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	FY 2020 Radionuclide Migration Project Status
	December 2020
	EA Cordova EC Golovich
	U.S. DEPARTMENT OF
	ENERGY Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

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Pacific Northwest National Laboratory Richland, Washington 99354

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) 2020 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. FY 2020 test conditions were chosen to provide direct comparison to both FY 2018 and FY 2019 testing. FY 2018 tests were conducted in a single matrix solution using three sizes of concrete monoliths in contact with a single contaminant at three concentrations. In FY 2019 tests were conducted in a modified matrix solution using the same three concrete monolith sizes, however, the concentration range was increased for both contaminant spikes. Based on FY 2019 test results, it was not evident which of the changes had the most impact on the sorption capacity of the concrete monoliths. FY 2020 testing was divided into two smaller sets utilizing each of the two matrix solutions, only one monolith size, and the same concentration range as FY 2019. The two test solutions included a saturated calcium hydroxide [Ca(OH)₂] solution and a modified solution with a composition more representative of the Hanford site groundwater.

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.

lodine partition coefficients calculated for FY 2020 ranged from 9.201 mL/g for the 1-month tests using the saturated Ca(OH)₂ solution to 23.221 mL/g for the 3-month tests with the modified Ca(OH)₂ saturated solution with simulated groundwater. FY 2020 K_d values are comparable to FY 2018 K_d values at both test durations for unmodified solution matrix tests. The results are reasonable given a \pm 20% for analytical results. Comparison of FY 2020 and FY 2019 shows a decrease in K_d values for FY 2020 of approximately 50%. Based on these results it is highly probable that addition of simulated groundwater components does not improve sorption capacity. For all iodine tests in this report iodine is added in the form of iodate (IO₃⁻). In general, iodate adsorbs more strongly than iodide and adsorption is inversely proportional to pH.¹ Measurement of pH was taken for all tests at each test duration and pH of the test solutions both before testing (12.33) and after was between 12.23 and 12.49 indicating no change in pH due to testing. Starting solutions are saturated with respect to Ca(OH)₂ and some precipitate was noted in test containers. It is possible that the higher K_d values for FY 2019 may not be attributable to sorption but may include some precipitation due to the addition of groundwater components.

Tc-99 partition coefficients calculated for FY 2020 ranged from 0.0719 mL/g for the 1-month tests using the modified Ca(OH)₂ saturated solution with simulated groundwater to 0.2448 mL/g for the 3-month tests with the saturated Ca(OH)₂. Due to an incorrect calculation for spiking the matrix solutions all three target concentrations were 10x lower than intended. As a result, the data is limited to the single 1 ppb starting concentration for FY 2020 that can be compared to the same concentration in FY 2019 for the modified Ca(OH)₂ saturated solution with simulated groundwater and the same concentration in FY 2018 for unmodified saturated Ca(OH)₂ solution. Comparison of K_d values for tests at the same concentration in the modified Ca(OH)₂ saturated solution with simulated groundwater show a decrease in K_d value for FY 2020 at 1 month and an increase in K_d value at 3 months. FY 2018 K_d values for tests in unmodified Ca(OH)₂ solution are higher at both 1 and 3 months than FY 2020 tests in the same matrix. The results are not consistent over the three years of testing in either matrix and both test durations, however, given the lower concentration and small data set, the tests should be repeated in the unmodified saturated Ca(OH)₂ matrix at the correct concentrations for direct comparison.

Experimental evidence clearly supports the association of iodine with inorganic carbonate minerals,^{2,3,4} the existence of calcium iodate phases such as lautarite,⁵ and the incorporation of iodate into calcite over iodide.⁶

Future work considerations should include longer test durations to confirm steady state, the extent of iodine incorporation and the evaluation of precipitates by imaging techniques such as SEM. In a saturated $Ca(OH)_2$ solution the precipitate should be recognized as calcium-carbonate, or calcium-iodate-carbonate phases.

¹ Kaplan, DI, ME Denham, S Zhang, C Yeager, C Xu, KA Schwehr, HP Li, YF Ho, D Wellman, and PH Santschi. 2014. "Radioiodine Biogeochemistry and Prevalence in Groundwater." *Critical Reviews in Environmental Science and Technology* 44:20 2287–2335.

² Claret, F, C Lerouge, T Laurioux, M Bizi, T Conte, JP Ghestem, G Wille, T Sato, EC Gaucher, E Giffaut, and C Tournassat. 2010. "Natural iodine in a clay formation: Implications for Iodine Fate in Geological Disposals." *Geochimica et Cosmochimica Acta* 74:16–29.

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⁴ Xu, C, DI Kaplan, S Zhang, M Athon, YF Ho, HP Li, CM Yeager, KA Schwehr, R Grandbois, D Wellman, and PH Santschi. 2015. "Radioiodine Sorption/Desorption and Speciation Transformation by Subsurface Sediments from the Hanford Site." *Journal of Environmental Radioactivity* 139:43–55.

⁵ Ghose S and C Wan. 1978. "The Crystal Structure of Synthetic Lautarite, Ca(IO₃)₂." Acta Crystallographica B34:84–88.

⁶ Lu, Z, HC Jenkyns, and REM Ricaby. 2010. "Iodine to Calcium Ratios in Marine Carbonate as a Paleo-Redox Proxy During Oceanic Anoxic Events." Geology 38:12 1107–1110.

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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
HFS	Hanford fine sands
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 nearfield environment.

This report presents results from a set of sorption experiments completed in fiscal year (FY) 2020 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. In FY 2020, experimental conditions from FY 2018 and FY 2019 were repeated to minimize the variability in how monolith samples and test conditions throughout the experiments. One batch of concrete was used to create a set of monoliths to use with two test solutions. Test solutions included a saturated calcium hydroxide [Ca(OH)₂] solution and a composition more representative of the Hanford site groundwater.

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 $6.0 \pm 1.5\%$. The nominal proportions and material specifications based on this initial design are listed in Table 2.1.

		Specified	Normalized
Material	Specifications	Field Mix	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%	

Table 2.1. Concrete material specifications and composition

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, an iron powder (when applicable); 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.

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 (tracer test) or Portland Type III (carbonation test)
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 (0.149 to 0.177 mm) $^{\rm (a)}$
PolyHeed 997		0.00375	Water-entraining agent
Air Content	6.0 ± 1.5%	6.0 ± 1.5%	

Table 2.2. Laboratory-scale material specification and composition

(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 concentrations for I and Tc-99, two test durations, and two test solutions as shown in Table 2.3. The modified ground water test solutions were prepared using a saturated Ca(OH)₂ solution to which several groundwater constituents representative of groundwater from the Hanford site were introduced. Table 2.4 lists the groundwater constituents used for the test solutions prior to addition of I or Tc-99. A total of 24 different test conditions, shown in Table 2.3, were run in duplicate. The 48 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.

Variable	Conditions
Species	Tc-99 or I
Concentrations (µg/L)	l: 1, 5, and 10 Tc-99: 0.1, 0.5, and 1
Duration (months)	1 and 3
Test solution	Saturated Ca(OH) ₂ and modified groundwater

Table 2.3. Batch sorption test variables

Table 2.4. Modified groundwater composition

Constituent	Concentration (g/L)
Ca(OH) ²	1.038
H ₄ SiO ₄	0.0153
KCI	0.0082
NaCl	0.015
CaSO ₄	0.067
рН	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 FY 2020 to evaluate partition coefficients (K_d) for iodine and Tc-99 using intact concrete monoliths. The test matrix included three solution concentrations, two test solutions, and two test durations. 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 the saturated $Ca(OH)_2$ solution (Table A.1) and the modified $Ca(OH)_2$ saturated solution with simulated groundwater (Table A.2). Tables A.3 and A.4 present the data previously reported for FY 2019 and FY 2018 respectively. Figure 3.1 shows the iodine sorbed to the monoliths as a function of the final iodine concentrations for both solution matrices and test durations.



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

A summary of the partition coefficients calculated for iodine is given in Table 3.1. Partition coefficients calculated for FY 2020 ranged from 9.201 mL/g for the 1-month tests using the saturated Ca(OH)₂ solution to 23.221 mL/g for the 3-month tests with the modified Ca(OH)₂ saturated solution with simulated groundwater. The results from FY 2018 and FY 2019 are also shown in Table 3.1 for comparison. The results for the saturated Ca(OH)₂ solution are similar for

FY 2018 and FY 2020. Approximately a factor of two increase is observed for both test solutions as the test duration increases for all experiments. Partition coefficients are larger for the modified groundwater tests in the FY 2020 data. The difference in results from last year's experiments may be due to the addition of groundwater constituents and precipitation processes that may be interfering with sorption.

Test Duration	Test Solution	FY 2020	FY 2019	FY 2018			
1 month	Saturated Ca(OH) ₂	9.201		7.631			
	Modified groundwater	12.581	22.816				
3 months	Saturated Ca(OH) ₂	15.340		13.7390 ^(a)			
	Modified groundwater 23.221 47.031						
a – Test duration for this set of data was 4 months							

Table 3.1. Summary of calculated iodine partition coefficients

3.2 Tc-99 Sorption

Specific test conditions including monolith characteristics, starting and final concentrations, and calculations of Tc-99 sorbed are provided in Appendix Section A.2. Separate tables contain details for tests using the saturated $Ca(OH)_2$ solution (Table A.5) and the modified $Ca(OH)_2$ saturated solution with simulated groundwater (Table A.6). Tables A.7 and A.8 present the data previously reported for FY 2019 and FY 2018 respectively. Figure 3.2 shows the Tc-99 sorbed to the monoliths as a function of the final Tc-99 concentrations for both solution matrices and test durations.

A summary of the partition coefficients calculated for Tc-99 is given in Table 3.2. Partition coefficients calculated for FY 2020 ranged from 0.0719 mL/g for the 1-month tests using the modified Ca(OH)₂ saturated solution with simulated groundwater to 0.2448 mL/g for the 3-month tests with the saturated Ca(OH)₂. The results from FY 2018 and FY 2019 are also shown in Table 3.2 for comparison. Due to an incorrect calculation for spiking the matrix solutions all three target concentrations were 10x lower than intended. Final concentrations from ICP-MS analysis reflect the lower concentrations although concentrations remained above detection limits. This resulted in direct comparison only for tests with a starting concentration of 1 ppb Tc-99. In FY 2020, unlike iodine, K_d values were slightly smaller for the modified groundwater than the saturated Ca(OH)₂ solution. An increase in K_d values is observed with increasing test duration. The addition of simulated groundwater components to the solution matrix resulted in higher K_d values but this may also be attributed to other processes rather than uptake by the monoliths.





Test Duration	Test Solution	FY 2020 ^(a)	FY 2019 ^(b)	FY 2018 ^(c)			
1 month	Saturated Ca(OH) ₂	0.0988		0.3508			
	Modified groundwater	0.0719	0.3778				
3 months	Saturated Ca(OH) ₂	0.2448		1.0092 ^(d)			
	Modified groundwater	0.2351	0.1042				
a – Initial Tc-99 concentrations were 0.1, 0.5 and 1.0 ppb							

Table 3.2. Summary of calculated Tc-99 partition coefficients

b - Initial Tc-99 concentrations were 1.0, 5.0, and 10 ppb

c - Initial Tc-99 concentrations were 0.5, 1.0, and 5.0 ppb d - Test duration for this set of data was 4 months

3.3 Conclusions

Regarding iodine, FY 2020 Kd values are comparable to FY 2018 Kd values at both test durations for unmodified solution matrix tests. The results are reasonable given a ±20% for analytical results. Comparison of FY 2020 and FY 2019 shows a decrease in K_d values for FY 2020 of approximately 50%. Based on these results it is highly probable that addition of simulated groundwater components does not improve sorption capacity. For all iodine tests in this report iodine is added in the form of iodate (IO₃-). In general, iodate adsorbs more strongly than iodide and adsorption is inversely proportional to pH (Kaplan et al. 2014). Measurement of pH was taken for all tests at each test duration and pH of the test solutions both before testing

(12.33) and after was between 12.23 and 12.49 indicating no change in pH due to testing. Starting solutions are saturated with respect to Ca(OH)₂ and some precipitate was noted in test containers. It is possible that the higher K_d values for FY 2019 may not be attributable to sorption but may include some precipitation due to the addition of groundwater components.

Due to an incorrect calculation for spiking the matrix solutions all three target concentrations were 10x lower than intended. As a result, the Tc-99 data is limited to the single 1 ppb starting concentration for FY 2020 that can be compared to the same concentration in FY 2019 for the modified Ca(OH)₂ saturated solution with simulated groundwater and the same concentration in FY 2018 for unmodified saturated Ca(OH)₂ solution. Comparison of K_d values for tests at the same concentration in the modified Ca(OH)₂ saturated solution with simulated groundwater show a decrease in K_d value for FY 2020 at 1 month and an increase in K_d value at 3 months. FY 2018 K_d values for tests in unmodified Ca(OH)₂ solution are higher at both 1 and 3 months than FY 2020 tests in the same matrix. The results are not consistent over the three years of testing in either matrix and both test durations, however, given the lower concentration and small data set, the tests should be repeated in the unmodified saturated Ca(OH)₂ matrix at the correct concentrations for direct comparison.

Experimental evidence clearly supports the association of iodine with inorganic carbonate minerals (Claret et al. 2010; Zhang et al. 2013; Xu et al. 2015), the existence of calcium iodate phases such as lautarite (Ghose et al. 1978) and the incorporation of iodate into calcite over iodide (Lu et al. 2010).

It is also of interest to determine whether the test durations that have been examined are indicating the full extent of iodine incorporation. For example, Xu et al, (2015) report ranges of iodine measured in association with calcite between 2.9 - 39.4% of total iodine concentrations in various samples.

Future work considerations should include longer test durations to confirm steady state, the extent of iodine incorporation and the evaluation of precipitates by imaging techniques such as SEM. In a saturated $Ca(OH)_2$ solution the precipitate should be recognized as calcium-carbonate, or calcium-iodate-carbonate phases.

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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 $\pm 10\%$ variability for standards and $\pm 20\%$ 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 table. The estimated quantification limit (EQL) of the analytical method used to detect iodine is $8.30E-05 \ \mu g/mL$.

Table A.1. Characteristics of FY 2020 monoliths, concentration results, and calculated values of sorbed materials in 1 month and 3 month iodine sorption tests in the saturated Ca(OH)₂ solution

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)	pН	Starting I Concentration (µg/mL)	Final I Concentration (µg/mL)	I Sorbed (µg/g solid)
C-20-101	4.048	3.267	58.28	33.92	72.270	2.13	1	582.8	12.477	1.09E-03	5.09E-04	4.69E-03
C-20-102	3.958	3.271	57.45	33.24	71.557	2.15	1	574.5	12.487	1.09E-03	5.90E-04	4.01E-03
C-20-103	4.102	3.280	59.14	34.64	72.199	2.08	1	591.4	12.477	4.92E-03	2.23E-03	2.20E-02
C-20-104	4.118	3.276	59.20	34.68	71.810	2.07	1	592.0	12.473	4.92E-03	2.12E-03	2.31E-02
C-20-105	4.021	3.267	58.01	33.69	72.621	2.16	1	580.1	12.464	9.68E-03	4.49E-03	4.15E-02
C-20-106	3.767	3.264	55.33	31.50	66.648	2.12	1	553.3	12.453	9.68E-03	4.63E-03	4.19E-02
C-20-201	3.899	3.271	56.84	32.75	69.810	2.13	3	568.4	12.373	1.09E-03	4.41E-04	5.28E-03
C-20-202	3.988	3.266	57.63	33.38	71.778	2.15	3	576.3	12.399	1.09E-03	4.11E-04	5.45E-03
C-20-203	4.239	3.275	60.43	35.69	76.112	2.13	3	604.3	12.328	4.92E-03	1.72E-03	2.54E-02
C-20-204	3.738	3.260	54.94	31.18	66.219	2.12	3	549.4	12.356	4.92E-03	1.60E-03	2.75E-02
C-20-205	4.210	3.271	60.04	35.36	74.685	2.11	3	600.4	12.371	9.68E-03	3.44E-03	5.02E-02
C-20-206	3.775	3.263	55.39	31.55	67.515	2.14	3	553.9	12.385	9.68E-03	3.33E-03	5.21E-02

Table A.2. Characteristics of FY 2020 monoliths, concentration results, and calculated values of
sorbed materials in 1 month and 3 month iodine sorption tests in the modified
Ca(OH) ₂ saturated solution with simulated groundwater

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)	pН	Starting I Concentration (µg/mL)	Final I Concentration (µg/mL)	l Sorbed (μg/g solid)
C-20-301	3.911	3.268	56.89	32.78	69.251	2.11	1	568.9	12.434	1.12E-03	5.09E-04	5.02E-03
C-20-302	4.101	3.275	59.00	34.52	73.213	2.12	1	590.0	12.434	1.12E-03	6.16E-04	4.06E-03
C-20-303	3.949	3.268	57.29	33.11	69.604	2.10	1	572.9	12.410	5.14E-03	1.92E-03	2.65E-02
C-20-304	4.001	3.274	57.95	33.66	70.022	2.08	1	579.5	12.421	5.14E-03	2.30E-03	2.35E-02
C-20-305	3.934	3.273	57.24	33.07	68.726	2.08	1	572.4	12.419	1.04E-02	4.40E-03	5.00E-02
C-20-306	3.962	3.265	57.36	33.16	69.489	2.10	1	573.6	12.388	1.04E-02	4.02E-03	5.27E-02
C-20-401	4.034	3.269	58.17	33.83	71.983	2.13	3	581.7	12.326	1.12E-03	4.66E-04	5.29E-03
C-20-402	4.330	3.272	61.30	36.39	76.596	2.10	3	613.0	12.326	1.12E-03	3.70E-04	6.00E-03
C-20-403	3.848	3.270	56.29	32.29	68.699	2.13	3	562.9	12.317	5.14E-03	1.79E-03	2.74E-02
C-20-404	3.737	3.265	55.05	31.27	67.461	2.16	3	550.5	12.334	5.14E-03	1.51E-03	2.96E-02
C-20-405	3.817	3.261	55.77	31.85	67.880	2.13	3	557.7	12.279	1.04E-02	2.84E-03	6.21E-02
C-20-406	4.115	3.272	59.07	34.57	75.081	2.17	3	590.7	12.233	1.04E-02	2.72E-03	6.04E-02

Table A.3. Characteristics of FY 2019 medium monoliths, concentration results, and calculated values of sorbed materials in 1 month and 3 month iodine sorption tests in the modified Ca(OH)₂ saturated solution with simulated groundwater

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)	l Sorbed (µg/g solid)
C-19-103	4.119	3.281	59.34	34.81	75.097	2.16	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	2.04	1	597.5	9.68E-04	2.95E-04	5.62E-03
C-19-109	3.852	3.274	56.43	32.41	68.206	2.10	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	2.14	1	641.9	4.75E-03	1.31E-03	2.66E-02
C-19-115	4.216	3.283	60.37	35.66	76.180	2.14	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	2.16	1	588.6	9.33E-03	2.47E-03	5.44E-02
C-19-203	4.179	3.274	59.79	35.16	75.850	2.16	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	2.12	3	593.3	9.68E-04	3.42E-04	5.03E-03
C-19-209	4.181	3.280	59.95	35.31	75.743	2.15	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	2.17	3	609.3	4.75E-03	7.65E-04	3.09E-02
C-19-215	3.966	3.275	57.62	33.39	71.256	2.13	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	2.13	3	586.3	9.33E-03	1.51E-03	6.29E-02

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm ²)	Volume (cm³)	Weight (g)	Density (g/cm ³)	Test Duration (months)	Solution Volume (mL)	Starting I Concentration (µg/mL)	Final I Concentration (µg/mL)	l Sorbed (μg/g solid)	
C-18-208	4.119	3.281	59.34	34.81	75.097	2.16	1	593.4	1.08E-03	9.21E-04	1.22E-03	
C-18-209	4.169	3.277	59.75	35.13	71.512	2.04	1	597.5	1.08E-03	8.78E-04	1.65E-03	
C-18-214	3.852	3.274	56.43	32.41	68.206	2.10	1	564.3	1.08E-03	2.79E-03	-1.42E-02	
C-18-215	4.587	3.283	64.19	38.80	82.988	2.14	1	641.9	1.08E-03	2.70E-03	-1.26E-02	
C-18-210	4.216	3.283	60.37	35.66	76.180	2.14	4	603.7	4.75E-03	5.40E-04	3.33E-02	
C-18-211	4.062	3.286	58.86	34.43	74.251	2.16	4	588.6	4.75E-03	5.60E-04	3.32E-02	
C-18-216	4.179	3.274	59.79	35.16	75.850	2.16	4	597.9	4.75E-03	1.86E-03	2.27E-02	
C-18-217	4.147	3.269	59.33	34.78	73.841	2.12	4	593.3	4.75E-03	1.86E-03	2.32E-02	

Table A.4. Characteristics of FY 2018 monoliths, concentration results, and calculated values of sorbed materials in 1 month and 4 month iodine sorption tests in the saturated Ca(OH)₂ solution

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. The EQL of the analytical method used to detect Tc-99 is 2.6E-05 μ g/mL.

Table A.5. Characteristics of FY 2020 monoliths, concentration results, and calculated values of sorbed materials in 1 month and 3 month Tc-99 sorption tests in the saturated Ca(OH)₂ solution

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm²)	Volume (cm ³)	Weight (g)	Density (g/cm ³)	Test Duration (months)	Solution Volume (mL)	pН	Starting Tc-99 Concentration (µg/mL)	Final Tc-99 Concentration (µg/mL)	Tc-99 Sorbed (µg/g solid)
C-20-107	3.807	3.261	55.68	31.78	67.914	2.14	1	556.8	12.31	1.45E-04	1.48E-04	-2.31E-05
C-20-108	4.067	3.268	58.50	34.10	73.502	2.16	1	585.0	12.33	1.45E-04	1.43E-04	1.64E-05
C-20-109	3.799	3.270	55.78	31.88	67.809	2.13	1	557.8	12.31	6.64E-04	6.57E-04	5.41E-05
C-20-110	4.058	3.264	58.31	33.93	71.676	2.11	1	583.1	12.28	6.64E-04	6.61E-04	2.44E-05
C-20-111	3.829	3.274	56.18	32.21	67.560	2.10	1	561.8	12.29	1.29E-03	1.27E-03	1.55E-04
C-20-112	3.920	3.266	56.95	32.82	69.066	2.10	1	569.5	12.16	1.29E-03	1.28E-03	8.54E-05
C-20-207	3.939	3.266	57.14	32.98	69.625	2.11	3	571.4	12.18	1.45E-04	1.29E-04	1.31E-04
C-20-208	3.472	3.266	52.34	29.06	62.488	2.15	3	523.4	12.20	1.45E-04	1.30E-04	1.09E-04
C-20-209	3.803	3.268	55.79	31.88	67.653	2.12	3	557.9	12.20	6.64E-04	6.25E-04	2.86E-04
C-20-210	3.655	3.269	54.28	30.65	64.040	2.09	3	542.8	12.20	6.64E-04	6.23E-04	3.36E-04
C-20-211	3.875	3.275	56.69	32.63	70.584	2.16	3	566.9	12.15	1.29E-03	1.22E-03	5.31E-04
C-20-212	4.309	3.278	61.21	36.34	77.046	2.12	3	612.1	12.22	1.29E-03	1.26E-03	2.72E-04

	Ca(OH) ₂ saturated solution with simulated groundwater													
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)	pН	Starting Tc-99 Concentration (µg/mL)	Final Tc-99 Concentration (µg/mL)	Tc-99 Sorbed (μg/g solid)		
C-20-307	3.856	3.271	56.39	32.38	69.887	2.16	1	563.9	12.22	1.50E-04	1.47E-04	2.44E-05		
C-20-308	3.908	3.270	56.90	32.79	68.799	2.10	1	569.0	12.13	1.50E-04	1.44E-04	4.66E-05		
C-20-309	3.904	3.263	56.70	32.62	68.621	2.10	1	567.0	12.26	6.67E-04	6.58E-04	7.33E-05		
C-20-310	3.646	3.263	54.06	30.46	64.710	2.12	1	540.6	12.24	6.67E-04	6.53E-04	1.08E-04		
C-20-311	3.714	3.267	54.85	31.11	64.527	2.07	1	548.5	12.22	1.31E-03	1.29E-03	1.60E-04		
C-20-312	3.822	3.263	55.88	31.94	68.375	2.14	1	558.8	12.30	1.31E-03	1.30E-03	8.04E-05		
C-20-407	4.119	3.271	59.10	34.60	71.500	2.07	3	591.0	12.14	1.50E-04	1.30E-04	1.64E-04		
C-20-408	3.716	3.263	54.78	31.05	65.456	2.11	3	547.8	12.09	1.50E-04	1.27E-04	1.64E-04		
C-20-409	3.624	3.266	53.91	30.35	64.635	2.13	3	539.1	12.05	6.67E-04	6.24E-04	3.37E-04		
C-20-410	4.014	3.269	57.98	33.67	71.727	2.13	3	579.8	12.18	6.67E-04	6.38E-04	2.49E-04		
C-20-411	3.825	3.271	56.08	32.13	68.421	2.13	3	560.8	12.14	1.31E-03	1.25E-03	4.96E-04		
C-20-412	3.734	3.265	55.01	31.24	66.557	2.13	3	550.1	12.16	1.31E-03	1.26E-03	3.66E-04		

Table A.6. Characteristics of FY 2020 monoliths, concentration results, and calculated values of sorbed materials in 1 month and 3 month Tc-99 sorption tests in the modified Ca(OH)₂ saturated solution with simulated groundwater

Table A.7. Characteristics of FY 2019 medium monoliths, concentration results, and calculated values of sorbed materials in 1 month and 3 month Tc-99 sorption tests in the modified Ca(OH)₂ saturated solution with simulated groundwater

Core ID	Length (cm)	Diameter (cm)	Surface Area (cm ²)	Volume (cm³)	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	2.12	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	2.12	1	589.3	9.90E-04	9.33E-04	4.57E-04
C-19-127	4.256	3.277	60.64	35.87	76.019	2.12	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	2.09	1	606.8	4.79E-03	4.51E-03	2.22E-03
C-19-133	4.077	3.278	58.82	34.38	73.043	2.12	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	2.15	1	617.7	9.96E-03	9.35E-03	4.77E-03
C-19-221	4.179	3.273	59.76	35.13	73.725	2.10	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	2.18	3	571.6	9.90E-04	9.63E-04	2.10E-04
C-19-227	4.063	3.274	58.60	34.19	72.451	2.12	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	2.11	3	660.5	4.79E-03	5.13E-03	-2.67E-03
C-19-233	4.072	3.275	58.70	34.27	72.471	2.11	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	2.14	3	594.6	9.96E-03	9.77E-03	1.51E-03

Table A.8. Characteristics of FY 2018 monoliths, concentration results, and calculated values of sorbed materials in 1 month and 4 month Tc-99 sorption tests in the saturated Ca(OH)₂ solution

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-18-227	3.821	3.292	56.51	32.51	69.580	2.14	1	565.1	1.01E-03	1.02E-03	-8.12E-05
C-18-228	3.888	3.295	57.27	33.14	70.730	2.13	1	572.7	1.01E-03	9.72E-04	3.08E-04
C-18-234	3.674	3.290	54.94	31.21	67.320	2.16	1	549.4	5.19E-03	4.95E-03	1.96E-03
C-18-235	3.811	3.290	56.36	32.38	69.060	2.13	1	563.6	5.19E-03	4.94E-03	2.04E-03
C-18-229	3.961	3.294	58.00	33.74	71.880	2.13	3	580.0	1.01E-03	9.34E-04	6.13E-04
C-18-230	3.892	3.316	57.78	33.58	71.340	2.12	3	577.8	1.01E-03	9.34E-04	6.16E-04
C-18-236	3.977	3.299	58.28	33.98	72.190	2.12	3	582.8	5.19E-03	4.63E-03	4.52E-03
C-18-237	3.940	3.296	57.83	33.60	71.530	2.13	3	578.3	5.19E-03	4.56E-03	5.09E-03

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