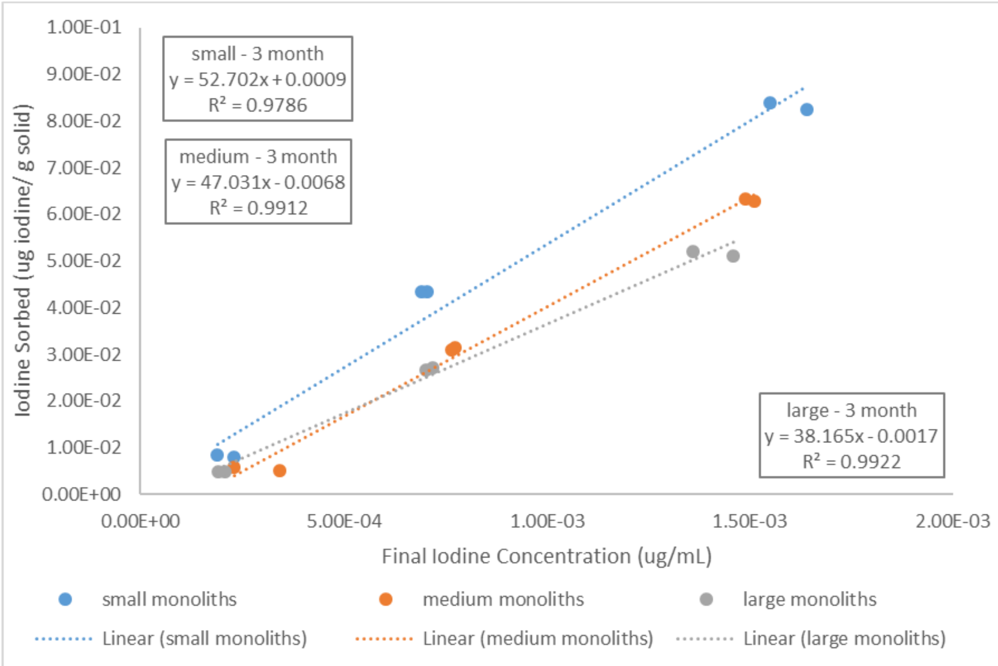

Figure 3.5. Calculated sorbed iodine as a function of final iodine concentration with 3-month test durations shown with linear fits for small, medium, and large monoliths.

Table 3.1. Summary of Calculated Iodine Partition Coefficients

Iodine Test Conditions	K_d (mL/g)
All Monolith Sizes	
1 Month	25.17
3 Month	46.96
1-Month Test Duration	
Small Monoliths	33.23
Medium Monoliths	22.82
Large Monoliths	19.51
3-Month Test Duration	
Small Monoliths	52.70
Medium Monoliths	47.03
Large Monoliths	38.17

3.2 Tc-99 Sorption

Specific test conditions including monolith characteristics, starting and final concentrations, and calculations of sorbed Tc-99 are provided in Appendix Section A.2. Separate tables contain details for tests using small monoliths (Table A.4), medium monoliths (Table A.5), and large monoliths (Table A.6). Figure 3.6 and Figure 3.7 show the Tc-99 sorbed to the monoliths by size as a function of the final Tc-99 concentrations for the 1-month and 3-month test durations respectively. The values quantified for many of the test conditions, especially those with the mid-range starting concentration of Tc-99 and both test durations, are suspect. For 12 tests, final concentrations were larger than initial concentrations resulting in the calculation of negative amounts of Tc-99 sorbed to the monoliths. In these cases, the data was not used for the linear fits to calculate the K_d values. For the 3-month test duration experiments, there were not enough positive data values to fit, therefore no linear fits (K_d values) were calculated.

A summary of the partition coefficients calculated for Tc-99 is given in Table 3.2. The partition coefficients that were able to be calculated for Tc-99 were similar to those observed in the FY 2018 experiments. Additional analyses were completed on the test solutions and differences in observed effects compared to iodine are discussed further in Section 3.3.

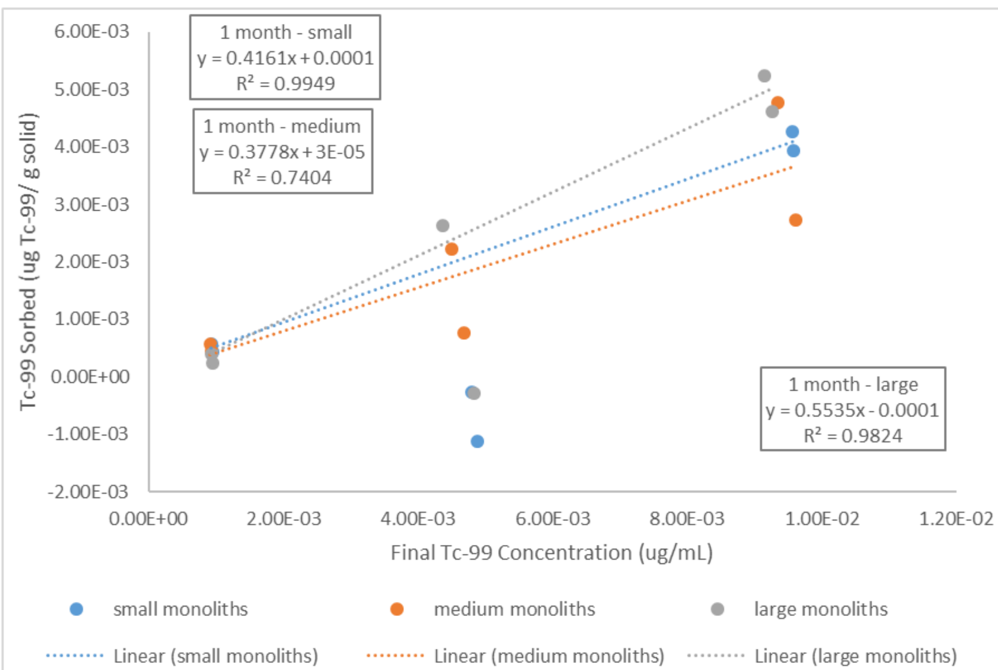

Figure 3.6. Calculated sorbed Tc-99 as a function of final Tc-99 concentration with 1-month test durations shown with linear fits for small, medium, and large monoliths.

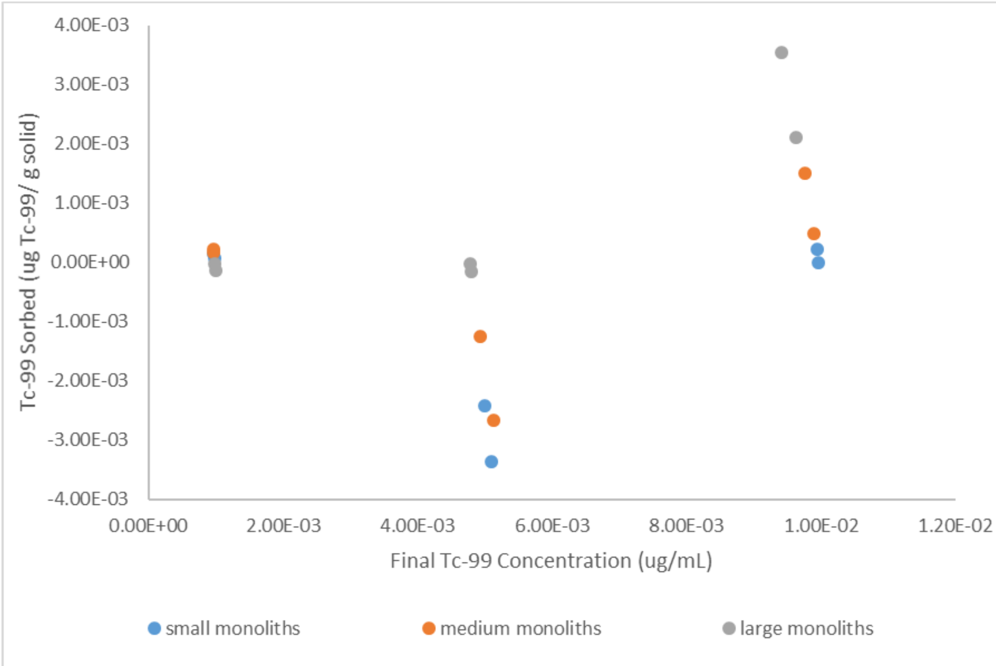

Figure 3.7. Calculated sorbed Tc-99 as a function of final Tc-99 concentration with 3-month test durations shown with linear fits for small, medium, and large monoliths.

Table 3.2. Summary of Calculated Tc-99 Partition Coefficients

Tc-99 Test Conditions	K_d (mL/g)
1 Month Test Duration	
Small Monoliths	0.4161
Medium Monoliths	0.3778
Large Monoliths	0.5535

3.3 Additional Analysis

The test solution used in FY 2018 experiments was a saturated $Ca(OH)_2$ solution spiked with I or Tc-99. The solution used in the FY 2019 tests was a modified groundwater composition as described in Table 2.4 adding more chemical species and creating more interactions within the solutions and with the monolith surface. A comparison of the pH, ionic strength, and alkalinity of the two base starting solutions (prior to I or Tc-99 spike) is shown in Table 3.3. The ionic strength for the saturated $Ca(OH)_2$ solution was slightly lower and the alkalinity was higher than the modified groundwater solution. An evaluation of the change of pH and alkalinity from the starting solution to the end solution had not been completed during testing this year, but is planned as part of the FY 2020 testing scope to see how this may have impacted overall surface interactions.

Table 3.3. Summary of Solution Characteristics from FY 2018 and FY 2019

	FY 2018	FY 2019
pH	12.33	12.35
Ionic Strength (M)	0.042	0.051
Alkalinity (μg/mL)	1400	1280

Additional analysis was completed on the test solutions by inductively coupled plasma - optical emission spectrometry (ICP-OES) and ion chromatography (IC) to evaluate cation and anion concentrations to identify other possible interactions that may have caused the increase in I K_d values. Results for calcium, magnesium, potassium, and sodium cation concentrations are presented in Appendix A in Table A.7 and Table A.8 for the I and Tc-99 tests respectively. Appendix A also contains concentration tables for chloride, nitrate, and sulfate as shown in Table A.9 and Table A.10 for the I and Tc-99 tests respectively.

Figure 3.8 shows cation (solid lines) and anion (dashed lines) concentrations plotted for a series of tests using the middle starting concentration of iodine and medium-sized monoliths for the starting solution and solutions from the end of the 1-month test duration (C-19-109) and the 3-month test duration (C-19-209). Concentrations are shown on a log scale (y-axis) to fit all species on a single plot and show the trends. Trends are similar for both I and Tc-99 tests. Calcium, potassium, and sodium all showed a slight increase in concentration over the 3 months of testing. Magnesium showed a decrease in concentration within the first month to concentrations at or below the estimated quantification limits (EQL) of the technique. Chloride and sulfate concentrations both decreased over the 3-month test duration, though sulfate had a larger change. Nitrate was not detected within the starting solution, so the EQL was used as the starting concentration. The observed increase in nitrate is believed to be due to the fly ash within the concrete.

Some particulate material was observed within the test containers, but an analysis of the material was not able to be completed. Studies about the Hanford Site have shown that iodine species co-precipitate with calcium carbonate (Truex et al., 2017b). Concentrations of calcium remain at least an order of magnitude larger than that of all other species throughout the duration of testing. The alkalinity values measured within the starting solutions are also large. This could preferentially co-precipitate the iodine within the solutions causing the observed increased K_d values.

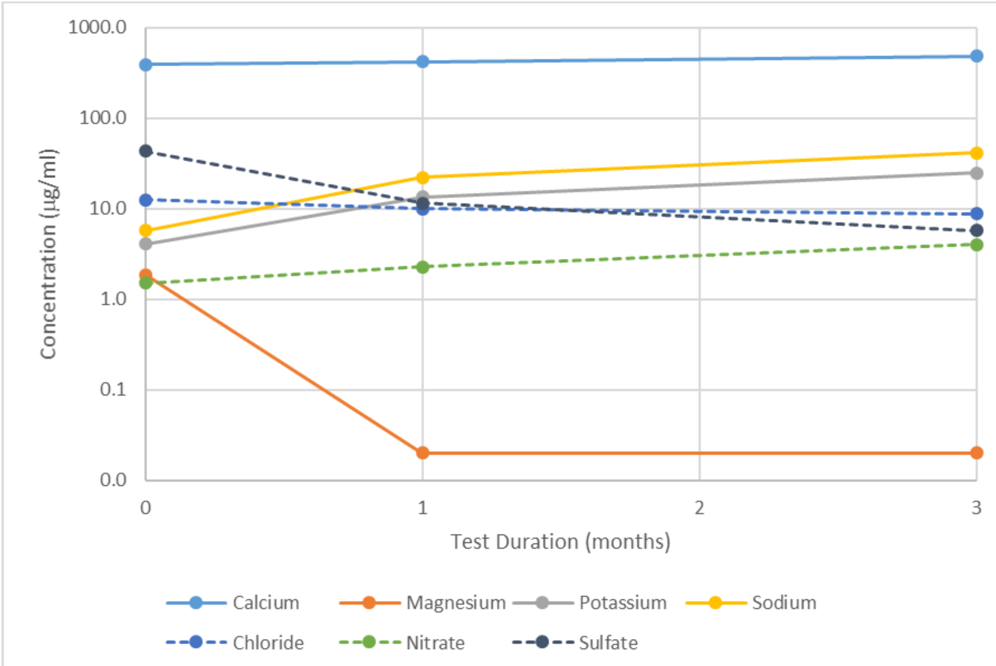

Figure 3.8. Concentrations of other ions evaluated for a test with a large monolith and the lowest concentration of iodine at starting conditions, 1-month, and 3-month test durations.

3.4 Conclusions

Additional analysis is needed to confirm the cause for the observed increase in K_d values for iodine and to obtain reliable data for Tc-99 for calculations. A set of experiments is planned for FY 2020 to repeat the studies using the $Ca(OH)_2$ and the modified groundwater solutions. A more detailed surface analysis (i.e., scanning electron microscopy) will be completed to better understand the changes in surface interactions in addition to a more complete analysis to evaluate pH and alkalinity throughout the test durations.

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Appendix A Sorption Testing Characteristics

Analytical techniques used for this project followed a process stating that instrument check measurements are within ±10% variability for standards and ±20% for duplicate runs.

A.1 Iodine Experiments

Monolith characteristics, test conditions, measured iodine concentrations and calculated values of iodine sorbed are given in the following tables for small (Table A.1), medium (Table A.2), and large (Table A.3) monoliths. The estimated quantification limit (EQL) of the analytical method used to detect iodine is 8.30E-05 μg/mL.

Table A.1. Characteristics of small-sized monoliths, concentration results, and calculated values of sorbed materials in 1-month and 3-month iodine sorption tests

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm^2)	Volume (cm^3)	Weight (g)	Density (g/cm^3)	Test Duration (months)	Solution Volume (mL)	Starting I Concentration (μg/mL)	Final I Concentration (μg/mL)	I Sorbed (μg/g solid)
C-19-101	3.870	2.207	34.46	14.79	32.096	2.170	1	344.6	9.68E-04	3.92E-04	6.18E-03
C-19-102	4.099	2.209	36.09	15.70	34.010	2.166	1	360.9	9.68E-04	3.16E-04	6.91E-03
C-19-201	3.665	2.207	33.05	14.01	30.601	2.184	3	330.5	9.68E-04	2.29E-04	7.97E-03
C-19-202	3.824	2.199	34.00	14.52	31.519	2.171	3	340.0	9.68E-04	1.89E-04	8.40E-03
C-19-107	3.956	2.209	35.09	15.15	33.403	2.205	1	350.9	4.75E-03	1.23E-03	3.70E-02
C-19-108	4.311	2.218	37.74	16.64	35.741	2.148	1	377.4	4.75E-03	1.18E-03	3.77E-02
C-19-207	4.037	2.207	35.62	15.44	33.240	2.153	3	356.2	4.75E-03	7.05E-04	4.34E-02
C-19-208	4.182	2.208	36.64	16.00	34.260	2.142	3	366.4	4.75E-03	6.93E-04	4.34E-02
C-19-113	4.147	2.210	36.44	15.89	34.520	2.172	1	364.4	9.33E-03	2.31E-03	7.41E-02
C-19-114	4.024	2.211	35.60	15.44	32.842	2.128	1	356.0	9.33E-03	2.45E-03	7.46E-02
C-19-213	3.879	2.210	34.59	14.87	32.109	2.159	3	345.9	9.33E-03	1.55E-03	8.38E-02
C-19-214	3.714	2.199	33.23	14.09	31.000	2.200	3	332.3	9.33E-03	1.64E-03	8.24E-02

Table A.2. Characteristics of medium-sized monoliths, concentration results, and calculated values of sorbed materials in 1-month and 3-month iodine sorption tests

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm^2)	Volume (cm^3)	Weight (g)	Density (g/cm^3)	Test Duration (months)	Solution Volume (mL)	Starting I Concentration (μg/mL)	Final I Concentration (μg/mL)	I Sorbed (μg/g solid)
C-19-103	4.119	3.281	59.34	34.81	75.097	2.157	1	593.4	9.68E-04	2.83E-04	5.41E-03
C-19-104	4.169	3.277	59.75	35.13	71.512	2.035	1	597.5	9.68E-04	2.95E-04	5.62E-03
C-19-203	4.179	3.274	59.79	35.16	75.850	2.157	3	597.9	9.68E-04	2.31E-04	5.81E-03
C-19-204	4.147	3.269	59.33	34.78	73.841	2.123	3	593.3	9.68E-04	3.42E-04	5.03E-03
C-19-109	3.852	3.274	56.43	32.41	68.206	2.104	1	564.3	4.75E-03	1.13E-03	2.99E-02
C-19-110	4.587	3.283	64.19	38.80	82.988	2.139	1	641.9	4.75E-03	1.31E-03	2.66E-02
C-19-209	4.181	3.280	59.95	35.31	75.743	2.145	3	599.5	4.75E-03	7.75E-04	3.15E-02
C-19-210	4.281	3.278	60.93	36.11	78.464	2.173	3	609.3	4.75E-03	7.65E-04	3.09E-02
C-19-115	4.216	3.283	60.37	35.66	76.180	2.136	1	603.7	9.33E-03	2.38E-03	5.51E-02
C-19-116	4.062	3.286	58.86	34.43	74.251	2.157	1	588.6	9.33E-03	2.47E-03	5.44E-02
C-19-215	3.966	3.275	57.62	33.39	71.256	2.134	3	576.2	9.33E-03	1.49E-03	6.34E-02
C-19-216	4.062	3.276	58.63	34.22	72.914	2.131	3	586.3	9.33E-03	1.51E-03	6.29E-02

Table A.3. Characteristics of large-sized monoliths, concentration results, and calculated values of sorbed materials in 1-month and 3-month iodine sorption tests

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm^2)	Volume (cm^3)	Weight (g)	Density (g/cm^3)	Test Duration (months)	Solution Volume (mL)	Starting I Concentration (μg/mL)	Final I Concentration (μg/mL)	I Sorbed (μg/g solid)
C-19-105	4.429	4.298	88.76	64.21	135.572	2.111	1	887.6	9.68E-04	2.68E-04	4.58E-03
C-19-106	4.355	4.303	87.91	63.30	132.800	2.098	1	879.1	9.68E-04	2.83E-04	4.53E-03
C-19-205	4.296	4.300	87.02	62.34	132.116	2.119	3	870.2	9.68E-04	2.07E-04	5.01E-03
C-19-206	4.266	4.302	86.68	61.98	134.957	2.178	3	866.8	9.68E-04	1.91E-04	4.99E-03
C-19-111	4.517	4.297	89.92	65.46	141.140	2.156	1	899.2	4.75E-03	1.31E-03	2.19E-02
C-19-112	4.658	4.300	91.92	67.61	144.852	2.142	1	919.2	4.75E-03	1.24E-03	2.23E-02
C-19-211	3.994	4.300	82.94	57.96	123.292	2.127	3	829.4	4.75E-03	7.19E-04	2.71E-02
C-19-212	3.880	4.311	81.69	56.59	124.377	2.198	3	816.9	4.75E-03	7.03E-04	2.66E-02
C-19-117	4.085	4.302	84.24	59.35	130.627	2.201	1	842.4	9.33E-03	2.46E-03	4.43E-02
C-19-118	4.385	4.291	87.99	63.38	139.518	2.201	1	879.9	9.33E-03	2.16E-03	4.52E-02
C-19-217	4.350	4.309	88.01	63.40	135.215	2.133	3	880.1	9.33E-03	1.46E-03	5.12E-02
C-19-218	4.232	4.305	86.30	61.57	132.194	2.147	3	863.0	9.33E-03	1.36E-03	5.20E-02

A.2 Tc-99 Experiments

Monolith characteristics, test conditions, measured iodine concentrations and calculated values of technetium-99 (Tc-99) sorbed are given in the following tables for small (Table A.4), medium (Table A.5), and large (Table A.6) monoliths. The EQL of the analytical method used to detect Tc-99 is 2.6E-05 μg/mL.

Table A.4. Characteristics of small-sized monoliths, concentration results, and calculated values of sorbed materials in 1-month and 3-month Tc-99 sorption tests

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm^2)	Volume (cm^3)	Weight (g)	Density (g/cm^3)	Test Duration (months)	Solution Volume (mL)	Starting Tc-99 Concentration (μg/mL)	Final Tc-99 Concentration (μg/mL)	Tc-99 Sorbed (μg/g solid)
C-19-119	3.652	2.204	32.89	13.92	30.731	2.208	1	328.9	9.90E-04	9.36E-04	5.73E-04
C-19-120	4.018	2.198	35.31	15.23	32.561	2.138	1	353.1	9.90E-04	9.49E-04	4.39E-04
C-19-219	4.067	2.208	35.85	15.56	35.174	2.260	3	358.5	9.90E-04	9.83E-04	6.63E-05
C-19-220	3.938	2.217	35.12	15.19	33.463	2.203	3	351.2	9.90E-04	9.76E-04	1.42E-04
C-19-125	4.165	2.207	36.50	15.92	34.092	2.142	1	365.0	4.79E-03	4.81E-03	-2.68E-04
C-19-126	4.089	2.208	36.00	15.65	33.780	2.159	1	360.0	4.79E-03	4.89E-03	-1.12E-03
C-19-225	3.903	2.212	34.79	14.99	32.176	2.146	3	347.9	4.79E-03	5.01E-03	-2.43E-03
C-19-226	3.994	2.206	35.30	15.25	33.048	2.167	3	353.0	4.79E-03	5.10E-03	-3.36E-03
C-19-131	3.835	2.206	34.20	14.64	31.273	2.136	1	342.0	9.96E-03	9.57E-03	4.26E-03
C-19-132	4.008	2.203	35.34	15.27	33.221	2.176	1	353.4	9.96E-03	9.59E-03	3.94E-03
C-19-231	4.172	2.207	36.56	15.95	34.200	2.144	3	365.6	9.96E-03	9.94E-03	2.14E-04
C-19-232	3.777	2.208	33.83	14.45	30.962	2.143	3	338.3	9.96E-03	9.96E-03	0.00E+00

Table A.5. Characteristics of medium-sized monoliths, concentration results, and calculated values of sorbed materials in 1-month and 3-month Tc-99 sorption tests

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm^2)	Volume (cm^3)	Weight (g)	Density (g/cm^3)	Test Duration (months)	Solution Volume (mL)	Starting Tc-99 Concentration (μg/mL)	Final Tc-99 Concentration (μg/mL)	Tc-99 Sorbed (μg/g solid)
C-19-121	4.349	3.287	61.85	36.89	78.071	2.117	1	618.5	9.90E-04	9.18E-04	5.66E-04
C-19-122	4.092	3.276	58.93	34.46	72.926	2.116	1	589.3	9.90E-04	9.33E-04	4.57E-04
C-19-221	4.179	3.273	59.76	35.13	73.725	2.099	3	597.6	9.90E-04	9.69E-04	1.66E-04
C-19-222	3.922	3.275	57.16	33.01	72.036	2.182	3	571.6	9.90E-04	9.63E-04	2.10E-04
C-19-127	4.256	3.277	60.64	35.87	76.019	2.119	1	606.4	4.79E-03	4.69E-03	7.58E-04
C-19-128	4.277	3.269	60.68	35.88	75.160	2.095	1	606.8	4.79E-03	4.51E-03	2.22E-03
C-19-227	4.063	3.274	58.60	34.19	72.451	2.119	3	586.0	4.79E-03	4.94E-03	-1.25E-03
C-19-228	4.772	3.281	66.05	40.31	85.201	2.113	3	660.5	4.79E-03	5.13E-03	-2.67E-03
C-19-133	4.077	3.278	58.82	34.38	73.043	2.125	1	588.2	9.96E-03	9.62E-03	2.74E-03
C-19-134	4.368	3.276	61.77	36.79	78.927	2.145	1	617.7	9.96E-03	9.35E-03	4.77E-03
C-19-233	4.072	3.275	58.70	34.27	72.471	2.114	3	587.0	9.96E-03	9.90E-03	4.86E-04
C-19-234	4.130	3.282	59.46	34.91	74.884	2.145	3	594.6	9.96E-03	9.77E-03	1.51E-03

Table A.6. Characteristics of large-sized monoliths, concentration results, and calculated values of sorbed materials in 1-month and 3-month Tc-99 sorption tests

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm^2)	Volume (cm^3)	Weight (g)	Density (g/cm^3)	Test Duration (months)	Solution Volume (mL)	Starting Tc-99 Concentration (μg/mL)	Final Tc-99 Concentration (μg/mL)	Tc-99 Sorbed (μg/g solid)
C-19-123	4.364	4.303	88.03	63.43	134.744	2.124	1	880.3	9.90E-04	9.30E-04	3.89E-04
C-19-124	4.520	4.300	90.06	65.61	141.743	2.161	1	900.6	9.90E-04	9.51E-04	2.45E-04
C-19-223	4.313	4.307	87.45	62.81	130.877	2.084	3	874.5	9.90E-04	9.93E-04	-2.34E-05
C-19-224	4.080	4.306	84.28	59.39	125.325	2.110	3	842.8	9.90E-04	1.01E-03	-1.38E-04
C-19-129	4.442	4.305	89.14	64.62	136.045	2.105	1	891.4	4.79E-03	4.83E-03	-2.95E-04
C-19-130	4.512	4.312	90.27	65.84	138.877	2.109	1	902.7	4.79E-03	4.38E-03	2.63E-03
C-19-229	4.138	4.296	84.79	59.95	127.165	2.121	3	847.9	4.79E-03	4.79E-03	-3.33E-05
C-19-230	4.608	4.307	91.43	67.09	140.515	2.095	3	914.3	4.79E-03	4.81E-03	-1.63E-04
C-19-135	4.052	4.290	83.46	58.53	126.819	2.167	1	834.6	9.96E-03	9.26E-03	4.61E-03
C-19-136	4.325	4.312	87.75	63.13	133.884	2.121	1	877.5	9.96E-03	9.16E-03	5.24E-03
C-19-235	4.344	4.301	87.70	63.07	132.589	2.102	3	877.0	9.96E-03	9.64E-03	2.12E-03
C-19-236	4.384	4.309	88.45	63.88	135.082	2.114	3	884.5	9.96E-03	9.42E-03	3.54E-03

A.3 Cation Results from Iodine Experiments

Concentrations for calcium, magnesium, potassium, and sodium are shown in Table A.7. Average values for the starting solution are given in the first row and the values presented in the following rows are from iodine experiment solutions at the end of the given test duration. EQL values are given in the final row.

Table A.7. Cation concentrations from iodine test solutions

Test #	Monolith Size	Test Duration (months)	Calcium Concentration (µg/ml)	Magnesium Concentration (µg/ml)	Potassium Concentration (µg/ml)	Sodium Concentration (µg/ml)
Starting Solution	-	-	393	1.86	4.09	5.74
C-19-101	small	1	398	ND	10.8	17.6
C-19-107	small	1	374	ND	12.2	19.3
C-19-113	small	1	526	ND	11.8	19.6
C-19-201	small	3	403	ND	0.403	31.2
C-19-207	small	3	471	ND	0.471	31.1
C-19-213	small	3	548	ND	0.548	32.1
C-19-103	medium	1	362	ND	13.6	22.3
C-19-109	medium	1	420	ND	13.5	22.3
C-19-115	medium	1	516	ND	13.8	22.5
C-19-203	medium	3	432	ND	0.432	39.2
C-19-209	medium	3	482	ND	0.482	41.7
C-19-215	medium	3	569	ND	0.569	39.8
C-19-105	large	1	416	ND	16	25
C-19-111	large	1	434	ND	14.7	22.8
C-19-117	large	1	318	ND	14.7	23
C-19-205	large	3	445	ND	0.445	38.7
C-19-211	large	3	498	0.03	0.498	39.3
C-19-217	large	3	490	0.02	0.490	40.2
EQL	-	-	0.17	0.02	0.602	0.17

ND - Not detected

A.4 Cation Results from Tc-99 Experiments

Concentrations for calcium, magnesium, potassium, and sodium are shown in Table A.8. Average values for the starting solution are given in the first row and the values presented in the following rows are from Tc-99 experiment solutions at the end of the given test duration. EQL values are given in the final row.

Table A.8. Cation concentrations from Tc-99 test solutions

Test #	Monolith Size	Test Duration (months)	Calcium Concentration (µg/ml)	Magnesium Concentration (µg/ml)	Potassium Concentration (µg/ml)	Sodium Concentration (µg/ml)
Starting Solution	-	-	380	1.33	4.20	5.93
C-19-119	small	1	412	ND	12.6	21.3
C-19-125	small	1	444	ND	12	20.1
C-19-131	small	1	380	ND	11.5	19.8
C-19-219	small	3	427	ND	0.427	32.2
C-19-225	small	3	410	ND	0.41	30.6
C-19-231	small	3	444	ND	0.444	32.9
C-19-121	medium	1	387	ND	13.9	22.5
C-19-127	medium	1	346	ND	14.5	23.9
C-19-133	medium	1	382	ND	15.5	26.2
C-19-221	medium	3	361	ND	0.361	41.4
C-19-227	medium	3	432	0.03	0.432	34
C-19-233	medium	3	357	ND	0.357	39.4
C-19-123	large	1	416	ND	15.4	23.3
C-19-129	large	1	361	ND	15.1	22.3
C-19-135	large	1	409	ND	16.4	25.4
C-19-223	large	3	431	0.18	0.431	47.8
C-19-229	large	3	457	ND	0.457	43.5
C-19-235	large	3	456	0.02	0.456	46.8
EQL	-	-	0.17	0.02	0.60	0.17

ND - Not detected

A.5 Anion Results from Iodine Experiments

Concentrations for chloride, nitrate, and sulfate are shown in Table A.9. Average values for the starting solution are given in the first row and the values presented in the following rows are from iodine experiment solutions at the end of the given test duration. EQL values are given in the final row.

Table A.9. Anion concentrations from iodine test solutions

Test #	Monolith Size	Test Duration (months)	Chloride Concentration (µg/ml)	Nitrate Concentration (µg/ml)	Sulfate Concentration (µg/ml)
Starting Solution	-	-	12.5	ND	43.5
C-19-101	small	1	10.4	1.78	9.53
C-19-102	small	1	10.3	1.94	9.16
C-19-107	small	1	10.8	2.08	12
C-19-108	small	1	10.3	2.11	12
C-19-113	small	1	10.4	2.04	16.1
C-19-114	small	1	10.6	1.69	18.1
C-19-201	small	3	9.13	2.87	5.14
C-19-202	small	3	8.87	3.32	4.91
C-19-207	small	3	9.18	3.18	5.91
C-19-208	small	3	8.73	3.66	5.52
C-19-213	small	3	9.2	3.28	8.03
C-19-214	small	3	9.55	2.93	8.99
C-19-103	medium	1	9.59	2.5	8.22
C-19-104	medium	1	9.91	2.53	8.79
C-19-109	medium	1	9.99	2.29	11.6
C-19-110	medium	1	10.1	2.18	12.1
C-19-115	medium	1	10.1	2.38	15.3
C-19-116	medium	1	10.1	2.09	16.1
C-19-203	medium	3	8.38	4.07	4.38
C-19-204	medium	3	8.39	4.44	4.61
C-19-209	medium	3	8.76	4.04	5.75
C-19-210	medium	3	8.87	3.78	5.93
C-19-215	medium	3	9.09	4.16	7.45
C-19-216	medium	3	9.26	3.6	8.13
C-19-105	large	1	10	2.66	10.5
C-19-106	large	1	9.81	2.68	10.7
C-19-111	large	1	10.4	2.02	14.6
C-19-112	large	1	10.5	2.04	14.9
C-19-117	large	1	10.2	2.15	13.8
C-19-118	large	1	10.1	2.15	13.4
C-19-205	large	3	9.08	3.47	5.77
C-19-206	large	3	9.22	3.8	5.92
C-19-211	large	3	9.3	3.71	6.97
C-19-212	large	3	9.26	3.54	7.04
C-19-217	large	3	9.27	3.7	6.84
C-19-218	large	3	9.18	3.53	6.8
EQL	-	-	0.75	1.5	0.5

ND - Not detected

A.6 Anion Results from Tc-99 Experiments

Concentrations for chloride, nitrate, and sulfate are shown in Table A.10 Average values for the starting solution are given in the first row and the values presented in the following rows are from Tc-99 experiment solutions at the end of the given test duration. EQL values are given in the final row.

Table A.10. Anion concentrations from Tc-99 test solutions

Test #	Monolith Size	Test Duration (months)	Chloride Concentration (µg/ml)	Nitrate Concentration (µg/ml)	Sulfate Concentration (µg/ml)
Starting Solution	-	-	12.9	ND	45.5
C-19-119	small	1	10.4	1.87	13.2
C-19-120	small	1	10	1.91	14.2
C-19-125	small	1	10.8	1.97	14.3
C-19-126	small	1	10.4	2.08	14.9
C-19-131	small	1	10.3	1.97	15
C-19-132	small	1	10.2	1.94	13.5
C-19-219	small	3	8.97	3.43	5.9
C-19-220	small	3	8.78	3.44	5.5
C-19-225	small	3	9.67	2.62	7.97
C-19-226	small	3	9.2	3.15	6.95
C-19-231	small	3	9.04	3.33	6.41
C-19-232	small	3	9.3	3.15	7.06
C-19-121	medium	1	10	2.22	16.8
C-19-122	medium	1	9.95	2.41	11.5
C-19-127	medium	1	9.9	2.19	15
C-19-128	medium	1	9.95	2.11	18.4
C-19-133	medium	1	10	2.36	13.3
C-19-134	medium	1	9.96	2.46	13
C-19-221	medium	3	8.65	3.83	5.89
C-19-222	medium	3	8.93	3.44	6.83
C-19-227	medium	3	9.52	3.13	8.42
C-19-228	medium	3	9.06	3.1	7.09
C-19-233	medium	3	8.78	3.46	6.53
C-19-234	medium	3	8.45	3.93	5.58
C-19-123	large	1	10.2	2.26	26.3
C-19-124	large	1	10.9	ND	36.7
C-19-129	large	1	9.91	2.01	23.5
C-19-130	large	1	9.96	1.97	23.3
C-19-135	large	1	10.5	2.1	20.3
C-19-136	large	1	10.5	1.6	31
C-19-223	large	3	8.96	4.15	12
C-19-224	large	3	9.32	4.03	15.8
C-19-229	large	3	8.57	4.05	9.63
C-19-230	large	3	7.04	4.09	7.42
C-19-235	large	3	8.77	4.22	9.19
C-19-236	large	3	8.94	3.87	17.2
EQL	-	-	0.75	1.5	0.5

ND - Not detected

Pacific Northwest National Laboratory

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99354
1-888-375-PNNL (7665)

www.pnnl.gov