PNNL-24297 Rev. 1
RPT-SWCS-010, Rev. 0

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Extended Leach Testing of Simulated LAW Cast Stone Monoliths

August 2016

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U.S. DEPARTMENT OF
ENERGY

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Executive Summary

This revision to the original report[1] adds two longer term leach sets of data to the report and provides more discussion and graphics on how to interpret the results from long-term laboratory leach tests. The leach tests were performed at Pacific Northwest National Laboratory (PNNL) for Washington River Protection Solutions (WRPS) to evaluate the release of key constituents from monoliths of Cast Stone prepared with four simulated low-activity waste (LAW) liquid waste streams.

Specific objectives of the Cast Stone long-term leach tests described in this report focused on four activities:

1. Extending the leaching times of monoliths made with LAW simulants for selected ongoing EPA-1315 tests beyond the conventional 63-day time period, up to 1042 days reported herein (with some tests continuing that will be documented later). This effort was to evaluate long-term leaching properties of Cast Stone to support future performance assessment activities.

2. Starting new EPA-1315 leach tests using two eluents (deionized water [DIW] and simulated Hanford Integrated Disposal Facility (IDF) Site vadose zone pore water [VZP]) on archived Cast Stone monoliths made with four LAW simulants. The use of VZP is to more realistically leach Cast Stone in a solution that represents IDF sub-surface recharge water.

3. Evaluating the impacts of varying the iodide loading (starting iodide concentrations) in one LAW simulant (7.8 M Na Hanford Tank Waste Operations Simulator [HTWOS] Average) by manufacturing new Cast Stone monoliths and repeating the EPA-1315 leach tests using DIW and the VZP eluents. Past iodide loading into Cast Stone was unrealistically high to make sure iodine could be measured in eluates but past leaching results showed high releases of iodide that might be caused by the unrealistically high iodide loadings.

4. Evaluating the impacts of using a non-pertechnetate form of Tc that is present in some Hanford tanks. In this activity one LAW simulant (7.8 M Na HTWOS Average) was spiked with a Tc(I)-tricarbonyl gluconate species and then solidified into Cast Stone monoliths. Cured monoliths were leached using the EPA-1315 leach protocol with DIW and VZP. The leach results for the Tc-Gluconate Cast Stone monoliths were compared to Cast Stone monoliths containing pertechnetate. Non-pertechnetate species are present in double-shell tank supernates and there is concern that these non-pertechnetate species may exist in liquid wastes destined for solidification in grouts.

Results from the screening tests (see Westsik et al. 2013a) that leached 26 different Cast Stone mixes for 91-days found that there were no statistically significant correlations between the leach properties of any of the key constituents (technetium, iodide, chromium, nitrate, nitrite, sodium and uranium) with the type of waste simulant, the source of the dry blend materials, the free-water-to-dry blend ratio, or wet slurry properties. So despite finding a two order of magnitude range in ^{99}Tc and Cr observed diffusivities and almost an order of magnitude range in observed diffusivities for the more leachable anions (nitrate, nitrite, and iodide) and sodium we have not identified correlations with the Cast Stone or waste simulants that shed light on the controlling mechanism(s) that lead to the variation in leach properties.

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[1] This report is an updated revision of RPT-SLAW-001, Rev. 0 (PNNL-24297), which is available online at: http://www.osti.gov/scitech/biblio/1208783-extended-leach-testing-simulated-law-cast-stone-monoliths. This updated report adds data and discussion for monoliths leached beyond the timeframes in that original report, as well as updating terminology and analyses from the original report. The project report number now has “SWCS” in it, rather than “SLAW” because the funding project has changed for the revision. The PNNL report number is the same with “Rev. 1” added for the revision.

Thus monolith leach studies were continued with current results documented herein for the same key constituents. Updated conclusions were derived after leaching four different suites of Cast Stone, one suite to address each of the four objectives just described. Leaching data for uranium is hardly mentioned in the main text because almost all the eluates, regardless of eluent type used (VZP or DIW), had non-detectable uranium concentrations. That is, uranium is sequestered strongly in Cast Stone and leaches so slowly that eluate concentrations are not detectable for almost all the Cast Stone monoliths studied. We hypothesize that a very insoluble calcium uranate solid phase is keeping the uranium from leaching from the Cast Stone monoliths.

One universal observation from these extended leach studies on LAW Cast Stone monoliths, when leached in VZP, is that the leach rates of Tc, Cr, I, nitrate, nitrite and sodium are lower than when leached in DIW. One potential cause for the lower leach rates for these constituents when leached in VZP is the formation of significant quantities of secondary precipitates on the surface of the monoliths contacted with VZP while the monoliths contacted with DIW show only trace quantities of secondary precipitates that are associated with surface micro-cracks present after curing and/or extended leaching times. We hypothesize that more (higher local concentrations) Ca diffuses out of the monoliths along the micro-cracks, causing supersaturated conditions with the small concentrations of dissolved CO_2 right at the micro-cracks. Finding that Cast Stone releases lower amounts of the four key groundwater risk drivers (Tc, Cr, I, nitrate, nitrite) and sodium when leached in the more representative VZP will lower the observed diffusivity numbers that will be used in future predictive modeling of Cast Stone performance in comparison to the diffusivity values based on leaching in DIW and used in past IDF PA analyses.

Up through 34 months of leaching in DIW the leach rates for the mobile constituents (nitrate, nitrite, iodide, and sodium) showed a continual decrease; faster decrease in the first seven days followed by a continual slow decrease. There is a net weight loss of monoliths leached in DIW suggesting net dissolution that might lead to crack development at long times. For Cast Stone monoliths leached in VZP there is a net gain in mass as the leaching progresses, which undoubtedly is caused by the precipitates that form on the monoliths' outer surfaces. These precipitates may be the main cause for the lower observed leach rates when Cast Stone is leached in VZP.

Tables of interval averaged observed diffusion coefficients (D_{obs} values[1])—a way of quantifying leach rates—for ^{99}Tc show a factor of about 50 change from the best performing Cast Stone mix to the worst performing mix, regardless of which eluent is used. The same range of interval averaged D_{obs} values for best to worst Cast Stone mixes for Na, I, and nitrate differ by a factor of about 6, and for nitrite by a factor of only 3. Chromium interval averaged D_{obs} values for VZP eluents are generally not quantifiable because eluate Cr concentrations were below detection limits. Cr leach rates from Cast Stone leached in DIW are also very low but quantifiable. This means that Cr leach rates from Cast Stone are very low such that the potential risk to groundwater from Cr release from buried Cast Stone waste forms is unlikely.

The best and worst performing Cast Stone monolith mixes leached in VZP have moderate correlation with the best and worst performing monoliths when leached in DIW for each constituent of interest. For ^{99}Tc, Cast Stone Mix 24 is the best performing mix and Mixes 17 and 18 are among the worst performing. Leach results for iodide show that Cast Stone Mixes 10 and 24 are the best performing and Mix 21 is the worst performing. For nitrate and nitrite, Cast Stone Mixes 8, 10, 18 and 24 leach less and Mix 21 leaches the most. As mentioned Cr leach data are qualitative because many of the eluates from the various mixes had no detectable Cr such that choosing the best and worst performing Cast Stone mixes

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[1] Different reports name the calculated diffusion coefficients from laboratory leach tests differently (names used in the literature to represent the calculated diffusion coefficients include "observed", "effective", and "apparent"). Because we are using EPA Method 1315 protocol we use the term "observed" diffusion coefficient, D_{obs} as defined in the protocol. See Section 3.3 for more discussion.

for Cr release is not useful, all mixes retain Cr well. The same is true for uranium when leached in VZP where almost all the eluates had no detectable uranium that yielded D_{obs} values $<1 \times 10^{-16}$ to $<3 \times 10^{-16}$ cm^2/s. For two Mixes (8 and 16) leached in DIW, the eluate uranium concentrations were detectable yielding D_{obs} values of 1×10^{-15} and 4×10^{-16} cm^2/s, respectively.

Regardless of the Cast Stone mix composition, the D_{obs} values for the anions iodide, nitrate and nitrite and the cation sodium are very similar. The range in averaged interval diffusion coefficient values for these four constituents varies by only a factor of three to six. Cr leaches the slowest especially when VZP eluent is used, followed by ^{99}Tc and then the four more mobile constituents. Because the cumulative mass of mobile (iodide, nitrate, nitrite, and sodium) constituents released from the monoliths starts to exceed 20% of their starting inventories when either eluent is used generally between 49 to 63-d of leaching, we are recommending that the VZP leached averaged 28- to 63-d interval diffusion coefficient values be used for future IDF performance assessment predictive modeling, if an empirical diffusivity conceptual model is chosen for the release from LAW Cast Stone.

We recommend using the 28- to 63-d interval observed diffusion coefficient values from the tests using VZP eluent because it better represents water that will percolate through the IDF facility. Further, the early leach periods (0.08 d up to 14 d) are often biased high likely because of monolith surface salt wash-off and diffusion from matrix locations directly connected to the relatively small monoliths' outer surface. That is, the leach results from the early times are also not good indications of diffusion from full-sized Cast Stone monoliths destined for burial in IDF. Thus we chose the 28- to 63-d interval D_{obs} values as most representative of IDF burial conditions.

The white precipitate on the surfaces of monoliths leached in VZP has been identified by X-ray diffraction (XRD) as predominantly aragonite (a polymorph of calcium carbonate). Calcium carbonate is likely present covering some of the micro-cracks on monoliths leached in DIW, but we have not recovered adequate precipitate mass off the DIW leached monoliths to get a good XRD spectrum.

The iodide leach results from the Iodide Suite of monoliths with four different starting iodide inventories (from 1.6×10^{-5} to 7.7×10^{-3} wt%) show no significant differences. Thus, we conclude that at iodide loadings well below 0.14 wt% (the lowest loading used in previous Cast Stone studies that did show differing iodide leach rates as a function of iodine loading) will not impact the interval derived D_{obs} values measured in leach tests. HTWOS predictions of ^{129}I masses (converted from activity) for future LAW and Effluent Treatment Facility (ETF) treated secondary wastes are several orders of magnitude below this threshold value of 0.14 wt%. Thus, we believe that there should be no iodide loading sensitivity in actual Cast Stone waste forms. The lower limit iodide loading in our study (1.6×10^{-5} wt%) resulted in eluate iodide concentrations that were nearing detection limits (using inductively coupled plasma mass spectrometry [ICP-MS] analysis of stable ^{127}I as a surrogate for ^{129}I). Thus, it should not matter what iodide loading is used in future simulant based Cast Stone leach tests as long as the iodide loading value is kept below ~0.14 wt%. In fact based on our test results it is possible to use initial total iodine loadings below 0.01 wt% but above 5×10^{-5} wt% and still get detectable iodine concentrations in all EPA Method 1315 eluates. The earlier literature, including Lockrem (2005) that suggested that iodide loading does impact cement/grout leach tendencies were performed at total iodine loadings much higher than will ever occur at Hanford using actual liquid waste streams. Although we have not spiked Cast Stone with other possible forms of iodine (e.g., iodate) we do not expect loading impacts for other iodine species as long as low total iodine loadings (below 0.14 wt% and ideally below 0.01 wt%) are used in future Cast Stone formulations. One other key conclusion that results from all the Cast Stone leach testing performed to date at Hanford is that iodide leaches more rapidly than pertechnetate and chromate but at about the same rate as nitrate, nitrite and sodium. We hypothesize that the pertechnetate and chromate lower leach rates are controlled by their reduction by the blast furnace slag (BFS) followed by slow time-dependent re-oxidation to more mobile oxidized forms. The iodide, nitrate, nitrite, and sodium releases are not controlled by a similar redox process.

The leaching results for Tc-bearing monoliths with differing starting technetium species—T4 monoliths with Tc(I)-tricarbonyl gluconate [referred to herein as Tc-Gluc] and T5 monoliths with pertechnetate [TcO_4^-])—suggest that Tc-Gluc monoliths do in fact leach more Tc than T5-pertechnetate monoliths in both VZP and DIW eluents. Whether the Tc in eluates from the Tc-Gluc monoliths remains in a non-pertechnetate form has not been determined because the concentration of total Tc in the Tc-Gluc monolith eluates is orders of magnitude below concentrations needed for direct speciation determinations. At this time a plausible hypothesis for the faster leaching of Tc out of the Tc-Gluc monoliths is that most of the pertechnetate in the companion monoliths was reduced to an insoluble Tc-bearing solid that only leaches out of the monoliths when the Tc-bearing solid is re-oxidized by oxygen ingress into the monolith interior. This re-oxidation process is slow such that the observed diffusion coefficient of Tc in Cast Stone monoliths containing pertechnetate are lower than monoliths containing Tc-gluconate and other soluble Tc(I) species that we hypothesize remain soluble in the Cast Stone pore water and simply diffuse out based on their molecular size. More discussion is found in Section 4.4.

In February 2015, about half the monoliths in the Extended, Archive and Tc-Gluconate Suites of leach tests were put into “wet” storage in a small volume of their final eluate and many are currently undergoing detailed solid phase (post-leaching) characterization to gain information on the mineralogical changes that occur during leaching, and to estimate the rate of oxygen and carbon dioxide ingress into the monoliths’ interiors, which quantifies the re-oxidation and carbonation processes that impact the release of the key groundwater risk driving contaminants. The post-leached monoliths selected for detailed characterization were carefully sectioned into small pieces with known locations as a function of distance from the outer surface using a diamond bladed saw. The sectioning was performed in an anoxic glove bag to minimize exposure to air (re-oxidation concerns) and cutting was performed without water cooling the blade to minimize water leach artifacts. Finally state-of-the-art solid phase characterization instrumentation is being used on pieces with known location to ascertain valence state information on redox sensitive elements (e.g., Tc and Cr) as a function of distance from the monoliths outer surfaces. The goal of all the solid phase characterization activities and eluate analyses will be used to elucidate the weathering reactions caused by the leaching process. All the information is important to improve the technical defensibility of waste form performance in the upcoming 2017 IDF performance assessment (PA). The results of the first solid phase characterization activities will be published in a technical report at the end of FY16. Within the post long-term leached solid phase characterization will be recommendations on whether additional solid phase characterization activities would be useful and how to improve the current long-term contaminant release conceptual models for Cast Stone/grouts that are currently being studied to solidify liquid secondary wastes and/or encapsulate solid secondary wastes for ultimate disposal in the IDF.

This report also discusses recommended follow-on leach testing of some of the monoliths that have been leaching for approximately three years and new types of leach testing that could be useful to improve our understanding of the long-term leach properties of Cast Stone. The need to act on the recommendations for additional experimental studies are predicated on whether the 2017 IDF system performance assessment (PA) shows that Cast Stone contaminant release predictions, based on the available data and conceptual contaminant release models, would benefit from the additional data, in order to show that Cast Stone/ETF grout contaminant release predictions are adequate, i.e., show an acceptable margin of safety and good technical defensibility.

Acknowledgments

This multi-year project was completed as part of the Secondary Waste Cast Stone Formulation and Waste Form Qualification project. Support for this project was provided through Washington River Protection Solutions (WRPS).

The authors wish to acknowledge Dave Swanberg (WRPS) for programmatic guidance, direction, and support. We also acknowledge Dave Swanberg and Elvie Brown(WRPS), Kevin Fox, Christine Langton, Roger Seitz, Greg Flach, and Walt Kubilius (SRNL), John Mahoney and Felix Miera (YAHGS LLC), Bob Andrews and Mick Apted (INTERA), and Wooyong Um, Matt Asmussen, Gary Smith, and Dave MacPherson (PNNL) for their technical review of the document.

The authors also acknowledge the following PNNL staff from the Environmental Sciences Lab, which is part of the Geosciences Group, for performing the leach tests, making all the analytical measurements on the eluates and simulants and performing the characterization studies on secondary minerals that precipitated on the surfaces of the monoliths: SR Baum, RE Clayton, BN Gartman, KJ Geiszler, AR Lawter, C Iovin, and II Leavy.

The authors also wish to acknowledge following PNNL staff—MK Edwards, TC Levitskaia, and BM Rapko - for synthesizing the Tc(I)-tricarbonyl gluconate material used to spike one of the LAW waste simulants in order to evaluate differences in the release of Tc(I) from Tc(VII) species in Cast Stone.

The authors wish to thank Heather Culley, Mary Stover. Veronica Perez, and Chrissy Charron for editorial review and document production. Finally, the authors thank EB Baer, WC Dey, KJ Geiszler, MM Valenta-Snyder, M Asmussen and GD Tartakovsky for performing all the analytical data and calculation QA reviews.

Acronyms and Abbreviations

ANS	American Nuclear Society
ANSI	American National Standards Institute
ASTM	ASTM International (formerly the American Society for Testing and Materials)
atm	atomosheres (unit of pressure)
BET	Brunauer–Emmett–Teller **(**BET**)** theory
BFS	blast furnace slag
bgs	below ground surface (typically ft bgs) to describe location where samples were retrieved from boreholes
CBP	Cementitious Barriers Partnership
CFR	Code of Federal Regulations
COC	constituents or contaminants of concern
COPC	contaminant of potential concern
D_e, or D_{eff}	effective diffusivity (observed diffusion coefficient) per ANSI/ANS-16.1-2003; see Eq. 3.2 in main text for definition
DIW	deionized water
D_{obs}	observed diffusivity (observed diffusion coefficient) per EPA Method 1315; see Eq. 3.1 in main text for definition
DOE	U. S. Department of Energy
DOE-ORP	U. S. Department of Energy Office of River Protection
DST	double-shell tanks
EDS	energy dispersive spectroscopy
EM	DOE Office of Environmental Management
EPA	U. S. Environmental Protection Agency
EQL	estimated quantification limit
eSTOMP	Subsurface Transport Over Multiple Phases (massively parallel version)
ETF	Effluent Treatment Facility
EXAFS	extended X-ray absorption fine structure
HLW	high-level waste
HTWOS	Hanford Tank Waste Operations Simulator (computer code)
IC	ion chromatography
ICP-OES	inductively coupled plasma optical emission spectroscopy
ICP-MS	inductively coupled plasma mass spectrometry
IDF	Integrated Disposal Facility
ILAW	immobilized low-activity waste
k_d	distribution coefficient (measure of sorption)
k_{sp}	solubility product (thermodynamic solubility value)

L	length (when used in units definitions)
LAW	low-activity waste
LI	Leach Index; see Eq. 3.2 in main text for definition
L/S	liquid-to-solid ratio
M	mass (when used in units definitions)
ND	non-detect; not detectable
NMR	nuclear magnetic resonance spectroscopy
OPC	ordinary Portland cement
PA	performance assessment
PNNL	Pacific Northwest National Laboratory
ppm	parts per million (generally mg/L when discussing aqueous concentrations)
PUF	pressurized unsaturated flow
QA	quality assurance
R&D	research and development
RCRA	Resource Conservation and Recovery Act
RPL	Radiochemical Processing Laboratory –PNNL facility
SEM	scanning electron microscopy
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
SST	single-shell tank
TCLP	EPA Toxicity Characteristic Leaching Procedure
TC&WM EIS	Tank Closure and Waste Management Environmental Impact Statement
TEM	transmission electron microscopy
TSPA	total system performance assessment
UTS	universal treatment standards
VZP	Hanford IDF Site vadose zone pore water
WRPS	Washington River Protection Solutions
WTP	Hanford Tank Waste Treatment and Immobilization Plant
WWFTP	WRPS Waste Form Testing Program
XANES	X-ray absorption near edge structure
XAS	X-ray absorption spectroscopy
XRD	X-ray diffraction

Preface

In this document, the term cementitious waste form is used as a generic term for any waste form that is created by mixing pozzolanic dry solids with liquid wastes that upon setting and curing becomes a physically hardened solid that has relatively low permeability and generally low solubility in water. Thus the most likely process controlling release of sequestered contaminants is diffusion.

The term Cast Stone refers to a particular mix of dry solids that are used to solidify various Hanford Site liquid wastes. The dry solids mix is predominately a blend of 8 wt% ordinary Portland cement (OPC), 45% class F fly ash, and 47% blast furnace slag (BFS). The Cast Stone dry blend mix is often modified by including small quantities of water reducing agents, porosity modifiers such as xypex, and “getters” whose function is to improve the sequestration of particular contaminants of concern (COC). The term LAW Cast Stone is used to specify the use of the Cast Stone dry blend mix to solidify low-activity waste (LAW) simulants as opposed to any other Hanford Site liquid waste stream.

In this document the term grout waste form is used to describe the product of mixing secondary liquid wastes from various Hanford Site processes with a dry blend with a different composition than that called Cast Stone. Currently the composition of dry materials in use to solidify Hanford Site secondary liquid wastes that are or will be treated in the ETF consists of a mixture of lime [$Ca(OH)_2$], BFS, and OPC. The ratios of these three dry solids are being varied to find a composition that has good rheologic and COC release properties once the grout has cured/hardened.

Saltstone is the waste form used at the Savannah River Site (SRS) to solidify liquid wastes that originate in SRS storage tanks. The dry blend mix for Saltstone is quite similar to Hanford’s Cast Stone. The Saltstone dry blend consists of 10% OPC, 45% class F fly ash and 45% BFS. The SRS liquid wastes that are being solidified differ somewhat from the liquid LAW that may be solidified in Cast Stone at the Hanford Site.

Two other terms used in this report are eluent and eluate, which in many reports and the ANS16.1 procedure are called leachant and leachate[1]. Eluent is the starting solution used to interact with the waste form monoliths. We use two eluents in this testing, deionized water (DIW) and an IDF vadose zone pore water simulant (VZP). The resultant solutions after contact with the waste form monoliths are called eluates. Eluates contain the species that diffuse or dissolve out of the waste forms. The difference between the concentrations of any constituent in the eluate from a container that had a monolith present and the concentration in the eluate from the blank container (contains only the monolith holder) represents the net concentration released from the monolith.

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[1] Note that Garrabrants et al. (2010) has defined 4 terms that differentiate the solutions that eventually contact the solid as follows:

Term	Definition
leachant	The contacting liquid in the **natural environment** (in the field) before contact with the solid of concern.
leachate	The contacting liquid in the **natural environment** (in the field) after contact with the solid of concern.
eluent	The contacting liquid in the **laboratory** before contact with the solid of concern.
eluate	The contacting liquid in the **laboratory** after contact with the solid of concern.

But, few reports or other leach test procedures make the distinction between leachant and eluent or between leachate and eluate. To remain consistent with EPA-1315 protocol we use eluent and eluate in this report.

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1.0 Introduction

The Hanford Site has approximately 56 million gallons of radioactive waste stored in 177 underground storage tanks. The Hanford Tank Waste Treatment and Immobilization Plant (WTP) currently under construction includes a pretreatment facility to separate the waste into a small volume of high-level waste (HLW) containing most of the radioactivity and a larger volume of low-activity waste (LAW) containing most of the nonradioactive chemicals. The HLW will be converted to glass in the HLW vitrification facility for ultimate disposal at an offsite Federal repository. Through the treatment and vitrification processes at the WTP, aqueous secondary waste streams will be generated that will be treated and solidified outside the WTP at the Effluent Treatment Facility (ETF). Current plans are to solidify the treated secondary wastes in a cementitious waste form that will subsequently be disposed in the Integrated Disposal Facility (IDF) on the Hanford Site. Washington River Protection Solutions (WRPS) and its contractors at Pacific Northwest National Laboratory (PNNL) and Savannah River National Laboratory (SRNL) are conducting a development program to develop and refine the cementitious waste form for the waste treated at the ETF, as well as to provide the data needed to support the IDF performance assessment (PA).

At least a portion (~35%) of the LAW will be converted to glass in the LAW vitrification facility and will be disposed of onsite at the IDF. The pretreatment and HLW vitrification facilities will have the capacity to treat and immobilize the waste destined for each facility. However, a second LAW immobilization facility will be needed for the expected volume of LAW requiring immobilization. One supplemental waste form called Cast Stone is being considered as an option to solidify the excess LAW. Further, a waste form called Saltstone that is used at the Savannah River Site to solidify their LAW tank wastes is nearly identical in dry blend composition to the Hanford Cast Stone that is being studied herein as a supplemental LAW waste form. The grout that is currently being studied for solidification of WTP aqueous secondary wastes treated at the ETF at Hanford uses a modified dry blend containing more cement, hydrated lime, and no fly ash, but most COPC leach results are similar to those found for Cast Stone, especially for mobile constituents such as sodium and nitrate.

Cast Stone was originally developed as one of three supplemental immobilization technologies as part of a mission acceleration initiative to provide the needed capacity to complete the Hanford tank waste cleanup mission in a timely manner (Raymond et al. 2004). The three technologies were containerized grout (now being called Cast Stone), bulk vitrification, and fluidized bed steam reforming. It was noted that limited data were available on Cast Stone for the immobilization of Hanford LAW. This lack of data was the impetus for initiating the Cast Stone development program for Hanford LAW.

1.1 Objectives of Cast Stone Development Program

The initial objectives of the Cast Stone development program work were to:

1. determine an acceptable formulation for the LAW Cast Stone waste form,
2. evaluate sources of dry materials for preparing the LAW Cast Stone,
3. demonstrate the robustness of the waste form for a range of LAW compositions and for variability in the Cast Stone process, and
4. provide Cast Stone contaminant release data that could be used in PA and risk assessment evaluations.

The Cast Stone development program for immobilization of Hanford LAW has been underway since late 2012. About the same time Cast Stone was selected as the preferred waste form for future WTP aqueous secondary wastes at the Hanford Site, which will be sent to the ETF for treatment and solidification. Since then a grout, whose dry blend consists of a mixture of lime, BFS, and fly ash, is currently being developed specifically to address elevated sulfate in aqueous wastes to be treated in the ETF. Enhancements to the Cast Stone formulation are being pursued to further improve retention of key risk driver contaminants of concern (COC) such that it could reasonably be considered as a supplemental immobilization technology for Hanford LAW.

Both grout waste forms, secondary and, if selected, supplemental LAW, must be acceptable for disposal in the IDF. Each Cast Stone/grout waste form and immobilization process must be tested to demonstrate that the final cementitious waste form can comply with waste acceptance criteria for the IDF. That is, it must be demonstrated that any Cast Stone or grout waste form can meet the waste acceptance criteria of the selected disposal facility for any application, and that the process can be controlled to consistently provide an acceptable waste form product. Further, the waste form must be tested to provide the technical basis for understanding the long-term performance of the waste form in the disposal environment. These waste form performance data are needed to support risk assessment and PA analyses of the long-term environmental impact of the waste disposal in the IDF. The PA is needed to satisfy both Washington State IDF Permit and U.S. Department of Energy (DOE) Order requirements. More details on the IDF and the history of IDF PAs and future refinements can be found in a recently published document (Yabusaki et al. 2015) called "Technical Approach for Determining Key Parameters Needed for Modeling the Performance of Cast Stone for the Integrated Disposal Facility Performance Assessment."

Activities that support the IDF PA will require a long-term testing program. Elements of the testing program include measuring release rates for key risk driver contaminants over long periods of time, developing an understanding of the long-term evolution or weathering of Cast Stone/grout in the disposal environment, developing an understanding of the mechanism by which radionuclides and other contaminants are retained in the Cast Stone/grout and the mechanism(s) of release, developing accelerated test methods and other test methods to characterize and predict the long-term performance of the Cast Stone/grout, and characterizing transport properties of the key contaminants solidified in Cast Stone/grout as water migrates through the IDF and the disposal packages contained within.

To support Cast Stone or grout performance testing, WRPS has contracted with several organizations, including PNNL, to conduct a Cast Stone testing program with several objectives. One of the first reports on this work that included monolith leaching results was called the "Screening Test Report" authored by staff at WRPS, PNNL, and SRNL (Westsik et al. 2013a).

The screening tests were performed to evaluate the effects of key parameters on the properties of the Cast Stone as it is initially prepared and after curing. The test parameters and their ranges that were investigated in the screening tests included simulants representing a range of LAW compositions (Average, single-shell tank [SST] Blend, High Al, and High SO_4 [all defined in Section 3.0 of Westsik et al. 2013a]), waste concentrations (5 M and 7.8 M Na), Class F fly ash source (NW = High Ca, SE = Low Ca)[1], BFS source (NW, SE), and free-water-to-dry-blend solids mix ratio (0.4, 0.6).

A total of 26 different Cast Stone mixes were prepared, and both wet slurry and cured properties were characterized in detail. Key findings from the screening tests were that no bleed/free water was found in 24 of the 26 mixes after curing, compressive strength of 28-d cured specimens for the 26 mixes were

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[1] The Class F fly ash included a relatively high-Ca content material available in the Pacific Northwest (designated NW) and a lower Ca content material available in the southeastern (designated SE) part of the country. Blast furnace slags from the northwest (designated NW) and southeast (designated SE) were also selected for the screening tests.

above 500 psi (3.45 MPa), all mixes met Universal Treatment Standards in 40 CFR 268 for land disposal restrictions, and leachability indices (LIs) for each of the constituents studied (nitrate, nitrite, chromium, technetium, uranium, and iodide) were not statistically different for the 24 mixes deemed acceptable. That is, the cured Cast Stone monoliths that were leach tested did not appear to be overly sensitive to the simulant compositions and total solute concentrations, or to the source of the dry blend ingredients, or to the free-water-to- dry blend ratio. This suggests that Cast Stone can be used to solidify a large range of waste compositions over a significant range of waste loadings while maintaining good slurry “processability” attributes. Whether the measured leaching properties of the key risk driving constituents are low enough to result in acceptable risks to the general public and environment after disposal in the IDF will require long-term predictive modeling to be performed on the whole IDF system.

1.2 Objectives of This Task

Specific objectives of the Cast Stone tests described in this report (both Rev. 0 and Rev. 1) focus on four activities:

1. Extend the leaching times for selected ongoing EPA-1315 tests on Cast Stone monoliths made with LAW simulants beyond the conventional 63-day time period in an effort to evaluate long-term leaching properties of Cast Stone to support future PA activities.

2. Start new EPA-1315 leach tests on archived Cast Stone monoliths made with four LAW simulants using two eluents (deionized water [DIW] and simulated Hanford IDF Site vadose zone pore water [VZP]) that will be continued long-term.

3. Evaluate the impacts of varying the iodide loading (starting iodide concentrations in one LAW simulant, 7.8 M Na average simulant) by manufacturing additional Cast Stone monoliths and repeating the EPA-1315 leach tests using DIW and the IDF Site VZP as eluents, leaching for long time periods.

4. Evaluate the impacts of using a non-pertechnetate form of Tc that is present in some Hanford tanks. In this activity, the LAW 7.8 M Na average simulant was spiked with a Tc(I) tricarbonyl gluconate species that was solidified into Cast Stone monoliths and subsequently leached using the EPA-1315 leach protocol with DIW and IDF Site VZP as the eluents. The leach results for Cast Stone monoliths using the LAW simulant but with the Tc(I)-gluconate spike are compared to Cast Stone monoliths made with the same LAW simulant spiked with pertechnetate at nearly the same initial concentration as the Tc(I)-gluconate, which were leached in both deionized and IDF Site VZP for long periods.

One concern with the iodide-bearing Cast Stone leach testing performed to date (e.g., Westsik et al. 2013a) is that, in order to have measurable concentrations of iodine in the eluates, the iodine concentration in the Cast Stone specimens were spiked up to two orders of magnitude higher than the projected average concentration in the LAW waste.

A second concern is that all the previous Cast Stone leach tests were performed using DIW as the eluent. DIW is generally considered an aggressive eluent that does not purport to represent the fluids that will interact with buried wastes within the IDF facility. Thus, to more realistically evaluate Cast Stone leach properties an IDF relevant eluate, simulated Hanford formation VZP was used in many of the leach tests described in this progress report.

1.3 Report Organization

Rev. 0 of this report extended the leach tests on many of the Cast Stone monoliths described in Westsik et al. (2013a) and on new Cast Stone monoliths made with the 7.8 M Na Hanford Tank Waste Operations Simulator (HTWOS) average simulant. In Rev. 0 Appendix C, one will find tables with corrected D_e values for some of the data presented in Westsik et al. (2013a) Appendix D that were found to be in error after its publication. The corrected D_e values do not impact any of the conclusions in Westsik et al. (2013a). Details on the errors and their impacts are discussed in Appendix C of Rev. 0. In Rev. 1 of this, this report, the Westsik et al. (2013a) correction tables are deleted and replaced by graphics showing details on the long-term leach tests reported in Rev. 1.

Each of the Extended Leach Task objectives and the results are discussed in more detail in Sections 4.0. Section 2.0 describes the LAW simulant and VZP recipes, and Cast Stone monolith preparation details. Section 3.0 describes the leach test methodology and terminology to describe various types of diffusion and symbols used to describe the various types of diffusion. Section 5.0 summarizes the key findings and describes some future studies that would be useful to gain more understanding on Cast Stone long-term performance. Section 6.0 lists the references cited in the report. Appendix A contains details on the original 26 Cast Stone mixes used in Westsik et al. (2013a) and details on the LAW and VZP simulants used in this report. Appendix B contains tables of the interval observed diffusion coefficients for Na, nitrate, nitrite, iodide, Tc and Cr, cumulative fractions leached as a function of leach time and the pH and electrical conductivity of the eluates from all four Suites on monoliths as a function of cumulative leach time. Appendix C provides additional graphics on selected leach tests that allow several questions to be posed on how to interpret whether the long-term leach results abide by various researcher’s criteria to call the data to be controlled by diffusion.

1.4 Quality Assurance

The multi-year work described in Revision 0 and Revision 1 of this report was conducted with funding from two sources, WRPS and DOE Headquarters Office of Environmental Management (EM)-31, under several contract releases as follows:

Preparation of the Archived Suite of monoliths and preparation and leach testing, through the first 91 days, of the Extended Suite of monoliths was funded under PNNL project 62745 and described in test plan TP-62745-001. This work was conducted under the WRPS Waste Form Testing Program Quality Assurance Plan (QA-WWFTP-001, Rev. 0, NQA-1-2000 compliant program).

The leach testing of the Archived Suite of monoliths and the leach testing of the Extended Suite of monoliths beyond 91 days was funded from the DOE EM-31 Support Program (EMSP) “Production and Long-term Performance of Low Temperature Waste Forms” and described in test plan TP-EMSP-0011. This work was conducted under the Environmental Management Support Program Quality Assurance Plan (QA-EMSP-001, Rev. 0, NQA-1-2000 compliant program). The leach testing continued to September 2013.

The preparation and leach testing of the newer Iodide and Tc-Gluconate Suites of monoliths was funded from the DOE EM-31 Support Program (EMSP) “Production and Long-term Performance of Low Temperature Waste Forms” and described in test plan TP-EMSP-0011. This work was conducted under the Environmental Management Support Program Quality Assurance Plan (QA-EMSP-001, Rev. 0, NQA-1-2000 compliant program). The leach testing continued to September 2013.

In October 2013, all of the identified leach testing continued with funding from WRPS under PNNL project 66596 and described in test plans TP-66596-001, TP-SLAW-002, and TP-SLAW-003. This work was conducted under the WRPS Waste Form Testing Program Quality Assurance Plan (QA-WWFTP-001, Rev. 1, NQA-1-2000 compliant program).

In October 2014 funding for the leach testing was transferred to PNNL project 68334 and described by test plan TP-SWCS-010. Performance of this work and the preparation of this report were conducted under the WRPS Waste Form Testing Program Quality Assurance Plan (QA-WWFTP-001, Rev. 2, NQA-1-2008/2009a compliant program).

All research and development (R&D) work at PNNL is performed in accordance with PNNL's Laboratory-level Quality Management Program, which is based on a graded application of NQA-1-2000, Quality Assurance Requirements for Nuclear Facility Applications, to R&D activities. In addition to the PNNL-wide quality assurance (QA) controls, the QA controls of the WRPS Waste Form Testing Program (WWFTP) QA program were also implemented for the work. The WWFTP QA program consists of the WRPS Waste Form Testing Program Quality Assurance Plan (QA-WWFTP-001) and associated QA-NSLW-numbered procedures that provide detailed instructions for implementing NQA-1 requirements for R&D work. The WWFTP QA program is based on the requirements of NQA-1-2008, Quality Assurance Requirements for Nuclear Facility Applications, and NQA-1a-2009, Addenda to ASME NQA-1-2008 Quality Assurance Requirements for Nuclear Facility Applications, graded on the approach presented in NQA-1-2008, Part IV, Subpart 4.2, "Guidance on Graded Application of Quality Assurance (QA) for Nuclear-Related Research and Development".

Performance of this work and preparation of this report were assigned the technology level "Applied Research" and were conducted in accordance with procedure QA-NSLW-1102, Scientific Investigation for Applied Research. All staff members contributing to the work have technical expertise in the subject matter and received QA training prior to performing quality-affecting work. The "Applied Research" technology level provides adequate controls to ensure that the activities were performed correctly. Use of both the PNNL-wide and WWFTP QA controls ensured that all client QA expectations were addressed in performing the work.

Records of all of the experimental work will be filed with the records for the Supplemental Immobilization of Hanford LAW project and Secondary Waste Cast Stone (PNNL projects 66596 and 66334, respectively).

1.5 Relationship to Recent Grouted Secondary Waste Data Packages

The observed diffusion coefficients, calculated from the EPA Method 1315 tests extended to long leach times, tabulated in this Rev. 1 report, are for Cast Stone monoliths that used LAW simulants. The two recent IDF waste form data packages, Cantrell et al. (2016) and Flach et al. (2016), focus on cementitious waste forms that will be used to solidify liquid and solid secondary wastes, respectively, to be generated during future operation of the WTP. The LAW waste stream simulants solidified in Cast Stone are 5–7.5 M sodium solutions (25–35 wt% solids, pH >14) with nitrate and hydroxide as the major anions. The current liquid secondary waste (LSW) waste stream simulants are primarily ammonium sulfate (10–20% solids) and are near neutral pH. Therefore, the LSW simulants are being solidified in a different mix of dry ingredients than Cast Stone, and the subsequent hydration, weathering, and leaching reactions that occur may vary. The projected ETF LSW composition differs from LAW waste simulants solidified in Cast Stone in that it contains lower free hydroxide and higher sulfate concentrations. Accordingly, modifications to the Cast Stone dry blend of 47 wt% BFS/45 wt% fly ash/8 wt% cement formulation are being investigated to increase calcium to bind the sulfate and raise the pH in order to activate the BFS.

The modified dry blends include significant portions of hydrated lime, increased portions of cement, similar portions of BFS, and no fly ash (see Um et al. 2016 for more details). The observed diffusion coefficient values tabulated in this Rev. 1 report are similar to those found in the last columns in Table 3.1 in Cantrell et al. (2016), which lists the geometric mean and low to high range values for the 26 Cast Stone mixes made with four different LAW simulants. However, given the difference in both the liquid waste simulants and the modified dry blend composition, the D_{obs} reported in this Rev. 1 report should not be expected to be quantitatively identical to those for hydrated lime based solidified secondary liquid wastes (values shown in the first several columns in Table 3.1 of Cantrell et al. The same is true for observed diffusion's tables found in Section 8 of Cantrell et al.

On the other hand, in general, the numerical values for observed diffusion coefficients for the five COPCs most often of interest (Tc, I, Cr, nitrate, and nitrite) do qualitatively show the same trends amongst the three waste forms (Cast Stone solidified LAW liquid simulants, hydrated lime-based grout solidified ETF-LSW simulants, and cement/grout solidified secondary solid wastes).

When reviewing the diffusion coefficient tables in the two data packages, Cantrell et al. (2016) and Flach et al. (2016) Section 8, bear in mind that the former data package uses the term effective diffusion coefficient and the latter data package uses the term apparent diffusion coefficient. All three terms (observed, effective, and apparent) are referring to the same diffusion coefficient—a coefficient that includes impacts of all physical and chemical processes that influence the release of a COPC from the porous waste form.

In summary, the D_{obs} values reported in this Rev. 1 report for each COPC that originated in LAW simulants that were solidified in Cast Stone are similar to the values reported in the two data packages just published that focus on secondary waste streams (both liquid and solid) that will be solidified/encapsulated into cementitious/grout waste forms destined for burial in the IDF. Similarly, in general, we mean the D_{obs} values agree within an order of magnitude in units of cm^2/s.

2.0 LAW Waste Simulants, Cast Stone Monolith Preparation, and Description of the Vadose Zone Pore Water Simulant

2.1 LAW Waste Simulants

This section summarizes the LAW waste simulants, dry blend components, and slurry mixing details used in the original screening studies and also followed in this new work using the one LAW simulant -7.8 M Na Avg. More details are found in the first technical report produced on the WRPS Supplemental Immobilization of the Hanford Low-Activity Waste project (Westsik et al. 2013a).

The four liquid LAW simulants, discussed in Section 3.0 of Westsik et al. (2013a), were prepared. The Na concentrations of 5 M and 7.8 M were selected to represent a range of possible LAW waste concentrations that will be processed through the WTP. Table 2.1 shows the normalized concentrations (per M of sodium) for the major constituents for the four LAW simulants. The simulants were given generic names (SST Saltcake Blend, HTWOS[1] Overall Average, HTWOS High Al, and HTWOS High SO_4), which represent a wide range of the wastes expected to be processed. More details on selecting these four waste compositions are found in Westsik et al. (2013a) and details on preparing the simulants are found in Russell et al. (2013). The four LAW simulants were spiked with the hazardous metals, radionuclides, and stable iodide as shown in Table 2.2. Other Resource Conservation and Recovery Act (RCRA) and hazardous metals **not** spiked into the simulants included As, Ba, Hg, Se, Ag, Sb, Be, and Th because their projected maximum concentrations in LAW wastes are low enough that even if 100% of their mass leached out of Cast Stone during the Toxicity Characteristic Leaching Procedure (TCLP) testing, the resultant concentrations would not exceed allowable universal treatment standards (UTS).

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[1] HTWOS= Hanford Tank Waste Operations Simulator (a computer program that predicts tank waste concentrations from waste retrieval through WTP treatment and immobilization).

Table 2.1. LAW Simulants for Cast Stone Screening Tests

Waste Constituent	SST Blend Saltcake	HTWOS Overall Avg.	HTWOS High Al	HTWOS High SO_4
		Concentration (moles/mole Na)(a)		
Na	1.000	1.000	1.000	1.000
K	0.002	0.007	0.028	-
Al	0.013	0.061	0.112	0.047
Cl	0.009	0.008	0.018	0.007
F	0.006	0.006(b)	0.010	0.012(b)
SO_4	0.018	0.017	0.004	0.030
PO_4	0.010	0.010(b)	0.005	0.010(b)
NO_2	0.085	0.113	0.194	0.098
NO_3	0.502	0.324	0.287	0.367
CO_3	0.095	0.055	0.040	0.035
TOC Total	0.057	0.015	0.021	0.007
Free OH	0.097	0.312	0.293	0.306

(a) After charge balancing.

(b) Concentration of F and PO_4^{3-} reduced from HTWOS values because of solids formation observed in preliminary simulants.

Table 2.2. Spike Levels for Hazardous Metals and Radionuclides in Simulants for Cast Stone Screening Tests

Waste Constituent	HTWOS Overall Avg.	HTWOS Maximum	Other Considerations
Metals	moles/mole Na	moles/mole Na	moles/mole Na
Cd(II)	2.78E−06	3.19E−05	-
Cr(VI)	2.42E−03	9.99E−03	4.30E−03(a)
Pb(II)	1.16E−05	5.13E−05	-
Ni(II)	6.41E−05	6.61E−04	-
Radionuclides	Ci/mole Na	Ci/mole Na	Ci/mole Na
^{99}Tc	1.13E−05	4.13E−05	-
^{99}Tc(c)	(6.65E+02 µg/mole Na)	(2.43E+03 µg/mole Na)	added as pertechnetate Tc(VII)
^{129}I	1.44E−08	8.01E−08	3.54E−06(b)
^{127}I (stable)(c)	(8.14E+01 µg/mole Na)	(4.53E+02 µg/mole Na)	(2.00E+04 µg/mole Na)
Natural U(c)	-	(3.56+04 µg/mole Na)	-

(a) Cr concentration adjusted based on review of best basis inventory and previous simulant work.

(b) Iodine concentration increased to address possible detection limits issues in waste form leach tests. Iodine added as non-radioactive ^{127}I in iodide form.

(c) These COCs were added to simulants based on mass (shown in bold font).

The 7.8 M Na HTWOS Average simulant was chosen to make new Cast Stone monoliths to gather data on objectives 3 and 4; the effect of varying iodide concentrations and the comparison of leaching properties for Cast Stone prepared with the two Tc species. The new batch of 7.8 M Na Average simulant was prepared in early June 2013 following the recipe documented in Russell et al. (2013). Details on the chemical reagents used to prepare 4.5 liters of the simulant are provided in Table 2.3, and notes on its preparation are found in Table A.1. The reagents were added to DIW in the order shown, and the final volume brought to 4.5 L using DIW after heating the mixture for several hours at 90°C to promote complete dissolution. During the chemical reagent additions, some promoted vigorous reactions and some chemicals, such as the nickel nitrate, did not completely dissolve. After cooling the final simulant to room temperature, it exhibited a yellow color (from the sodium dichromate added) with a small amount of precipitates, as shown in Figure 2.1. The final specific density of the new simulant was 1.336 g/cm^3 compared to 1.343 g/cm^3in the batch made by Russell et al. (2013). An aliquot of the small amount of precipitate in the new simulant was filtered and characterized by X-ray diffraction (XRD); however, the only crystalline peak identified was nitratine ($NaNO_3$), which likely was residual simulant that evaporated during drying the material on the XRD slide. There were a few unidentified XRD peaks in the scan shown in Figure A.2. The remaining precipitates in the simulant were left and incorporated into the Cast Stone similar to the way residual solids in simulants were handled in Westsik et al. (2013a).

Table 2.3. Masses of Chemical Reagents Used to Make 4.5 L of 7.8M Na Average Simulant

Chemical Reagent (added in order listed)	**MW (g)**	**Amount Added (g)**
$Al(NO_3)_3•9H_2O$	375.13	**807.93**
KNO_3	101.11	**23.247**
$NaNO_2$	69	**273.61**
$NaNO_3$	85	**399.141**
$Na_3PO_4 •12H_2O$	380.12	**131.292**
Na_2SO_4	142.04	**85.266**
Na_2CO_3	105.99	**203.98**
NaF	41.99	**9.316**
NaCl	58.44	**17.307**
NaOH (50% wt. soln)	(50% wt soln)	**1565.15**
$NaC_2H_3O_2$	82.04	**22.084**
$Na_2Cr_2O_7•2H_20$	298.05	**22.317**
$Pb(NO_3)_2$	331.23	**0.595**
$Ni(NO_3)_2•6H_20$	290.8	**6.7**
$Cd(NO_3)_2•4H_20$	308.49	**0.341**

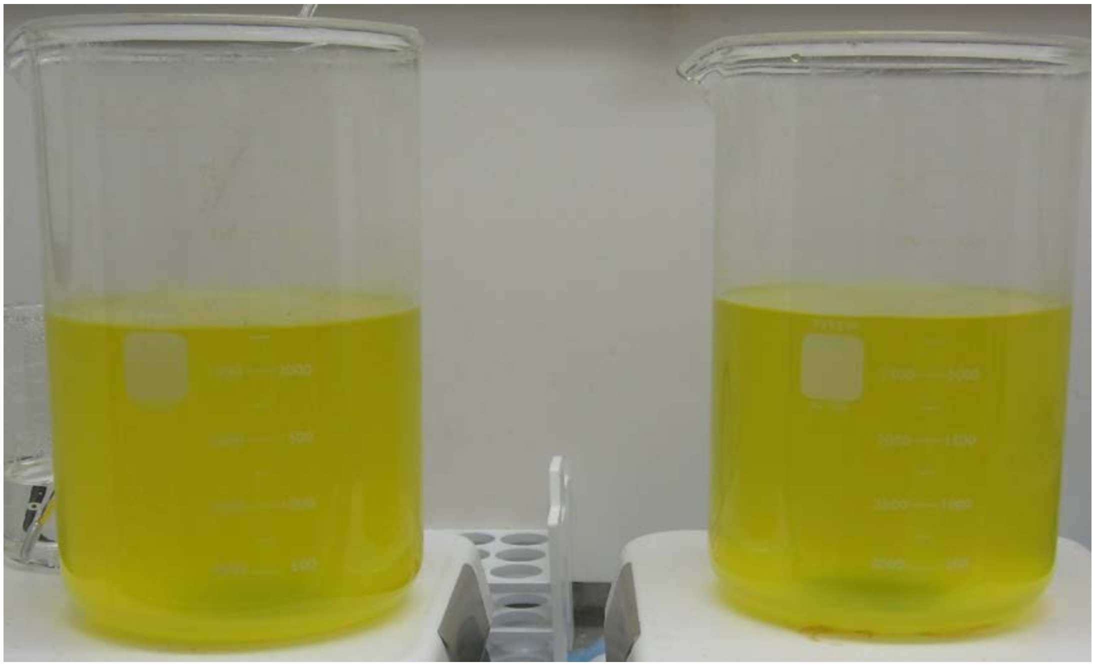

Figure 2.1. 7.8 M Na Average Simulant

The 4.5L batch of new simulant was split into five fractions and three were spiked with stable iodide at three different concentrations and two fractions were spiked with different forms of ^{99}Tc, as shown in Table 2.4. Reagent grade NaI was used to spike the three fractions of simulant used to investigate the impact of having different starting iodide concentrations in the cured Cast Stone. The three iodide-spiked simulants were labeled “Low I”, medium or “Med I,” and “High I”. Originally the three iodide fractions were also spiked with the short-lived radiotracer ^{125}I to allow sensitive and precise gamma counting to be performed. Unfortunately, soon after starting the leach tests, the auto-gamma counter that was being used broke and was not repairable. Therefore, the leach testing proceeded with only stable ^{127}I being measured via ICP-MS. Because the amounts of NaI reagent added to the simulant was very small for the “Low I” and “Med I” aliquots (0.5 mg and 4.5 mg, respectively), we elected to directly measure the starting iodide concentration in the simulants instead of relying on the mass of reagents used to make up the simulants. The latter method (using the mass of a constituent in the chemical reagents and Tc-spike stock solution used to make up the simulant), was used to calculate the starting inventory of each constituent of interest in the original screening test report, Westsik et al. (2013a), as well as in this report, with the exception of for iodide and ^{99}Tc. As mentioned, we made direct ICP-MS measurements of ^{127}I and ^{99}Tc in the simulant before mixing with the Cast Stone dry blend to calculate the starting concentration in our newly prepared and cured monoliths, which were used in the I-Loading and Tc-Gluconate Suite tests.

The ^{99}Tc spikes used in the other two aliquots of simulant were a mixture on Tc(I)-tricarbonyl species[1] representative of a reduced form of Tc thought to be present in some Hanford storage tanks and for the second aliquot, the more common oxidized Tc(VII) species, pertechnetate. Pertechnetate is the form of Tc found in most Hanford waste streams and in the Hanford subsurface environment below cribs, trenches, and tanks that released Tc to the environment as well as the groundwater. All past Cast Stone mixes containing Tc had the pertechnetate species spiked as either $Na^{99}TcO_4$ or $NH_4{}^{99}TcO_4$ dissolved in dilute base.

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[1] The non-pertechnetate species (i.e., Tc(I) in the spike were estimated using NMR to be a mixture of 50% of $Tc(CO)_3$Gluconate, 45% $Tc(CO)_3(OH)(H_2O)_2$, and 5% TcO_4^- dissolved in 0.75 M NaOH.

The non-pertechnetate spike was synthesized by PNNL staff in the Radiochemical Processing Laboratory (RPL). After some difficulty in getting good yield and a pure form, we received ~7.6 mg of mixed non-pertechnetate in ~7 mLs of 0.75 M NaOH solution. The 7.6 mg Tc value was measured by beta counting (liquid scintillation). Because the RPL staff creating the non-pertechnetate spike had difficulties making adequate spike, we got the non-pertechnetate on the very same day as we were preparing the Cast Stone monoliths. Thus, we had no time to verify the concentration of total ^{99}Tc in the non-pertechnetate spike, and therefore, we elected to directly measure the ^{99}Tc, using ICP-MS, after spiking the non-pertechnetate into 500 mL of the 7.8 M Na Average simulant. The final 500-mL aliquot of 7.8 M Na Average simulant was spiked with 0.761 mLs of 9987 µg ^{99}Tc /mL stock pertechnetate solution. The ^{99}Tc concentration in the mixed simulant was measured directly by ICP-MS. Thus, for all the Cast Stone monoliths prepared with the new simulant, the starting concentrations of the iodide and ^{99}Tc were based on direct measurements of concentrations in the simulant, while the starting concentrations of Na, Cr, nitrate, and nitrite were based on the total mass of each of these constituents in the reagents used to make the simulant. No U was added to the new Cast Stone monoliths made in 2013.

Table 2.4. Details on Iodide and Technetium Spikes

Simulant Name	Analyte	Result (µg/L)	EQL (µg/L)
Low I	Iodine 127	322	100
Med I	Iodine 127	3230	100
High I	Iodine 127	6030	100
Tc(I) species; herein called Tc- gluconate	Technetium-99	6710	410
Pertechnetate-Tc(VII)	Technetium-99	16000	410
Iodide added as NaI solids; Tc(I) was a mixture of 50% of $Tc(CO)_3$Gluconate, 45% $Tc(CO)_3(OH)(H_2O)_2$, and 5% TcO_4^- Dissolved in 0.75 M NaOH. Tc (VII) was NH_4TcO_4 dissolved in dilute NaOH.			

2.2 Cast Stone Monolith Preparation

These five spiked simulants shown in Table 2.4 were solidified by mixing in the standard Cast Stone dry blend, which consists of three materials, 47 wt% blast furnace slag (BFS), 45 wt% Class F fly ash, and 8 wt% ordinary Portland cement (OPC, Type I/II). This dry blend mixture is identical to the Cast Stone dry blend used in all recent Cast Stone studies (Westsik et al. 2013a; Mattigod et al. 2011; Sundaram et al. 2011). Further, the source of the BFS was from the southeast and the source for the fly ash was from the northwest. The same source of Portland cement from the northwest was used for the new Cast Stone as in the previous screening. See Westsik et al. (2013a) Section 4 for complete details and characterization of the dry materials. The new monoliths prepared for testing objectives 3 (iodide loading effects) and 4 (Tc speciation effects) were made with the 7.8 M Na Average simulant and Caste Stone dry blend at a free-water to dry blend ratio of 0.6. All the newly prepared (June 2013) monoliths thus have the same composition as the original screening test mixes #3 and #22 as shown in Table A.1.

The same mixing procedure, equipment, and lab staff used in screening tests were again used to make the new Cast Stone monoliths. The same size monoliths (2-in diameter by 4-in length; right cylinders) were prepared and cured at room temperature under high relative humidity, again following the Screening Test activities as close as possible. Figure 2.2 through Figure 2.5 show photographs of some of the key Cast Stone monolith preparation steps. Figure 2.2 shows the specially fabricated (at SRNL) mixer blade that optimally mixes the wet slurry within 15 minutes. Figure 2.3 shows the mixer motor (Caframo model

BDC3030 overhead mixer) and mixer blade shaft in the slurry mixing beaker. The size of the plastic beaker was chosen to be approximately twice the volume of the mix being prepared. Figure 2.4 is a close up photograph of wet slurry after 15 minutes of mixing just prior to pouring into the curing molds. Figure 2.5 is a photograph of one of the curing molds (just prior to being placed on curing stands in 5-gallon buckets partially filled with water) and the curing container just prior to placing an air tight lid on top to maintain high relative humidity. [1]Each of the monolith molds had the plastic cap perforated (several slashes through the cap) to allow moist air to interact with the monolith top surface during the curing step. After curing for between 29 and 33 days, the monoliths from the 26 mixes were removed from the 5-gallon buckets, removed from their molds, inspected for cracks, inspected for bleed water (none found), and the diameters and lengths were measured with calipers and cured weights were recorded. After the physical measurements, 2 of the 6 monoliths from each of the 26 mixes were used in EPA Method 1315 leach testing and the remaining 4 monoliths from each batch were placed in plastic bags. Each plastic bag, containing one or two monoliths from the same mix, was placed within another zip-lock plastic bag that also contained a wet paper towel. The inner plastic bag containing monoliths was open, but the outer plastic bag was closed to prevent the wet paper towel from drying out. The double plastic bags, with the inner bag containing a monolith, were stored at room temperature until they were used in the leach tests identified herein as the Archived Suite. Periodically during the storage phase, the wet towels in the outer plastic bags were inspected to ensure that they remained wet. If necessary, the paper towels were re-wetted to maintain a relative humidity near 100%. Figure 2.6 shows the double bag monolith storage method. More details on Cast Stone monolith preparation, curing, and storage are found in Westsik et al. (2013a) Section 5. The newly made monoliths, used in the Iodide Loading and Tc-Gluconate Suites, were prepared and cured in an identical fashion. These newly made monoliths were cured for 29 days (Iodide Loading) and 33 days (Tc-Gluconate), respectively. Then the monoliths had their physical dimensions and cured weights measured and then were immediately used in EPA Method 1315 leach tests.

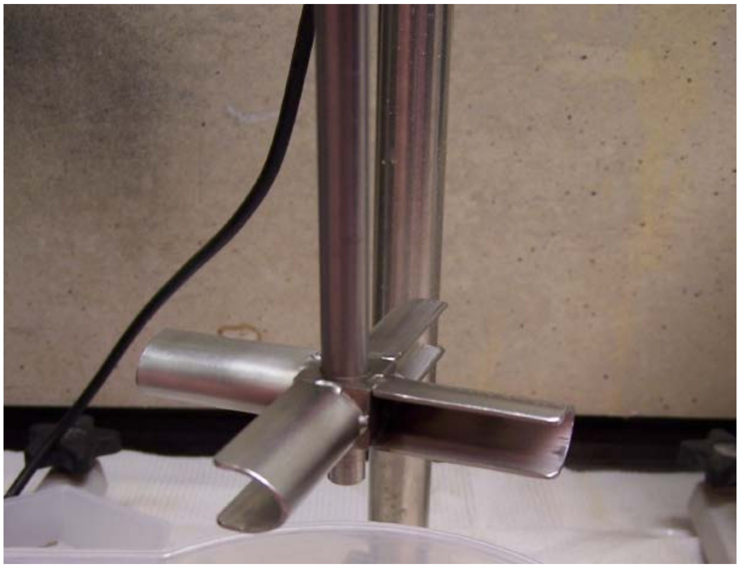

Figure 2.2. Mixer Blade on End of Mixer Shaft (Direction of Rotation is Clockwise)

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[1] In recent grout monolith testing, the relative humidity of the curing containers and individual plastic bags containing wet towels and curing monoliths have been monitored with a hygrometer and found to range between 85 to 100% relative humidity, depending on the wetness of the towel in the outer plastic bag.

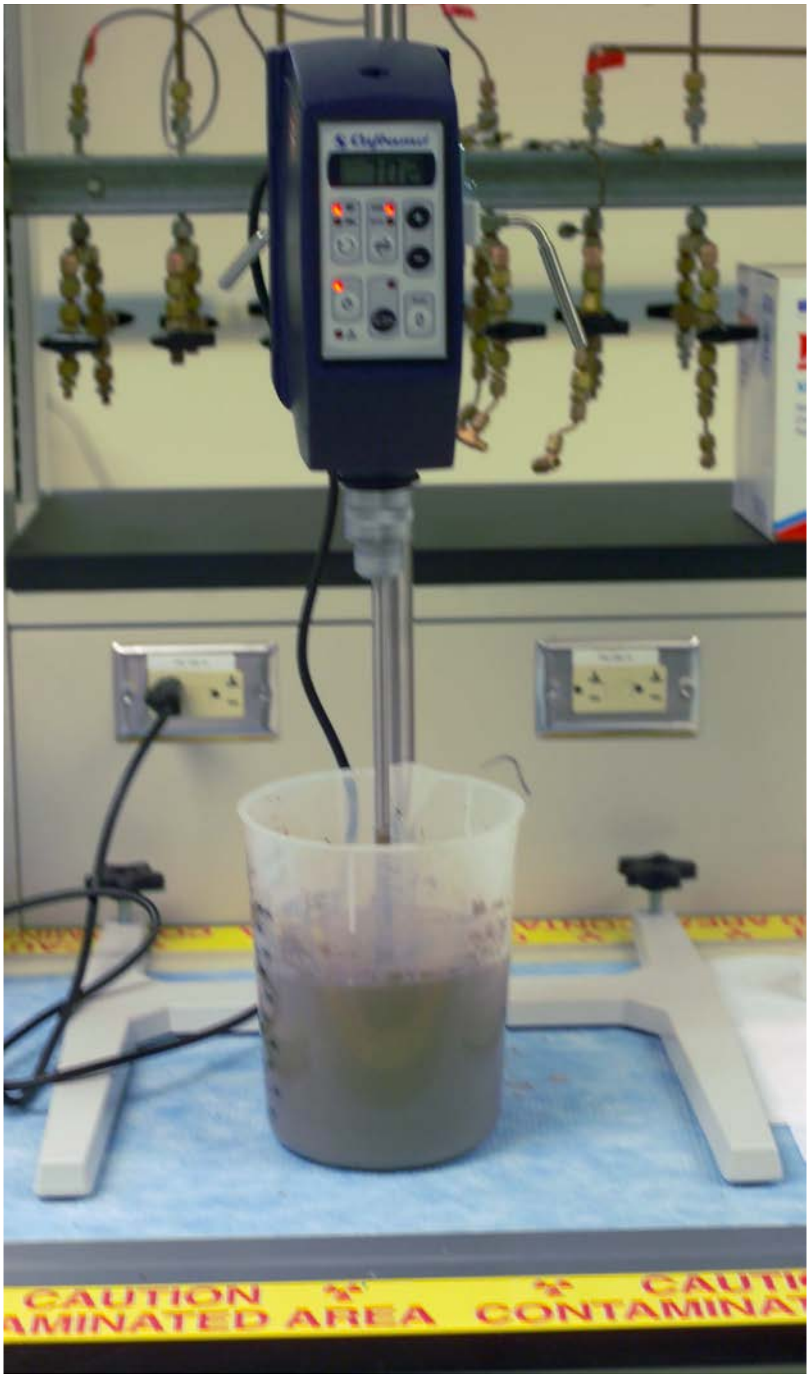

Figure 2.3. Mixer Pump & Assembly Immersed in Mixing Container

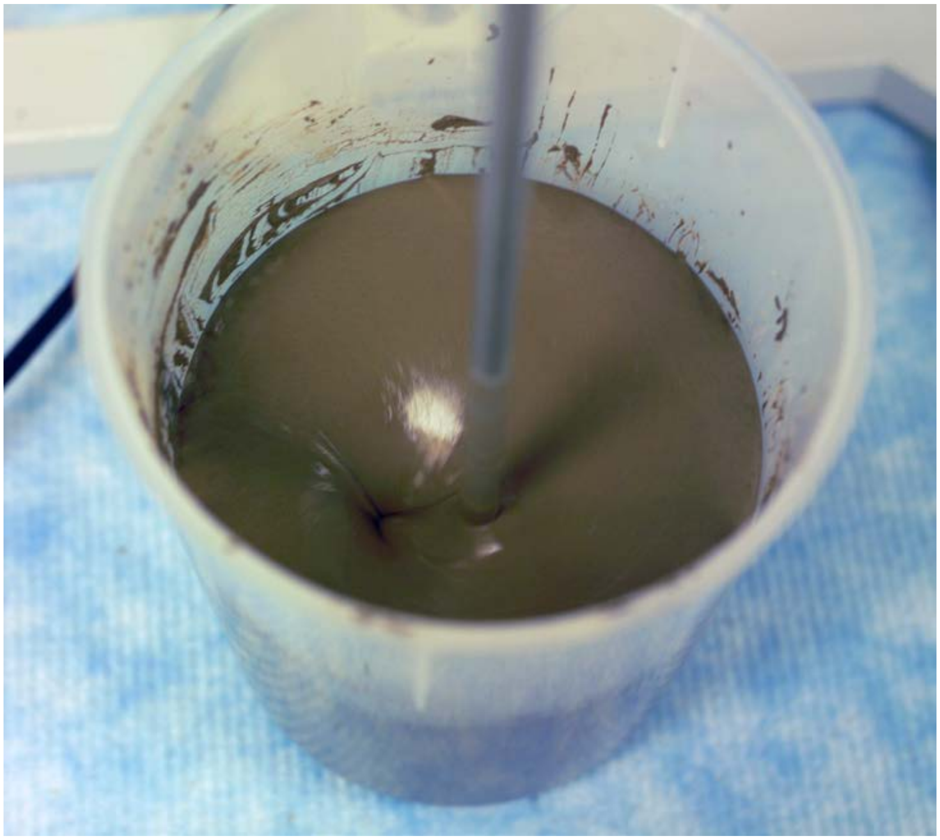

Figure 2.4. Close Up of Well Mixed Cast Stone Slurry Ready for Molding

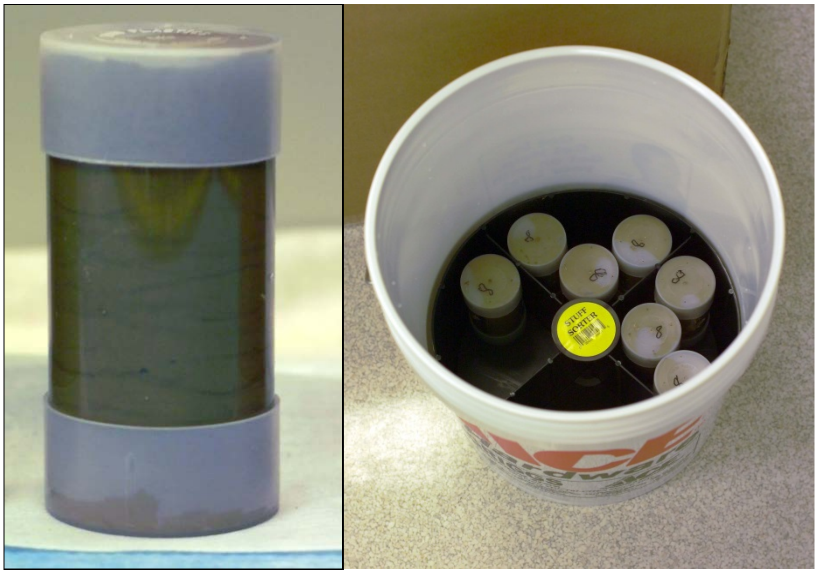

Figure 2.5. Curing Molds Filled with Cast Stone Slurry & Placement in Curing Containers

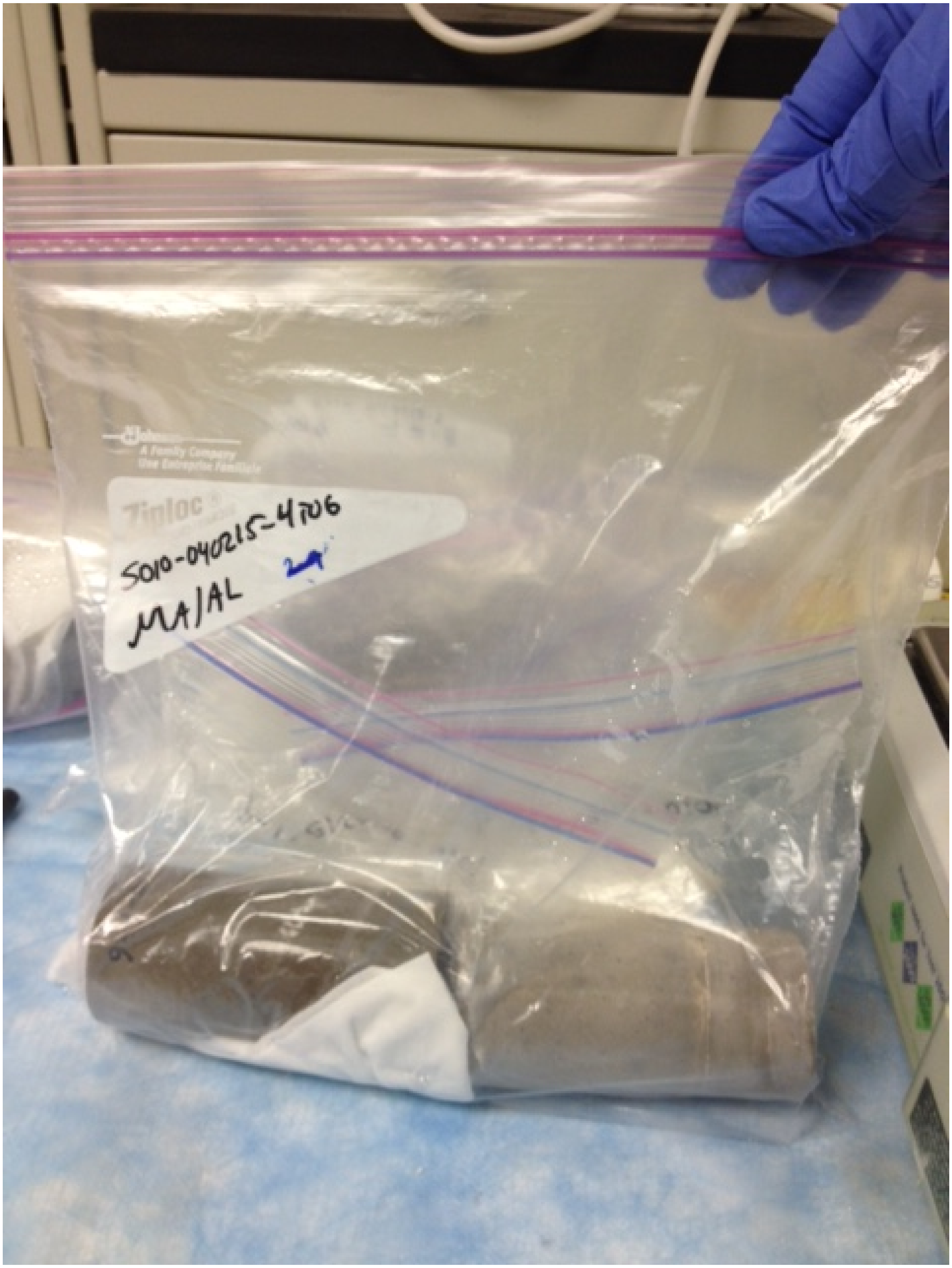

Figure 2.6. Storage Method-Monoliths in Double Lined Bags with Wet Paper Towel

2.3 Description of the Vadose Zone Pore Water Simulant

The VZP simulant recipe is shown in Table 2.5. Reagent grade chemicals were dissolved in deionized water. The recipe is loosely based on two direct measurements of actual VZP removed from Hanford formation sediments from a borehole in 200 E where the IDF is located. Several hundred grams of field moist sediment collected from borehole C4124 (299-E27-22) drilled into uncontaminated Hanford formation sediments using cable tool drive barreling were placed in special holders and ultra-centrifuged for several hours. The small volumes of VZP were forced through the sediment and collected at the bottom of the holders in small sampling cups. When approximately 30 to 50 mL of extracted water was collected from each sediment sample, the solution was immediately filtered through 0.45 µm membrane filters and analyzed for chemical composition. The results from characterizing the pore water from two 1-ft long core liners depths (47.5 to 48.5 and 81.5 to 82.5 ft below ground surface [bgs]) from borehole C4124 (299-E27-22) was averaged and then charge balanced (see Brown et al. 2008 for details on the borehole drilling and core liner). The analytical data for major cations and anions, pH and electrical conductivity for the two vadose pore waters are found in Table A.3. The sulfate values seemed peculiarly high so a portion of the sulfate was replaced by adding more chloride. Geochemical speciation calculations using the code MINEQL$^+$ also suggested that calcite would be oversaturated in the averaged ultra-centrifuged composition so alkalinity was reduced in the simulant Um et al. (2007). Although Si is present at ~23 mg/L in the actual pore waters, it was not added to the simulant recipe because the speciation of Si in actual vadose zone pore water has not been identified. Forms of Si that readily dissolve in simulants may not represent Si in actual pore water. The vadose simulant shown in Table 2.5 has been used since 2007 (Um et al. 2007) in our laboratory studies excepting recent work reported in Last et al. (2015) that used a VZP simulant recipe derived from the two direct measurements of actual VZP removed from Hanford formation sediments from a borehole C4124.

Table 2.5. Vadose Zone Pore Water Simulant

Order to Dissolve	M	Reagents Available	MW	g/L	g for 150 L	g for 600 L	g for 900 L
1	0.012	$CaSO_4 \bullet 2H_2O$	172.1723	2.0661	309.910	1239.641	1859.461
2	0.0017	NaCl	58.4430	0.0994	14.903	59.612	89.418
3	0.0004	$NaHCO_3$	84.0068	0.0336	5.040	20.162	30.242
4	0.0034	$NaNO_3$	84.9948	0.2890	43.347	173.389	260.084
5	0.0026	$MgSO_4$	120.3660	0.3130	46.943	187.771	281.656
6	0.0024	$MgCl_2 \bullet 6H_2O$	203.3034	0.4879	73.189	292.757	439.135
7	0.0007	KCl	74.5515	0.0522	7.828	31.312	46.967
Adjust pH to 7.0 to 7.2 with sodium hydroxide or sulfuric acid.							

Direct analyses of eight large batches of this VZP simulant showed that not all the Mg remained in solution (61% of expected concentration measured) and a small amount (4%) of Ca also appeared to precipitate. XRD scans of some of the un-dissolved solids showed the presence of only gypsum ($CaSO_4 \cdot 2H_2O$) despite the fact that the magnesium in solution was lower than expected. Table A.4 compares the “theoretical compositions based on masses of chemical reagents added” with the actual measured composition of the VZP.

3.0 Leach Test Details and Test Data Reduction

After curing for between 207 and 214 days, the Cast Stone monoliths[1] originally prepared for the screening tests that had been archived and are now called the Archive Leach Test Suite, and the newly (June 2013) prepared monoliths (called the I-Loading Suite and Tc-Gluconate Spiked Suite) were subjected to the EPA Method 1315, *Mass Transfer Rates of Constituents in Monolithic or Compacted Granular Materials Using as Semi-Dynamic Tank Leaching Test* (EPA 2013)[2]. The newly prepared Iodide- and Tc-Gluconate- suites monoliths were cured for 29 and 33 days, respectively before the Method 1315 leach tests started. A complete listing of the Cast Stone monolith's preparation dates and date for commencing the leach tests is provided in Table A.2. Two modifications to the EPA Method 1315 leach test were used for the Archive, Iodide, and Tc-Gluconate Suites. First, two eluents were used to leach the Cast Stone monoliths. As customary, distilled-deionized water (DIW) was one of the eluents, and a simulated Hanford formation sediment vadose zone pore water (VZP) was the second eluent. Second, the time intervals prescribed in the EPA-1315 method were extended, albeit with some large intervals, beyond 63 days up to 861 days for some monoliths. Some of the monolith leach tests continue.

Finally, 11 of the original Screening Test monolith leach tests in DIW were extended beyond 63 days (to 1037 days or 1042 days) and were named Screening Leach Tests –Extended Suite. For the Screening Leach Tests –Extended Suite, one of the two monoliths originally being leached for 11 of the mixes was chosen for extended leaching. The 11 mixes and specific replicates chosen are shown in Table 3.1 (see Figure A.1 for more details on the composition of these mixes). All of the leach tests were conducted at ambient room temperature in plastic containers on a bench top. Aside from having lids loosely fitted to the leach containers, no effort was made to exclude air from entering leach containers during sampling or during the time intervals between sampling (slow diffusion through the plastic containers). That is, no effort was made to preclude oxygen or carbon dioxide from entering the containers. As mentioned, the monoliths were all nominally 2-in. diameter × 4-in. long right cylinders. The physical dimensions of the cured monoliths were measured with calipers at three locations (top, middle, and bottom for diameter and at 120 degree angles along the sides for length). The average geometric surface area and volume of each monolith was calculated from averages of the caliper measured dimensions. The moisture content of the monoliths from the Screening Leach Tests –Extended Suite and Archive Leach Test Suite are documented in Westsik et al. (2013a). We merely extended the leach testing of these monoliths prepared by Westsik et al. (2013a). In retrospect, it might have been useful to re-measure the moisture content of the Archive Leach Test Suite of monoliths (made by Westsik et al. 2013a, but stored for an additional 176 or 181 days dependent on mix composition at room temperature under high relative humidity conditions). The newly made monoliths for the I-Loading Suite and Tc-Gluconate Spiked Suite did not have moisture contents directly measured. We used the average moisture content value for Mix #3 and #22 (replicates) from Westsik et al. (2013a). We justified this indirect moisture content determination because we made the

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[1] The shapes of the monoliths were right cylinders with dimensions of 2-in diameter by 4-in length. EPA Method 1315 states "The geometry of monolithic samples may be rectangular (e.g., bricks or tiles), cubes, wafers or cylinders. In all cases, a minimum sample size of 5 cm in the direction of mass transfer must be employed and the liquid- surface-area ratio (L/A) must be maintained at 9 ± 1 mL/cm^2." Further, compacted (following a modified Proctor compaction protocol) granular material can be used. However, the sample geometry of the compacted granular material must be open-faced cylinders due to limitations of mechanical packing. In all cases, the compacted granular sample size must be at least 5 cm in the direction of mass transfer and the L/A must be maintained at 9 ± 1 mL/cm^2'. Thus, the measured D_{obs} values reported herein should be applicable for predicting the diffusional release of COCs from the larger rectangular waste forms that are planned for burial in the IDF.

[2] These screening tests used the draft EPA 1315 method (published in 2012). EPA Method 1315 became a formally accepted EPA method in January 2015 with no significant changes in methodology between the draft and final version. At the time of this report, the final validated version is available at https://www.epa.gov/hw-sw846/validated-test-method-1315-mass-transfer-rates-constituents-monolithic-or-compacted.

new 7.8 M Na Average simulant using the identical recipe, used the same dry blend materials, the same mixing equipment and procedure for the wet slurry, and the same monolith curing conditions as used in the screening tests. The chosen gravimetric moisture content value (0.254) for the newly prepared monoliths was calculated by difference from the measured solids content, 0.746.

Table 3.1. Monoliths from Screening Test that had Leach Testing Extended

Number	Mix #	Monolith Final #s Extended in DIW
1	5	CS-T5HCS1-7.8AVG-2
2	8	CS-T8LCS1-5RAS-4
3	10	CS-T10HCS1-5HIS-6
4	13	CS-T13LCS2-5AVG-2
5	14	CS-T14LCS2-7.8HIS-3
6	15	CS-T15HCS1-7.8HIS-1
7	16	CS-T16HCS1-7.8RAS-3
8	17	CS-T17LCS2-5HIA-3
9	18	CS-T18LCS2-7.8RAS-6
10	21	CS-T21LCS1-7.8HIS-6
11	24	CS-T24HCS1-5HIA-1

Monolith naming convention:

CS	Cast Stone
T#	mix number (see Figure A.1)
HC	NW fly ash has high Ca content
LC	SE fly ash which has low Ca content
S1	NW BFS
S2	SE BFS
-7.8 or -5	Na molarity in waste simulant
AAA	simulant abbreviation; AVE= Average, RAS = saltcake blend, HIS=high sulfate, HIA= high aluminum
Final #	monolith number (six monoliths of each mix were prepared but only two were used in EPA Method 1315 leach testing for the original screening tests).

3.1 EPA Method 1315

The EPA Method 1315 (EPA 2013) is a semi-dynamic leach test that consists of iteratively submerging a monolithic sample in eluent at a fixed liquid-volume to solid-geometric-surface-area ratio. The initial sampling was done at fixed periods of time at cumulative leaching times of 0.08, 1, 2, 7, 14, 28, 42, 49, and 63 days. For the screening tests documented in Westsik et al. (2013a), two additional samplings were conducted at cumulative leaching times of 77 and 91 days. For this report, eluates were collected at longer times, listed in Table 3.2. At the end of each sampling interval, all the leaching fluid was removed and replaced with fresh fluid. A schematic of this process is shown in Figure 3.1.

Table 3.2. Sampling Interval Durations and Cumulative Time (days) for the Four Suites of Monoliths

Extended Suite (Mix 5, 8,10, 13, 14)		**Extended Suite (Mix 15, 16, 17, 18, 21, 24)**		**Archive Suite**		**Iodide Suite**		**Tc-Gluconate Suite**	
Interval (days)	Total (days)	Interval (days)	Total (days)	Interval (days)	Total (days)	Interval (days)	Total (days)	Interval (days)	Total (days)
0.08	0.08	0.08	0.08	0.08	0.08	0.08	0.08	0.08	0.08
0.92	1	0.92	1	0.92	1	0.92	1	0.92	1
1	2	1	2	1	2	1	2	1	2
5	7	5	7	5	7	5	7	5	7
7	14	7	14	7	14	7	14	7	14
14	28	14	28	14	28	14	28	14	28
14	42	14	42	14	42	14	42	14	42
7	49	7	49	7	49	7	49	7	49
14	63	14	63	14	63	14	63	14	63
14	77	14	77	30	93	37	100	31	94
14	91	14	91	287	380	269	369	267	361
28	119	28	119	47	427	45	414	47	408
33	152	28	147	163	590	155	569	161	569
35	187	35	182	271	861			272	841
35	222	35	217						
35	257	35	252						
305	562	305	557						
47	609	47	604						
161	770	161	765						
272	1042	272	1037						

Note: most of these leach tests were continued through February 2015 when about half of the Extended (2/18/2015), the Archive (2/19/2015), and Tc-Gluconate (2/18/2015) Suite monolith leach testing was terminated. All the Iodide Suite tests were terminated on February 13, 2015. The last sampling of the Extended, the Archive, and Tc-Gluconate Suite monoliths occurred in Nov 2015. Monoliths whose leach tests were terminated in February 2015 were stored in their final eluates and are being used for post-leaching solid phase characterization. A report describing the solid phase characterization will be prepared in September 2016.

At the end of each of the leaching intervals, the monolith mass was recorded, and the leaching solution was changed. This method is similar to ANSI/ANS-16.1 (2003) and ASTM-1308 (current version is C-1308-08; published in 2008), but the leaching intervals are modified, and the developers of EPA Method 1315 claim that the process of mass transfer can be interpreted by more complex release models that account for physical retention of the porous medium and chemical retention within the porous matrix.

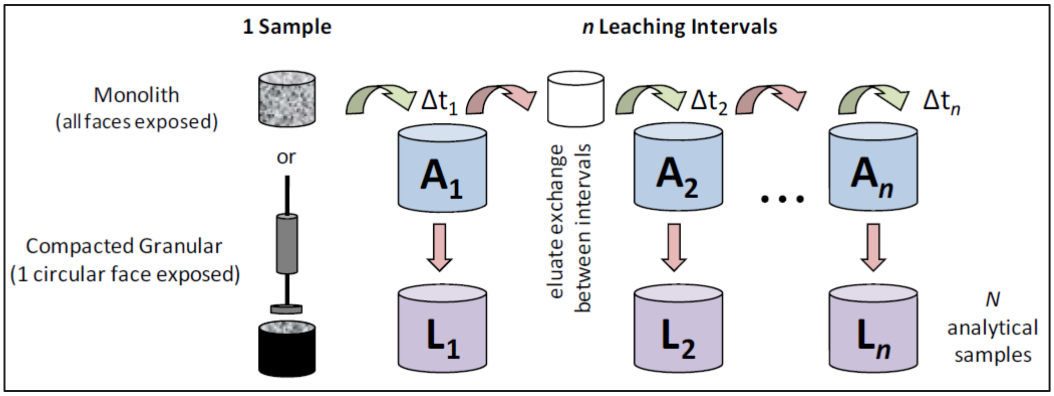

Figure 3.1. Schematic of the EPA Method 1315 Leach Protocol

The cylindrical monoliths were placed into the centers of leaching vessels containing sufficient eluent to maintain a solution-to-solid surface area ratio of 9 ± 1 mL of eluent per square centimeter of sample geometric surface area. Sample stands and holders (see Figure 3.2 and Figure 3.3) were used to maximize the contact area of the monolith sample with the leaching solution. Figure 3.4 shows one of the monoliths submerged in the leaching container. In between the sampling/replacement intervals, the leach vessels were covered with lids. Solution pH and electrical conductivities were measured on a small aliquot of unfiltered eluate at each leaching interval. Then a larger aliquot of eluate was removed from the leach container and split into various analytical aliquots, some preserved as noted in Table 3.3, and submitted for chemical analyses using the analytical method designated.

Figure 3.2. Photo of Monoliths on Stands and Leach Containers Prior to Start of Leach Tests

Figure 3.3. Close Up of Monoliths on Stands Prior to Leaching

Figure 3.4. Monolith in Leach Container During Leach Testing

Table 3.3. Chemical Analyses of Eluates

Method	Analyte	Volume (mLs)	Container/Preservation
ICP-OES	Cations (Na, K, Ca, Mg, S others)	20	Plastic vial; ultrapure nitric to pH ~2
ICP-MS	Cr, ^{99}Tc	--	Same Plastic vial as ICP-OES; ultrapure nitric to pH ~2
IC	Nitrate, nitrite, sulfate, (others)	20	Plastic vial; none
pH, EC probes	pH, EC	25	Unfiltered; Plastic vial; none
Titration	Alkalinity (phenolphthalein & total)	--	Same Plastic vial as IC; none
ICP-MS	^{127}I	20	Plastic vial; Spectrasol to pH ~11

3.2 Leach Test Data Reduction Terminology

There is a lack of consistency in the use of mass diffusion terminology in the literature. For example, Table 3.4 is a summary of definitions for specific diffusion coefficients from three leading references

.

Table 3.4. Definitions of Diffusivity in the Literature

Reference	D^{mol}	D^{eff}	D^{int}	D^{app}
van der Sloot et al. (2009)	**Molecular diffusivity** is a property of the diffusing species and temperature with no consideration for the physical or chemical effects. Three variants to the molecular diffusion coefficient may be seen in the porous media literature depending on incorporation of various physical and chemical influences.	The **effective diffusivity** describes the rate of diffusion of a species in a tortuous, porous medium relative to the pore area through which diffusion occurs. Thus, effective diffusivity accounts for tortuosity, but not for porosity or chemical effects.	**Intrinsic diffusivity** represents the rate of diffusional transport that is hindered by the effective surface area (e.g., porosity) and the tortuous pathway of the fluid phase (e.g., tortuosity). Intrinsic diffusivity does not account for chemical effects on mass transport.	The **apparent diffusivity** describes all physical and chemical effects that hinder the diffusion of constituents in a porous material. Classically, the associated form of the diffusion coefficient is derived from the closed form solution of the semi-infinite diffusion into an infinite bath (Crank, 1986) under the assumption that all chemical interactions may be described by a linear portioning coefficient. The apparent diffusivity is the rate of diffusion observed from experimental mass transfer tests.

Reference	D_{mol}	D_{eff}		D_{obs}
Garrabrants and Kosson, 2005	**molecular diffusivity** Denotes the rate of change in the concentration gradient for a species diffusing in aqueous media.	**effective diffusivity** Is used to describe the mass transport when physical retardation is present. Diffusion in a non-reactive porous medium retards the release of constituents in relationship to molecular diffusion through effective surface area and increased path length, or tortuosity τ.		**observed/apparent diffusivity** One measure of diffusivity in chemically reactive systems. In chemically reactive systems, the rate of diffusion for a constituent is slowed due to interactions with the surface, or chemical retardation R.

Reference	D	D_e	D_i	D_a
Seitz and Walton. 1993. NUREG/CR-6070	**tracer diffusion coefficient** in water (cm^2/s). Simply the diffusion coefficient in water. It assumes saturation and does not include tortuosity or sorption due to a porous media.	**effective diffusion coefficient** in porous media (cm^2/s). Includes the effects of tortuosity, but not the effects of sorption and moisture content.	**intrinsic diffusion coefficient** measured in steady-state leach test across thin slice of porous media (cm^2/s). Includes the effects of tortuosity and moisture content, but not sorption.	**apparent diffusion coefficient** obtained from batch leach tests (cm^2/s). Includes the effects of all the factors in columns 2, 3, and 4.
	For saturated systems, as in a concrete block, the effective and apparent diffusion coefficients are essentially equal for non-sorbing radionuclides or tracers because the tracer is all in the liquid phase.			

There is also inconsistency in the standard leach test methods for what is meant by the terms for derived diffusivities (observed or apparent, or in ANS16.1 effective) and Leachability Index. Table 3.5 is a summary of diffusivities and Leachability Index as used in four standard leach test methods.

Table 3.5. Observed/Effective Diffusivity and Leachability Definitions in Standard Leach Test Methods

<table>
<tr><th>Leach Test Method</th><th>Observed diffusivity/diffusion coefficient</th><th>Effective diffusivity/diffusion coefficient</th></tr>
<tr><td>EPA Method 1315</td><td>Observed mass diffusivity – The apparent, macroscopic rate of release due to mass transfer from a solid into a liquid as measured using a leaching test under conditions where mass transfer controls release. The observed diffusivity accounts for all physical and chemical retention factors influencing mass transfer and is typically expressed in units of cm2/s.</td><td>Effective mass diffusivity – The intrinsic rate of mass transfer in a porous medium accounting for physical retention. The effective mass diffusivity is typically expressed in units of cm2/s.</td></tr>
<tr><td rowspan="2">ANSI/ANS-16.1-2003</td><td>Neither observed diffusivity or observed diffusion coefficient is used.</td><td>Effective diffusivity is defined solely by Eq. 3.1 in the procedure and given the symbol D.
Effective diffusivity is first used in quotes on page 9:
Under these conditions the mass-transport equations permit the calculation [4] of an “effective diffusivity” by the following expression:
Note: Reference [4] in the procedure is ORNL-TM-4333 (1974). Effective diffusivity is defined there within equation 16 (similar to Eq. 3.1 in our report after re-arranging to solve for D). The ORNL report also adds the subscript, D_e to introduce the effective concept, which we and others prefer to call observed.</td></tr>
<tr><td colspan="2">Leachability Index for a constituent of concern i (L_i) is defined as:
$L_i = \frac{1}{7}\sum_1^7[log(\beta/D_i)]_n$ where: β is a defined constant of 1.0 cm^2/s and
D_i is the effective diffusivity of constituent i in interval n.
If all the initial and boundary conditions for the diffusion model used in this standard are met, the Leachability Index would be independent of time. However, the measurements of *L* can include the effects of unknown, perhaps intermittent, variations that might be insignificant for all practical purposes. This standard uses the confidence range and the correlation coefficient as measures of these discrepancies.</td></tr>
<tr><td>MT001.1 in Kosson et al. (2002)</td><td>What is meant by observed diffusivity is not defined.</td><td>Effective diffusivity is not used.</td></tr>
</table>

Leach Test Method	Observed diffusivity/diffusion coefficient	Effective diffusivity/diffusion coefficient
ASTM C1308-08	**diffusion coefficient (diffusivity)**—an intrinsic property of a species that relates (1) its concentration gradient to its flux in a given medium (Fick's first law), (2) its spatial rate of change in the direction of the concentration gradient to the time rate of change in its concentration in a given medium (Fick's second law), or (3) its mean square displacement to time in a given medium (the Einstein equation).	**effective diffusion coefficient (De)**—the diffusion coefficient as modified by other processes (for example, adsorption) or physical constraints (for example, tortuosity and constrictivity).

Past reports that tabulate diffusion coefficients for Hanford Site wastes that have been solidified in Cast Stone or other grout blends and then leached in laboratory tests use the term and symbol identified here in to quantify the calculated diffusion value.

Flach et al. (2016) -→ apparent diffusion coefficient (D_{app})
Cantrell et al. (2016) -→ effective diffusion coefficient (D_{eff})
Um et al. (2016) -→ effective diffusion coefficient (D_{eff})
Serne et al. (2015) -→ effective diffusion coefficient (D_e)
Fox et al. (2014) --→ reports only average leach indices (LI's)
Westsik et al. (2013)- -→ effective diffusion coefficient (no symbol used)
Mattigod et al. (2011)- -→ effective diffusion coefficient (D) with no subscript in Tables and Figures
Sundaram et al. (2011)- -→ effective diffusion coefficient (D) with no subscript in Tables and Figures
Pierce et al. (2010)- -→ observed diffusion coefficient (D) with no subscript in Tables and Figures

In summary, the interval-based diffusion coefficient obtained in the standard semi-dynamic eluent replacement tests that includes both physical and chemical effects on contaminant release is most often called "observed" or "apparent" in EPA Method 1315/Garrabrandts and Kosson/LEAF/Vanderbilt/van der Sloot and Seitz and Walton references. Their literature reserves the term "effective" to denote physical processes that retard the diffusion of contaminants out of porous waste forms.

Other literature uses the term intrinsic diffusion coefficient to represent the diffusion "retarding" effects generated by physical attributes of the porous waste form. We prefer using the term intrinsic diffusion coefficient to represent the diffusivity value controlled by physical processes such as tortuosity and constrictivity, which are discussed in the next section.

On the other hand, references/leach methods associated with ANS-16.1 and ASTM C1308 use the term "effective" to include both physical and chemical processes that impede contaminant release from porous waste forms in comparison to diffusion of the same constituent in water. Because most of the PNNL researchers performing laboratory leach tests had years of experience using the ANS-16.1 protocol, they continued to use the term effective diffusion coefficient when converting to using EPA Method 1315. They should have started using the term observed diffusion coefficient. Hopefully this oversight has been corrected in this Rev. 1 report. Neither ANS-16.1 or ASTM C1308 methods or their supporting documentation address or name a diffusion coefficient that represents solely physical processes (e.g., tortuosity, constrictivity) that impede a constituent's diffusion through a porous waste form in comparison to diffusion in water.

To avoid continuing the confusion in naming the diffusion coefficients, we use **observed diffusion coefficient (D_{obs})** to represent the diffusion coefficient that includes all physical **and** chemical processes that lower the diffusivity of a COC released from porous cementitious waste forms. We also use the term **intrinsic diffusion coefficient (D_i)** to represent the diffusivity of a COC that is **only impacted by physical processes** that are intrinsic to the physical attributes of the porous solid. For example, nitrate is often assumed to have no (or at least insignificant) chemical interactions (sorption or solubility, etc.) with cementitious waste forms. We also contend that literature using the term diffusivity is referring to diffusion coefficient (i.e., the two terms are synonymous.).

3.3 Observed Diffusion Coefficient (Diffusivity) Calculations

The leach testing discussed in this report followed protocols in EPA Method 1315. Data reduction follows from EPA Method 1315, although ANSI/ANS-16.1 data reduction techniques are also used to show “equivalency”. Garrabrants and Kosson (2005) observe that under assumed simple diffusion controlled release, Fick’s second law may be fit to cumulative release data to calculate an observed diffusivity, D^{obs} [m^2/s], using <u>either</u> of the following two simple equations for semi-infinite media:

$$D = \pi \left[\frac{a_n/A_0}{(\Delta t)_n}\right]^2 \left[\frac{V}{S}\right]^2 \left[\frac{1}{2}\left(t_n^{1/2} + t_{n-1}^{1/2}\right)\right]^2 \qquad \text{Eq. 3.1}$$

(ANS-16.1)

Where: D = effective diffusivity for a leaching interval (cm^2/s)
a_n = quantity of constituent released from the monolith during interval n (mass units)
A_0 = total quantity of given contaminant in the specimen at the beginning of the first leaching interval (same mass units as a_n)
$(\Delta t)_n$ = duration of leaching interval n (s) = t_n-t_{n-1}
V = volume of the solid monolith (cm^3)
S = geometric surface area of the monolith calculated from measured dimensions (cm^2)
t_n = cumulative leaching time since the beginning of the first leaching interval through interval n (s)

$$D_i^{obs} = \pi \left[\frac{M_{t_i}}{2\,\rho\,C_0\left(\sqrt{t_i}-\sqrt{t_{i-1}}\right)}\right]^2 \qquad \text{Eq. 3.2}$$

(EPA Method 1315)

Where: D_{obs} = observed diffusivity of a specific constituent for leaching interval, *i* (m^2/s),
M_{ti} = mass of specific constituent released during leaching interval *i* (mg/m^2),
t_i = cumulative contact time at the end of the current leaching interval, *i* (s),
t_{i-1} = cumulative contact time at the end of the previous leaching interval, *i-1* (s),
ρ = Cast Stone bulk density (dry basis) (kg-dry/m^3), and
C_o = initial concentration of constituent in the dry Cast Stone (mg/kg-dry).

Eq. 3.1 was developed for data reduction within the ANS-16.1 protocol for radionuclide release (ANSI/ANS 2003), while Eq. 3.2 was developed for heavy metal and matrix constituent release using NEN 7345 (Dutch Standardization Institute 1995) and MT001 in Kosson et al (2002). Both Eq. 3.1 and Eq. 3.2 are based on the same diffusion model assuming (i) a homogeneous solid matrix, (ii) constant release parameters, (iii) constant driving force for diffusion (i.e., zero concentration at the solid’s surface boundary), (iv) no depletion of the constituent at the inner core of the solid matrix, and (v) less than 20% of the initial content is leached (which allows the use of the semi-infinite media assumption given in the equations).

Because our leach testing followed the EPA Method 1315 protocols, we stay consistent with that method’s nomenclature and its observed diffusivity equation (Eq. 3.2 here, and page 1315-17 in the EPA method).

Note that the most common units for observed or effective diffusion coefficients when used in transport modeling is cm^2/s such that the value calculated in Eq. 3.1, Eq. 3.2, or Eq. 3.3 is multiplied by 10,000 cm^2/m^2 to convert the value to cm^2/s. Also in this report, the C_0 (initial concentrations of COCs) were calculated solely from the concentrations present in the liquid waste simulant. It was assumed that any COC, if present, in the dry blend was in a more inert form that was not prone to leaching.

For this report, the constituents of interest (often called contaminant of concern) include Cr, I, Na, Tc, U, nitrate, and nitrite.

The leachability index (LI) for a COC, is defined in ANS-16.1 as a unitless parameter derived from averaging the interval leachability values (in ANS-16.1 there are 7 leaching intervals specified; thus n = 7 in that method), and is calculated using Eq. 3.3:

$$LI = \frac{1}{n}\sum_1^n \left[log\left(\beta/D^{obs}\right)\right]_n \qquad \text{Eq. 3.3}$$

where: $\beta = 1.0\ cm^2/s$ and D = observed (effective in ANS-16.1) diffusivity for interval n.

To more simply convert any interval observed diffusion coefficient to an interval leachability index, Eq. 3.4 is used

$$LI_n = -log[D_n^{obs}] \qquad \text{Eq. 3.4}$$

where LI_n = the LI for each leach interval (n) and D_n^{obs} is the observed diffusivity for the constituent of interest (cm^2/s) during the leach interval n.

EPA Method 1315 (EPA 2013) is similar to ANSI/ANS-16.1 (see ANSI/ANS 2003) and ASTM Method 1308-08 (ASTM 2008a), but the leaching intervals are modified, and the process of mass transfer supposedly can be interpreted by more complex release models that account for physical retention of the porous medium and chemical retention within the porous matrix. Mattigod et al. (2011) have shown that these three monolith leach test methods yield the same diffusion coefficient over time periods of 91 days for Cast Stone monoliths containing several secondary waste simulants.

In the ANS-16.1 method, if more than 20% of the mass of a given constituent of interest leaches from the cylindrical monoliths, effective diffusivity corrections (to account for depletion of the mass of the leaching constituent) are recommended; correction factors are presented in the method's Appendix A. ASTM-1308 also suggested that corrections should be made when the waste form leaches (depletes) more than 20% of the starting inventory of the constituent of interest. In our EPA-1315 leach tests reported herein many of the monoliths leached more than 20% of the starting inventories for the mobile COCs (Na, NO_2, NO_3, and iodide), but the EPA Method 1315 criteria for a diffusion-controlled mechanism was also satisfied. The fraction leached for all COCs in our tests as a function of cumulative leach time are shown in Appendix B.

ASTM-C1308 also promotes performing the leaching tests at three (two elevated above room temperature) temperatures so that the temperature effects on the empirical effective diffusion coefficient can be quantified. Diffusion increases with temperature such that leach testing at elevated temperatures can be used to accelerate the diffusion process, which can be used to extrapolate releases to longer time periods for disposal conditions in subsurface repositories. This method of accelerated testing may be valid as long as the release mechanism for each constituent of interest does not change as a function of temperature.

There are several types of constituent release, and ways of portraying them, that are mentioned in the literature in relation to semi-dynamic tank leaching tests. In Appendix C, we show plots of release log (mg/m^2) vs. log of leach time in days that describe data from semi-dynamic tank leaching tests for a) simple diffusion controlled release, b) delayed release, c) wash-out of mobile species, d) depletion of leachable species, e) surface wash-off, and f) release when there is a significant change in chemical conditions.

The EPA Method 1315 mentions the need to check raw interval diffusivity data to evaluate whether the data support (or "allow") averaged D_{obs} values to be calculated from the individual leach test interval using Eq. 3.2 (above). Fickian diffusion requires that the interval slope for the line, which is plotted as cumulative log of release versus cumulative log of time, be equal to 0.5. Per EPA Method 1315, only data from those intervals having slopes of 0.5 ±0.15 are to be used to calculate an observed diffusivity for the interval using Eq. 3.2, and those values of D_{obs} that meet this criterion can then to be used to determine the overall mean (or average) observed diffusivity. If the interval slope is not 0.5 ±0.15 for any given interval, then the interval D_{obs} value calculated from Eq. C.4 cannot be used to determine a mean observed diffusivity.

This interval slope checking criterion has seldom, if ever, been explicitly discussed in the Cast Stone, saltstone, or hydrated lime-based grout technical reports[1] containing experimental data. Thus, selecting only interval D_{obs} values from the Cast Stone monoliths EPA Method 1315 tests that meet the criteria so they can be considered for calculating mean D_{obs} values and leachability indices is problematic.

Appendix C in this report is our first attempt at performing such "criteria" checks. As shown in Appendix C, Table C.1, Table C.2, Table C.3, and Table C.4, many of the interval slopes for the key COPCs, especially for the mobile nitrate and iodide, do not have many interval slopes within the 0.5 ± 0.15 criterion. Whether this interval slope criterion and the <20% source inventory criterion must be met before using interval D_{obs} values to calculate averages/means for use in PA modeling is an open question, in our opinion. We suggest that this "criteria" checking issue be discussed between leaching experimentalists, PA modelers, and regulatory communities in the hope of developing a consensus path forward. Thus, we did not censor interval data that fails the "criteria" checks or modify our recommendations for what D_{obs} values to use in the next round of IDF calculations. We leave the decision up to the reader on whether to impose the EPA Method 1315 "criteria" checking procedures on available experimental leach data to remove interval D_{obs} values that do not meet the criteria and thus modify recommendations made in the cited reports.

Table C.1 and Table C.2 show the interval slope checks as well as when the leached inventory exceeds 20% for our Extended and Archived Suites of monoliths (see Table 3.1 and Table 4.3 in the main text for specifics) on the monolith compositions. Table C.3 and Table C.4 show the same interval slope checks as well as when the leached inventory exceeds 20% for the Tc-Gluconate and Iodide Suites. Details on their compositions are found in Table 2.4, Table 4.6, and Table 4.8.

The next section provides more details on conceptualizing the release of a constituent solidified in a cementitious porous waste form and how the various physical and chemical processes that influence the release of the constituent can be parameterized and in certain cases measured from companion testing.

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[1] See foot note 1 on page C.4, for a listing of reports that do not discuss performing "criteria" checks on interval leach data before tabulating the data or recommending specific values to get average D_{obs} values to recommend to IDF PA modelers.

3.4 Derivation of Diffusion Coefficients for Porous Media (Conceptual Release Model)

Diffusional release of species from cementitious waste forms, such as Cast Stone, is best treated as a combination of transport impacted by physical and chemical processes. Herein, we use a set of equations presented in the following references—Atkinson et al. (1986), Atkinson and Nickerson (1988) and van Brakel and Heertjes (1974)—that show the relationship between the various diffusion coefficients from diffusion of an ion in dilute water, to the diffusion of the same ion through a porous media such as Cast Stone, and finally diffusion of the same ion assuming that it chemically interacts with the porous medium through reversible sorption reactions. The conceptual model is based on the fact that cementitious waste forms consist of a complex porous matrix, which restricts free diffusion via physical processes as well as being a chemically reactive solid, with additional diffusion constraints caused by chemical interactions. The chemical interactions can have important retarding effects on final transport (or release) rates. The intrinsic diffusion coefficient (D_i) of each particular species through the porous media quantifies the physical constraints to diffusion, and conceptually depends on the tortuosity (τ), constrictivity (δ), and porosity (ε) of the cementitious waste form. These three physical parameters, attributes of the porous media, influence the diffusion coefficient of a solute in dilute water, D_f, described by:

$$D_i = D_f \frac{\varepsilon\delta}{\tau^2} \qquad \text{Eq. 3.5}$$

Tortuosity and constrictivity are two dimensionless parameters that are not readily measured, but conceptually represent physical attributes of the porous media. Tortuosity relates to the fact that a diffusing species inside the porous medium will have to travel a larger distance to reach the outer surface of the porous medium because the complex solid structure impedes direct migration down the concentration gradient that the ion would follow if diffusing through a fluid. Figure 3.5 is a portrayal of the tortuosity concept; it is obvious in this conceptualization that tortuosity has a numerical value greater than 1. Constrictivity, another dimensionless parameter, is viewed to depend on the ratio of the diameters of the smallest and largest pores in the porous media. The value of constrictivity is always less than 1. Constrictivity is not defined for a single pore, but as the parameter of the entire pore space within the porous media being considered. One numerical conceptualization of constrictivity assumes that constrictivity is related to a relationship between the maximum, minimum and mean pore diameters (see Figure 3.6. In practice, the constrictivity, tortuosity, and porosity are often used in models as purely empirical parameters to establish the observed diffusivities in porous media.

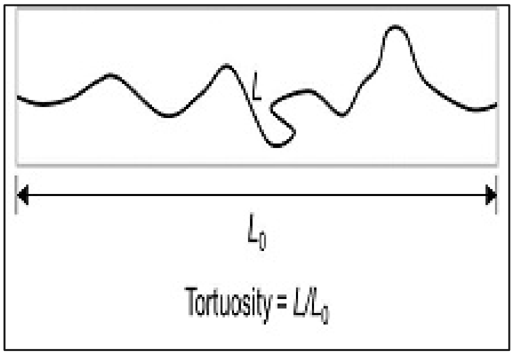

Figure 3.5. Conceptual Representation of Tortuosity (τ)

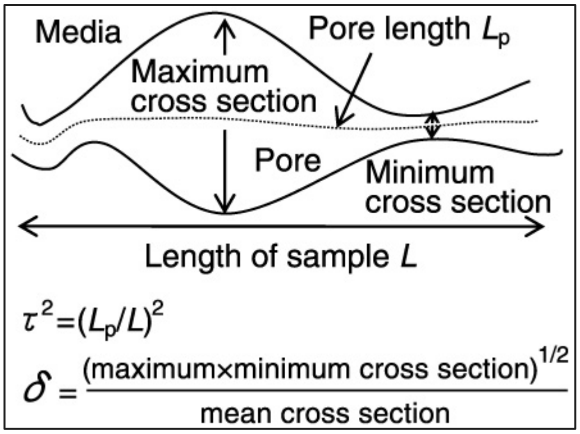

Figure 3.6. Conceptual Representation of Constrictivity (δ)[1]

Chemical interactions can be quite varied (ion exchange, precipitation, specific and irreversible adsorption, and each process may have fast or slow kinetics). The simplest process that is mathematically readily tractable is fully reversible ion exchange with fast kinetics that obeys a linear isotherm. This simple chemical process gives rise to the following equation:

$$D_{obs} = \frac{D_i}{\alpha} \qquad \text{Eq. 3.6}$$

where the modified diffusion coefficient is called the observed diffusion coefficient, D_{obs}[2], and is related to the intrinsic diffusion coefficient for the physically constrained porous media by a chemical capacity factor, α. The chemical capacity factor is the ratio of the moles of contaminant per unit volume of water-saturated solid (C_s) to the moles per unit volume of contaminant in the liquid, C_L. The chemical capacity factor for the simple conceptual model for reversible sorption is related to the K_d (mL/g) by the following equation:

$$\alpha = \varepsilon + \rho K_d \qquad \text{Eq. 3.7}$$

where ρ is the is the dry bulk density of the porous solid waste form. Again, we stress that this relationship requires fast and reversible chemical reaction processes and that sorption satisfies the linear isotherm constraint. Few chemical reactions for contaminants meet these requirements. Regardless, this simple construct is often applied in quantifying the release of contaminants from cementitious waste forms because it allows one to separate the physical and chemical processes that control transport of contaminants. There are several experimental methods that one can use to measure the K_d (and then compute α) after measuring the porosity and bulk density of the waste form. Conversely, one can measure the observed diffusion coefficient using through-diffusion cells, penetration profiles of a contaminant into a solid porous medium, or out diffusion of contaminants (leaching tests), and then attempt to calculate K_d values by comparing the D_{obs} values for a companion species that is assumed to

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[1] Figure taken from Takahashi, H. et al., 3D X-ray CT and Diffusion Measurements to Assess Tortuosity and Constrictivity in a Sedimentary Rock, diffusion-fundamentals.org, vol.11, issue 89, 2009, p.1-11, (http://www.uni-leipzig.de/diffusion/journal/contents_vol11.html).

[2] The term observed diffusion coefficient used in this report is called effective diffusion coefficient in ANSI/ANS16.1, which uses the symbol D with no subscript. ASTM C1308 method uses the symbol D_e and name effective diffusion coefficient. In some literature this parameter, D_{obs} defined in Eq. 3.1 through Eq. 3.3 and Eq. 3.7 (used to accommodate all physical and all chemical effects) is called apparent diffusion coefficient, D_a (see for example Grathwohl (1998). Despite being named differently in different literature when “quantified” in standard leach tests by use of Eq. 3.1 and Eq. 3.2 or Eq. 3.6, the same numerical value is obtained.

have a zero K_d and is present in the same waste form. If one also has an independent or direct measure of the porosity of the cement waste form, then the measured D_{obs} value for the non-sorbing constituent allows one to calculate the intrinsic (D_i) coefficient for the specific waste form using Eq. 3.5 and the known porosity because Eq. 3.7 reduces to the porosity (ε) when K_d is zero. Note that α is equivalent to the commonly used retardation factor, R, multiplied by porosity of the porous media as shown in Eq. 3.8 and Eq. 3.9.

$$\alpha = \varepsilon R \qquad \text{Eq. 3.8}$$

where R for water saturated porous media is

$$R = \left[1 + \left(\frac{\rho_b}{\varepsilon}\right)K_d\right] \qquad \text{Eq. 3.9}$$

Where ρ_b is the dry bulk density of the porous media with units of M/L^3 generally g/cm^3 and
ε is porosity as defined previously —unit-less and
K_d is the sorption distribution coefficient with units of L^3/M generally cm^3/g.

For Cast Stone, which contains the reductant BFS in its dry blend, we caution against assuming the measured D_{obs} values for redox sensitive COCs, such as Tc and Cr, represent chemical interactions controlled by reversible sorption. Thus for Cast Stone containing mobile contaminants that are generally assumed to have K_d values equal to zero one can estimate a desorption K_d using the logic just described wherein the D_{obs} value for the mobile COC (often nitrate) is used in Eq. 3.6 and Eq. 3.7 along with independent measurements of porosity and dry bulk density to calculate the Cast Stone's inherent porous media D_i value. This calculated D_i value should be the same for all COCs in any given Cast Stone monolith for a specific mix/composition. Then by taking measured D_{obs} for the chemically reactive COC such as Tc or Cr along with the calculated D_i value for Cast Stone monolith for the mix of interest one can calculate the α ratio for the reactive COC using Eq. 3.6. From the independently measured values for porosity and dry bulk density one can calculate the K_d for the constituent using Eq. 3.7. But as mentioned the release of reduced species of Tc, Cr, U, and other redox sensitive COCs from Cast Stone is not likely to be controlled by reversible sorption processes. Thus, despite the fact that the above logic can be used to calculate a K_d for these COCs, their release is not controlled by reversible desorption processes and therefore, predictions of future release based on the calculated desorption K_d may be unfounded.

4.0 Results of the Long-Term Leach Tests—Through FY 2016

The results of the extended leach testing on the Cast Stone monoliths presented in Rev. 0 of this report for the four suites of tests that were performed to gather data on the objectives delineated in Section 1.0 are now updated to include two more eluent replacements that occurred in February 2015 and November 2015. The order of presentation is Screening Leach Tests –Extended Suite, Archive Leach Test Suite, I-Loading Suite, and finally, the Tc-Gluconate Spiked Suite.

4.1 Screening Leach Tests –Extended Suite

The objective of this testing was to extend the leaching times for selected ongoing EPA-1315 tests on Cast Stone monoliths made with LAW simulants beyond the conventional 63-day time period in an effort to evaluate long-term leaching properties of Cast Stone to support future PA activities. Table 3.1 shows the eleven monoliths from the Screening Test activities (see Westsik et al. 2013a) that were chosen for extended leaching in de-ionized water. The EPA-1315 leach tests for the Screening Test activities were performed for 91 days on duplicates of 26 mixes of LAW simulants-Cast Stone dry blend as described in Westsik et al. (2013a) and herein shown in Figure A.1. Based on the detailed characterization of both the wet slurry and cured Cast Stone monoliths generated in the Screening Test, Mixes #1 and #23 were deemed unacceptable; thus these two mixes were not considered for the extended leach testing. The leaching results for the first 91 days for the remaining 24 mixes were evaluated and the 11 monoliths were chosen for further leaching. Selection was based on wanting to cover the entire range of leach performance. Therefore, one of the better-performing (i.e., having the lowest leach rate or D_{obs} value) monoliths, one of the worst performing (i.e., having nearly the highest leach rate or D_{obs} value) monoliths, and one of the median performing (i.e., having a leach rate or D_{obs} value in the middle of the 24 acceptable mixes) monoliths for each of the key constituents of concern (Cr, Tc, I, nitrate, nitrite, and sodium) were chosen. As mentioned, uranium did not leach out of most of the 26 Screening Test mixes at measureable concentrations, so no effort was made to rank Cast Stone mixes for uranium release. Further, because the leach rates of iodide, nitrate, nitrite, and sodium for any given mix were very similar, choosing six monoliths from the 48 monoliths (duplicates for each of the 24 acceptable mixes) that were leached in the screening tests satisfied our criteria of continuing to leach a best, worst, and median performing monolith. Adding in the other five monoliths captured the best, worst, and median performing monoliths for the Cr and Tc.

Table 4.1 shows the average of interval D_{obs} values for the constituents of interest for the eleven monoliths being leached in DIW up through either 1037 or 1042 days depending on the mix. The original screening tests broke the 26 mixes into two groups that initiated leach testing off-set by five days to allow sampling to be accomplished in one eight hour work day. After the selection of only eleven monoliths for continuation of leaching was accomplished, all the extended leach test sampling was accomplished on the same day, thus causing the five day difference in cumulative leach times for sampling times beyond 119 days.

Three averages are provided that include using the original Screening Test data for the duplicate monolith for each mix, which was leached up through 91 days. The first average of interval D_{obs} values used the interval leaching data from day 7 through the last sampling interval, either 1037 days or 1042 days. The second average of interval D_{obs} values includes only interval leach data for both monoliths over the time period 28 days through 63 days. This was the average used in the original screening report to perform detailed statistical analyses (see Westsik et al. 2013a for details). The third average of interval D_{obs} values provided in Table 4.1 uses the interval leach data from 28 days out through the last sampling (either 1037 days or 1042 days).

Table 4.1. Average of Interval D_{obs} Values (cm^2/s) for Designated Sampling Periods

Monolith	D_{obs} 7d to 1037/1042 d Avg.	σ	D_{obs} 28 d to 63 d Avg.	σ	D_{obs} 28 d to 1037/1042 Avg.	σ
	Tc(VII)					
T18LCS2-7.8RAS	1.90E-11	2.11E-11	5.69E-12	1.05E-12	2.02E-11	2.27E-11
T14LCS2-7.8HIS	9.44E-12	2.30E-12	9.60E-12	1.79E-12	8.88E-12	1.86E-12
T15HCS1-7.8HIS	5.49E-11	4.92E-11	4.62E-11	1.04E-11	3.47E-11	1.40E-11
T24HCS1-5HIA	7.06E-11	2.11E-11	6.52E-11	6.74E-12	7.24E-11	1.94E-11
T5HCS1-7.8AVG	4.51E-11	2.39E-11	7.16E-11	1.47E-11	4.40E-11	2.54E-11
T13LCS2-5AVG	7.38E-11	2.83E-11	8.27E-11	1.10E-11	8.08E-11	2.50E-11
T21LCS1-7.8HIS	1.63E-10	1.69E-10	9.40E-11	7.25E-11	1.91E-10	1.71E-10
T10HCS1-5HIS	1.44E-10	8.49E-11	1.20E-10	1.42E-11	1.13E-10	2.29E-11
T16HCS1-7.8RAS	1.45E-10	3.33E-11	1.57E-10	1.65E-11	1.41E-10	3.44E-11
T17LCS2-5HIA	2.15E-10	2.49E-10	1.76E-10	2.81E-11	2.24E-10	2.73E-10
T8LCS1-5RAS	1.84E-10	8.73E-11	2.16E-10	9.48E-12	1.56E-10	5.61E-11
	Iodide					
T8LCS1-5RAS	2.23E-09	1.05E-09	2.55E-09	3.87E-10	1.87E-09	6.55E-10
T18LCS2-7.8RAS	3.32E-09	1.49E-09	3.15E-09	4.39E-10	2.75E-09	5.62E-10
T10HCS1-5HIS	4.74E-09	4.97E-09	4.44E-09	1.59E-09	2.78E-09	1.70E-09
T5HCS1-7.8AVG	5.38E-09	6.91E-09	5.30E-09	7.89E-10	2.68E-09	2.75E-09
T15HCS1-7.8HIS	5.32E-09	5.49E-09	5.68E-09	2.01E-09	3.18E-09	2.47E-09
T14LCS2-7.8HIS	6.21E-09	8.22E-09	5.70E-09	2.26E-09	3.00E-09	2.57E-09
T17LCS2-5HIA	7.95E-09	7.81E-09	7.58E-09	2.52E-09	4.90E-09	3.14E-09
T16HCS1-7.8RAS	6.74E-09	4.61E-09	7.86E-09	1.80E-09	5.04E-09	2.69E-09
T24HCS1-5HIA	7.32E-09	8.52E-09	8.13E-09	2.92E-09	4.16E-09	3.78E-09
T13LCS2-5AVG	7.93E-09	8.28E-09	8.48E-09	2.89E-09	4.72E-09	3.73E-09
T21LCS1-7.8HIS	1.62E-08	2.26E-08	1.68E-08	7.63E-09	7.43E-09	8.86E-09
	Na					
T8LCS1-5RAS	2.12E-09	9.66E-10	2.40E-09	2.84E-10	1.78E-09	6.05E-10
T18LCS2-7.8RAS	2.75E-09	1.09E-09	2.85E-09	2.27E-10	2.36E-09	6.40E-10
T14LCS2-7.8HIS	3.93E-09	4.01E-09	4.45E-09	1.08E-09	2.43E-09	1.81E-09
T5HCS1-7.8AVG	5.27E-09	5.22E-09	5.58E-09	2.04E-09	3.32E-09	2.33E-09
T13LCS2-5AVG	5.81E-09	5.57E-09	6.43E-09	1.80E-09	3.69E-09	2.65E-09
T17LCS2-5HIA	6.05E-09	5.26E-09	6.67E-09	1.87E-09	4.06E-09	2.76E-09
T16HCS1-7.8RAS	5.83E-09	3.19E-09	7.07E-09	1.11E-09	4.73E-09	2.17E-09
T15HCS1-7.8HIS	6.03E-09	4.33E-09	7.35E-09	1.55E-09	4.47E-09	2.69E-09
T10HCS1-5HIS	7.32E-09	6.60E-09	7.54E-09	1.99E-09	4.72E-09	2.75E-09
T24HCS1-5HIA	7.88E-09	7.33E-09	9.79E-09	2.53E-09	5.26E-09	4.11E-09
T21LCS1-7.8HIS	9.51E-09	1.19E-08	1.05E-08	3.30E-09	4.83E-09	5.00E-09
	NO_2					
T8LCS1-5RAS	2.35E-09	1.36E-09	2.44E-09	4.29E-10	1.86E-09	6.22E-10
T18LCS2-7.8RAS	3.11E-09	1.46E-09	2.95E-09	2.93E-10	2.55E-09	5.69E-10

	D_{obs} 7d to 1037/1042 d Avg.	σ	D_{obs} 28 d to 63 d Avg.	σ	D_{obs} 28 d to 1037/1042 Avg.	σ
T10HCS1-5HIS	6.37E-09	7.70E-09	5.57E-09	1.84E-09	3.43E-09	2.17E-09
T14LCS2-7.8HIS	6.59E-09	9.50E-09	5.93E-09	2.11E-09	3.02E-09	2.71E-09
T5HCS1-7.8AVG	6.52E-09	8.28E-09	6.23E-09	3.08E-09	3.37E-09	3.08E-09
T15HCS1-7.8HIS	6.95E-09	6.58E-09	7.18E-09	2.73E-09	4.35E-09	3.06E-09
T16HCS1-7.8RAS	6.63E-09	4.64E-09	7.36E-09	1.71E-09	4.87E-09	2.57E-09
T17LCS2-5HIA	7.71E-09	7.72E-09	7.37E-09	2.25E-09	4.56E-09	2.97E-09
T13LCS2-5AVG	7.78E-09	9.39E-09	7.46E-09	2.51E-09	4.14E-09	3.28E-09
T24HCS1-5HIA	7.32E-09	7.83E-09	7.95E-09	1.63E-09	4.39E-09	3.29E-09
T21LCS1-7.8HIS	1.30E-08	1.65E-08	1.38E-08	4.12E-09	6.56E-09	6.45E-09
	NO_3					
T8LCS1-5RAS	2.58E-09	1.21E-09	2.60E-09	5.21E-10	2.14E-09	5.83E-10
T18LCS2-7.8RAS	3.74E-09	1.46E-09	3.33E-09	4.94E-10	3.18E-09	4.99E-10
T10HCS1-5HIS	6.88E-09	7.43E-09	6.10E-09	2.25E-09	3.97E-09	2.27E-09
T5HCS1-7.8AVG	6.62E-09	8.28E-09	6.18E-09	3.11E-09	3.40E-09	3.03E-09
T14LCS2-7.8HIS	6.71E-09	8.70E-09	6.26E-09	2.47E-09	3.36E-09	2.78E-09
T15HCS1-7.8HIS	6.86E-09	6.42E-09	7.02E-09	2.61E-09	4.35E-09	2.85E-09
T17LCS2-5HIA	8.71E-09	8.19E-09	7.71E-09	2.60E-09	5.64E-09	3.99E-09
T16HCS1-7.8RAS	7.25E-09	4.57E-09	8.04E-09	2.03E-09	5.56E-09	2.58E-09
T13LCS2-5AVG	8.53E-09	9.44E-09	8.40E-09	3.23E-09	4.82E-09	3.67E-09
T24HCS1-5HIA	8.71E-09	9.55E-09	9.62E-09	2.93E-09	5.20E-09	4.16E-09
T21LCS1-7.8HIS	1.42E-08	1.89E-08	1.51E-08	5.54E-09	6.95E-09	7.45E-09
	Cr					
T14LCS2-7.8HIS	9.64E-15	3.41E-15	9.91E-15	2.25E-15	9.22E-15	3.11E-15
T18LCS2-7.8RAS	<2.73E-14	1.73E-14	<1.84E-14	1.19E-14	<2.92E-14	1.82E-14
T13LCS2-5AVG	6.41E-14	3.86E-14	5.50E-14	9.00E-15	5.19E-14	1.97E-14
T5HCS1-7.8AVG	8.86E-14	1.03E-13	8.00E-14	2.82E-14	6.72E-14	2.36E-14
T24HCS1-5HIA	1.20E-13	1.33E-13	9.41E-14	3.68E-14	7.93E-14	3.14E-14
T21LCS1-7.8HIS	1.74E-13	1.10E-13	1.51E-13	6.57E-14	1.97E-13	1.04E-13
T17LCS2-5HIA	2.01E-13	1.81E-13	1.65E-13	2.53E-14	2.07E-13	1.98E-13
T16HCS1-7.8RAS	2.90E-13	2.48E-13	2.44E-13	5.39E-14	1.98E-13	8.21E-14
T15HCS1-7.8HIS	7.56E-13	1.41E-12	3.16E-13	3.07E-13	1.72E-13	2.22E-13
T10HCS1-5HIS	7.90E-13	9.98E-13	6.99E-13	3.99E-13	4.64E-13	3.33E-13
T8LCS1-5RAS	5.70E-13	3.24E-13	7.15E-13	1.02E-13	4.75E-13	2.42E-13

Note: None of the interval averaged D_{obs} values has been corrected for inventory depletion. Also, the averaged values include calculated values for the duplicate monoliths that were discarded after 91-d of leaching (see Westsik et al. (2013) Appendix D and Appendix C of Rev. 0 of this report (Serne et al. 2015) for some minor corrections to diffusion coefficients found in Appendix D of Westsik et al..

The average interval D_{obs} values for each constituent have been sorted using the interval average from 28 to 63 days (same sampling period used in the Westsik et al. 2013a screening report) to show the ranking from lowest value to highest value. In general, the D_{obs} range for each constituent varies by two orders of magnitude for Cr(VI) and Tc(VII) and by less than one order of magnitude for the more leachable constituents (iodide, sodium, nitrate, and nitrite). Interestingly, the rankings of the monoliths with the lowest and highest average interval D_{obs} values are quite similar for nitrate, nitrite, iodide, and to a lesser extent, sodium. This suggests that for a given Cast Stone mix ratio, the leach properties of these four constituents are quite similar. The monolith rankings for ^{99}Tc and Cr are also quite similar, but differ from the other four more leachable constituents. This suggests that whatever mechanism(s) are controlling ^{99}Tc and Cr leach properties are similar. We hypothesize that the controlling mechanism for these two constituents is re-oxidation, to higher solubility species from the initially reduced (low solubility) species, followed by diffusion through the tortuous Cast Stone matrix. Table 4.1 also shows the standard deviation for each of the three specified interval D_{obs} averages. In general, the standard deviations for the more leachable nitrate, nitrite, iodide, and sodium are large because the interval D_{obs} values are, in most instances, continually decreasing with increasing time. A key reason for the decrease in D_{obs} values for these more leachable constituents is the depletion of the starting inventory. For the most leachable monoliths (T13LCS2-5AVG, T21LCS1-7.8HIS, and T24HCS1-5HIA, shown in Table 4.1), nitrite, nitrate, and iodide reach >20% depletion between 7 and 14 days of cumulative leaching. Recall that both EPA Method 1315 and ANS16.1 protocol states that when a monolith releases >20% of its starting inventory for a constituent, that the observed D_{obs} calculated using Eq. 3.1 through Eq. 3.3 is no longer accurate. Even the two least leachable monoliths (T8LCS1-5RAS and T18LCS2-7.8RAS) deplete >20% of the nitrite, nitrate, sodium, and iodide generally between 42 and 77 days of leaching. However, the eleven monoliths leached in this test at most leached only 12.4% of the starting ^{99}Tc inventory and 1% of the starting Cr(VI) inventory. Regardless, we recommend using the interval averaged 28 to 63-day D_{obs} values in Table 4.1 (monoliths leached in DIW) as the upper range estimates for all key constituents for future predictive modeling activities. In the following subsections, we present similar interval averaged D_{obs} values for monoliths leached in VZP that are our recommended "base-case," or most probable, D_{obs} values (most representative of the IDF subsurface burial environment). Table 4.3, Table 4.4, Table 4.6, and Table 4.8 interval averaged D_{obs} values for monoliths leached in VZP have not been corrected for inventory depletion. Bear in mind that when the recommended D_{obs} values cited herein are used in future IDF PA calculations, different values of COC starting inventory and different waste form surface area-to-volume ratios will be used such that the percentage of mass leached likely will not exceed the 20% until long time periods have elapsed. Further, although not addressed in this work, temperature corrections (the IDF burial environment is cooler (~15°C) than the laboratory temperatures (~23°C) such that temperature correction for D_{obs} values should be considered in future IDF PA activities.[1]

A second plausible reason for the decrease in D_{obs} values versus time (shown in Table 4.1) is the long leaching intervals used in the extended leach tests out beyond 91 days. Such long intervals between refreshing the eluent (47, 161, 272 or as large as 305 days when the project was shut off in 2014) could have allowed the eluate concentrations of leachable constituents to increase enough to significantly lower the concentration gradient as a driving force for diffusion. Eluate concentrations for each sampling period are shown in Appendix B for each monolith leach test, the concentration of the mobile constituents (iodide, nitrate, nitrite, and sodium) in the eluates from the leach intervals beyond 91 days, are generally significantly lower than eluates from cumulative leach times between 1 day and 42 days, which were times recommended in EPA 1315 Methodology that should be adequate to avoid significant build-up in eluates to reduce the diffusion gradient. For most of the 11 monoliths whose leach testing continued beyond 91 days only the sampling at cumulative time 557 or 562 days shows eluate concentrations significantly larger than those observed during the first 42 days of leaching. The 557 to 562 day cumulative leaching interval had a 305 day gap between samplings (between 257 and 562 or between 252 and 557 cumulative days no eluent replacement occurred) such that the 557/562-d eluates contained

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[1] At this time we have no data on the impacts on D_{obs} values when leach tests are performed at lower temperatures (such as 15°C). The effect is likely small and within the uncertainties in the laboratory leach data generated to date.

higher concentrations than all other eluates. Therefore, because all but the 557/562 cumulative day leach intervals used in the extended leach testing resulted in lower eluate concentrations than early (-1 to 42-d cumulative eluates, the lowering of the diffusion gradient is likely less of the cause for the observed decrease in D_{obs} values as cumulative time increases. Rather, the depletion of starting inventories for the mobile constituents is likely the main cause for decreasing D_{obs} values.

A third explanation for the decrease in D_{obs} values versus time would be preferential diffusion of COCs from an outer ring out into the eluate with the interior center of the monolith not “feeding” constituents to the outer ring via diffusion gradients. This conceptual model would be equivalent to stating the physical structure of the monolith and location of the COCs within the monoliths are not homogeneous. We have no objective evidence that the cured monoliths are not homogeneous in physical structure or distribution of COCs at the start of the leach testing.

Photographs of some of the most leach resistant and least leach resistant monoliths after ~555 days of leaching are shown in Figure 4.1. The upper two photographs are monoliths from Mix 8 and Mix 18 that showed the lowest D_{obs} values for the mobile constituents, (nitrate, nitrite, iodide, and sodium), as shown in the rankings in Table 4.1. The bottom two photographs are monoliths from Mixes 21 and 24, which showed the highest D_{obs} values for the mobile constituents. All four photographs show that the monoliths have remained intact throughout the 550 to 555 days of leaching. Two of the photographs show some white precipitate either clinging to the monolith’s surface or concentrated along micro-cracks that were present when the monoliths were removed from the plastic molds. The white precipitate, found to be calcium carbonate, will be discussed in detail in the section on Archive Leach Test Suite monoliths (Section 4.2). Compared to monoliths leached in VZP, which was used in the Archive Leach Test Suite, I-Loading Suite, and Tc-Gluconate Spiked Suite, these monoliths leached in DIW show much less secondary precipitates on their surfaces. This might be expected, given that DIW contains no solutes to interact with the Cast Stone monoliths. The slight evidence of calcium carbonate precipitation on the monolith surfaces likely originated from carbon dioxide present in the container head space air and the fresh DIW eluent replenished with each sample exchange, as well as diffusion through the plastic leach containers. Calcium also needs to diffuse out to the monoliths before reacting with dissolved carbon dioxide in the DIW right at the surface of the monoliths. All-in-all, the photographs in Figure 4.1 suggest that the monoliths have little signs of significant degradation or extensive secondary mineral formation on the outer surfaces after the first 20 months of leaching in DIW. Photographs taken after 765/770 and 1037/1042 days of cumulative leaching show more secondary mineral formation on outer surfaces or along linear surface micro-cracks for these monoliths leached in DIW but the surface precipitates are less than observed on Archived Suite monoliths leached in VZP to be discussed in the next section.

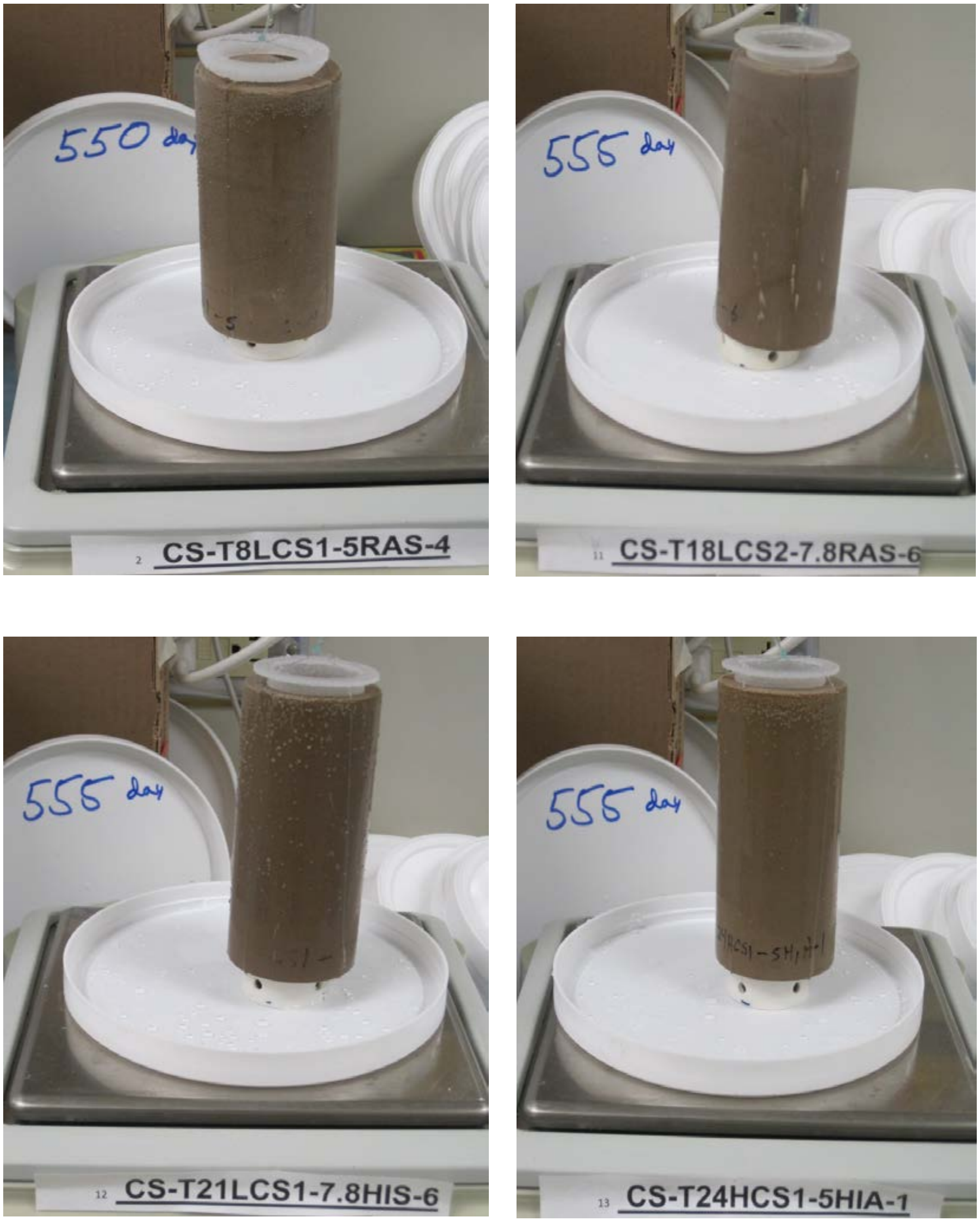


Figure 4.1. Photographs of •Screening Leach Tests –Extended Suite Monoliths after ~555 days of Leaching in DIW

Results from the screening tests (see Westsik et al. 2013a), which leached duplicate Cast Stone monoliths with the 26 mix compositions shown in Figure A.1 for 91 days, suggest that there were no statistically significant correlations between the leach properties of any of the key constituents with the type of waste simulant, the source of the dry blend, the free-water-to-dry blend ratio, or wet slurry properties. Therefore, despite finding a two order of magnitude range in ^{99}Tc and Cr observed diffusivities and almost an order of magnitude range in observed diffusivities for the more leachable anions (nitrate, nitrite, iodide) and sodium, we have not identified correlations with the Cast Stone or waste simulants that shed light on the controlling mechanism(s) that lead to the variation in leach properties.

After the 765 or 770-day sampling leach testing for some of the extended leach monoliths were placed in storage, within a small volume of their respective final eluate, so that solid phase characterization can be performed to gain insight into weathering processes and weathering rates. The results of the **solid phase** characterization will be documented in a technical report in the last quarter of 2016. The monoliths that were stored for solid phase characterization and those that continue to be leached are identified in Table 4.2.

Table 4.2. Status of Extended Leach Suite Monoliths

Number	Mix #	Monolith IDs Leached in DIW	Status (February 18, 2015) after 765/770 days leaching
1	5	CS-T5HCS1-7.8AVG-2	Stored for solid phase characterization
2	8	CS-T8LCS1-5RAS-4	Actively leaching in DIW
3	10	CS-T10HCS1-5HIS-6	Stored for solid phase characterization
4	13	CS-T13LCS2-5AVG-2	Stored for solid phase characterization
5	14	CS-T14LCS2-7.8HIS-3	Actively leaching in DIW
6	15	CS-T15HCS1-7.8HIS-1	Stored for solid phase characterization
7	16	CS-T16HCS1-7.8RAS-3	Stored for solid phase characterization
8	17	CS-T17LCS2-5HIA-3	Stored for solid phase characterization
9	18	CS-T18LCS2-7.8RAS-6	Actively leaching in DIW
10	21	CS-T21LCS1-7.8HIS-6	Actively leaching in DIW
11	24	CS-T24HCS1-5HIA-1	Actively leaching in DIW

4.2 Archive Leach Test Suite Results

The objective of the Archive Leach Test Suite of leach tests was to start using a more IDF-realistic leachant/eluent than the DIW eluent used throughout the screening tests. The more realistic eluent is the VZP described in Section 2.3. During the fabrication of the original 26 Cast Stone mixes for the Screening Test task, six Tc, I, Cr, and U spiked monoliths were poured and cured for each mix. Only two of the spiked monoliths for each mix were leach tested in the Screening Test task, thus four monoliths remained in storage at room temperature in containers that maintained high relative humidity. These stored monoliths are called archived monoliths for this report.

Table 4.3 lists the monoliths and eluent used for this Archive Leach Test Suite of tests. We chose two of the available archived monoliths from the original Screening Test task for leaching in VZP and one for leaching in DIW. Table A.2 lists the curing times for the Archive Suite of monolith that had been stored in double wrapped plastic bags with the outer bag containing moist paper towels that were re-wetted periodically. Because two monoliths of each mix were already leached in DIW during the Screening Tests, documented in Westsik et al. (2013a), we used only one of the archived monoliths (stored for an additional 176 to 181 days after the Screening Leach Tests were initiated). We leached two archived monoliths in VZP so that replicates would allow some measure of precision. For the original Screening Test mix #5, only two monoliths were still available, so there could not be two archived monoliths leached in VZP. Therefore, one Mix #5 monolith (T5HCS1-7.8 AVG-3) was leached in VZP and one (T5HCS1-7.8 AVG-6) in DIW in the Archive Leach Test Suite.

Table 4.4 presents average interval D_{obs} values for the Archive Leach Test Suite monoliths similar to the results shown in Table 4.1 for the Extended Suite of monoliths, which were leached solely in DIW. The Archive Suite leach test results for many of the Archived Suite monoliths are now available through 861 days of leaching in comparison to the Extended Suite, which has leach data through 1037/1042 days, dependent on the Cast Stone mix. Some of the Archived Suite monoliths were stored in their respective final eluates after 590 days of leaching to allow for post –leaching solid phase characterization. Again, the interval D_{obs} values for the two replicates leached in VZP for the four different sampling periods were averaged (28-, 42-, 49- and 63-days), as noted previously in the Extended Suite. For the monoliths leached in DIW, we have just the one datum per sampling interval and average across the noted sampling intervals. Table 4.4 suggests that there is more leaching occurring from monoliths of a particular mix composition when the eluent is DIW than when using VZP as the eluent. This is especially evident for the leaching of ^{99}Tc and Cr. Figure 4.2 shows bar charts for the averaged interval D_{obs} values for the 28-day to 63-day data. We wish to point out that the 28-d to 63-d averaged D_{obs} values plotted in Figure 4.2 (data from Table 4.4) show an average difference of a factor of ~16 larger Tc observed diffusivities when leached in DIW in comparison to values leached in VZP. The range in the ratio of 28-d to 63-d Tc D_{obs} values in DIW to D_{obs} values in VZP is 2.4 to 53, with Mix 14 having the lowest ratio and Mix 21 having the highest ratio. The range in the DIW-to-VZP D_{obs} ratio for the more mobile constituents is much lower but consistently shows iodide, sodium, nitrite and nitrate leases slightly faster in DIW than in VZP. The range in the DIW-to-VZP D_{obs} ratio for these four mobile constituents ranges from 1.06 to 2.03 and consistently is highest for Mix 17 and lowest for Mix 18 and/or Mix 8. No statistical evaluations were attempted to evaluate whether there are relationships between Mix composition and D_{obs} values for either eluents (VZP vs DIW) because extensive statistical evaluations of relationships between Mix simulant type, simulant total Na composition, Mix water-to-dry blend ratio, dry blend FA and BFS source, cured compressive strength, and Mix wet slurry properties performed by Westsik et al. (2013) did not find any significant correlations.

When leached in VZP, most of the averaged interval D_{obs} values for Cr are not readily determinable. Because many of the VZP eluates did not contain detectable concentrations of Cr, we resorted to assuming that these eluates contained Cr concentrations at the sample detection limit. This assumption led to designating the calculated D_{obs} values as “less than values”, e.g., < x where x is the numerical value of D_{obs} in units of cm^2/sec. In contrast, when leached in DIW, only one averaged interval Cr D_{obs} data set contained eluates with Cr concentrations below detection. Table 4.4 presents the average interval D_{obs} values for each COC sorted from low to high leaching as quantified by the 28- to 63-d averaged interval D_{obs} values.

The Tc in the Archive Leach Test Suite monoliths that were leached in VZP show a change of a factor of 40 from the best performing Cast Stone mix (T24) to the worst performing mix (T16) for the 28- to 63-day averaged interval D_{obs} values. The iodide in the Archive Leach Test Suite monoliths that were leached in VZP shows a change of a factor of 6 from the best performing Cast Stone mix (T10) to the worst performing mix (T21) for the 28- to 63-day averaged interval D_{obs} values. The sodium in the Archive Leach Test Suite monoliths that were leached in VZP also show a change of a factor of 6 from the best performing Cast Stone mix (T14) to the worst performing mix (T21) for the 28- to 63-day averaged interval D_{obs} values. The nitrite in the Archive Leach Test Suite monoliths that were leached in VZP show a change of a factor of 3 from the best performing Cast Stone mix (T18) to the worst performing mix (T21) for the 28- to 63-day averaged interval D_{obs} values. The nitrate in the Archive Leach Test Suite monoliths that were leached in VZP show a change of a factor of 5.4 from the best performing Cast Stone mix (T8) to the worst performing mix (T21) for the 28- to 63-day averaged interval D_{obs} values. The Cr in the Archive Leach Test Suite monoliths had many values less than the lower quantification limit for the VZP eluates; thus, no quantitative ranking of Cast Stone mixes is possible. Average interval D_{obs} values for Cr in DIW except Test 18 are real values. Test 18 D_{obs} values in DIW are less than values.

Table 4.3. Listing of Archived Monoliths and Eluent Type Used in Leach Tests

Test or Mix #	VPZ Eluent	DIW Eluent
Test 3	T3HCS2-7.8AVG-3 T3HCS2-7.8AVG-4	T3HCS2-7.8AVG-5
Test 5	T5HCS1-7.8 AVG-3	T5HCS1-7.8 AVG-6
Test 8	T8LCS1-5RAS-2 T8LCS1-5RAS-3	T8LCS1-5RAS-5
Test 10	T10HCS1-5HIS-3 T10HCS1-5HIS-4	T10HCS1-5HIS-5
Test 13	T13LCS2-5AVG-3 T13LCS2-5AVG-4	T13LCS2-5AVG-5
Test 14	T14LCS2-7.8HIS-4 T14LCS2-7.8HIS-5	T14LCS2-7.8HIS-6
Test 15	T15HCS1-7.8HIS-3 T15HCS1-7.8HIS-5	T15HCS1-7.8HIS-6
Test 16	T16HCS1-7.8RAS-4 T16HCS1-7.8RAS-5	T16HCS1-7.8RAS-6
Test 17	T17LCS2-5HIA-2 T17LCS2-5HIA-4	T17LCS2-5HIA-6
Test 18	T18LCS2-7.8RAS-2 T18LCS2-7.8RAS-3	T18LCS2-7.8RAS-4
Test 21	T21LCS1-7.8HIS-2 T21LCS1-7.8HIS-4	T21LCS1-7.8HIS-5
Test 24	T24HCS1-5HIA-4 T24HCS1-5HIA-5	T24HCS1-5HIA-6

Note: monoliths that were placed in storage after 590 days of leaching are highlighted in gray shading.

Table 4.4. Interval Averaged D_{obs} Values (cm^2/s) for Archived Monoliths Leached in Either VZP or DIW—Sorted Based on 28 to 63 Day Interval Average

	Eluent = VZP							**Eluent = DIW**					
	D_{obs} 7d up to 861d Avg.	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d up to 861d Avg.**	**σ**		**D_{obs} 7d up to 861d Avg.**	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d up to 861d Avg.**	**σ**
Monolith	**Tc**						**Monolith**	**Tc**					
T24H-5HIA	6.38E-13	2.45E-13	7.55E-13	2.38E-13	6.15E-13	2.54E-13	T24H-5HIA	1.21E-11	1.05E-11	5.96E-12	4.70E-13	1.42E-11	1.05E-11
T15H-7.8HIS	1.22E-12	5.89E-13	1.54E-12	2.95E-13	1.09E-12	5.62E-13	T14L-7.8HIS	7.19E-12	1.94E-12	7.13E-12	5.94E-13	7.11E-12	2.14E-12
T5H-7.8AVG	1.29E-12	6.76E-13	1.60E-12	2.63E-13	1.10E-12	6.03E-13	T18L2-7.8RAS	2.90E-11	1.25E-11	2.36E-11	3.10E-12	3.13E-11	1.28E-11
T3H2-7.8AVG	2.17E-12	1.04E-12	2.70E-12	5.94E-13	2.11E-12	1.13E-12	T15H-7.8HIS	3.08E-11	9.76E-12	3.39E-11	6.93E-12	3.36E-11	7.99E-12
T14L-7.8HIS	1.95E-12	1.71E-12	2.97E-12	2.17E-12	1.98E-12	1.89E-12	T13L-5AVG-5	8.59E-11	9.79E-11	3.50E-11	1.13E-11	1.05E-10	1.01E-10
T8L-5RAS	6.63E-12	4.06E-12	5.17E-12	8.70E-13	5.02E-12	2.15E-12	T5H-7.8AVG	3.72E-11	1.47E-11	4.00E-11	1.10E-11	4.18E-11	1.19E-11
T21L-7.8HIS	4.97E-12	2.75E-12	5.73E-12	1.85E-12	5.68E-12	2.54E-12	T3H2-7.8AVG	9.59E-11	7.48E-11	5.10E-11	1.43E-11	1.12E-10	7.39E-11
T18L2-7.8RAS	5.47E-12	1.84E-12	6.03E-12	7.74E-13	4.98E-12	1.66E-12	T10H-5HIS	7.80E-11	1.20E-11	8.73E-11	5.55E-12	8.13E-11	1.02E-11
T10H-5HIS	5.83E-12	3.00E-12	6.90E-12	1.08E-12	4.81E-12	2.37E-12	T8L-5RAS	1.11E-10	3.32E-11	1.41E-10	9.71E-12	1.12E-10	3.56E-11
T13L-5AVG-5	1.42E-11	1.29E-11	7.86E-12	2.72E-12	1.67E-11	1.33E-11	T16H-7.8RAS	1.35E-10	3.27E-11	1.58E-10	1.56E-11	1.42E-10	3.33E-11
T17L-5HIA	2.10E-11	1.33E-11	1.84E-11	4.65E-12	2.27E-11	1.45E-11	T17L-5HIA	2.17E-10	9.76E-11	1.89E-10	1.50E-11	2.36E-10	1.00E-10
T16H-7.8RAS	2.18E-11	1.08E-11	3.02E-11	5.88E-12	2.08E-11	1.17E-11	T21L-7.8HIS	2.31E-10	1.12E-10	3.05E-10	7.48E-11	2.62E-10	9.41E-11
	I							**I**					
T10H-5HIS	1.68E-09	5.56E-10	1.88E-09	1.77E-10	1.51E-09	4.69E-10	T10H-5HIS	2.31E-09	3.08E-10	2.48E-09	9.00E-11	2.26E-09	3.23E-10
T24H-5HIA	1.91E-09	1.29E-09	2.06E-09	9.48E-10	1.40E-09	7.18E-10	T24H-5HIA	2.34E-09	1.08E-09	2.48E-09	5.96E-10	1.96E-09	7.01E-10
T8L-5RAS	1.89E-09	7.45E-10	2.20E-09	2.85E-10	1.65E-09	6.05E-10	T8L-5RAS	2.19E-09	1.07E-09	2.62E-09	4.60E-10	1.85E-09	8.43E-10
T15H-7.8HIS	1.99E-09	1.09E-09	2.34E-09	4.26E-10	1.61E-09	8.43E-10	T18L2-7.8RAS	3.06E-09	8.52E-10	3.04E-09	4.76E-10	2.77E-09	5.81E-10
T13L-5AVG-5	2.97E-09	1.48E-09	2.68E-09	7.80E-10	2.34E-09	7.28E-10	T13L-5AVG-5	3.45E-09	1.38E-09	3.76E-09	2.37E-10	2.96E-09	9.94E-10
T5H-7.8AVG	2.63E-09	1.40E-09	2.77E-09	3.23E-10	2.07E-09	8.28E-10	T15H-7.8HIS	3.42E-09	1.39E-09	4.15E-09	9.42E-10	3.11E-09	1.38E-09
T18L2-7.8RAS	3.16E-09	1.43E-09	2.88E-09	7.75E-10	2.56E-09	6.77E-10	T14L-7.8HIS	4.17E-09	1.63E-09	4.25E-09	4.80E-10	3.54E-09	8.48E-10
T16H-7.8RAS	2.69E-09	1.26E-09	2.91E-09	4.23E-10	2.21E-09	8.71E-10	T5H-7.8AVG	3.96E-09	1.45E-09	4.45E-09	5.15E-10	3.56E-09	1.34E-09
T17L-5HIA	4.08E-09	2.32E-09	3.10E-09	8.72E-10	3.04E-09	8.82E-10	T16H-7.8RAS	4.25E-09	1.30E-09	4.84E-09	8.18E-10	4.01E-09	1.34E-09

	Eluent = VZP							Eluent = DIW					
	D_{obs} 7d up to 861d Avg.	σ	D_{obs} 28d to 63d Avg.	σ	D_{obs} 28d up to 861d Avg.	σ		D_{obs} 7d up to 861d Avg.	σ	D_{obs} 28d to 63d Avg.	σ	D_{obs} 28d up to 861d Avg.	σ
	I		I		I		I		I		I		I
T14L-7.8HIS	3.63E-09	1.24E-09	3.68E-09	7.97E-10	3.17E-09	7.81E-10	T17L-5HIA	5.65E-09	3.26E-09	6.30E-09	1.37E-09	4.47E-09	2.36E-09
T3H2-7.8AVG	4.69E-09	2.50E-09	4.90E-09	4.66E-10	3.76E-09	1.37E-09	T3H2-7.8AVG	6.42E-09	2.10E-09	6.63E-09	1.03E-09	5.76E-09	1.50E-09
T21L-7.8HIS	9.46E-09	6.79E-09	1.11E-08	2.93E-09	7.03E-09	4.67E-09	T21L-7.8HIS	1.28E-08	1.06E-08	1.58E-08	5.14E-09	9.11E-09	7.63E-09
	Na							Na					
T14L-7.8HIS	1.72E-09	7.67E-10	1.83E-09	3.32E-10	1.43E-09	4.58E-10	T14L-7.8HIS	2.30E-09	9.61E-10	2.47E-09	1.84E-10	1.99E-09	7.05E-10
T13L-5AVG-5	2.10E-09	1.25E-09	1.93E-09	3.56E-10	1.56E-09	5.17E-10	T18L2-7.8RAS	2.44E-09	7.27E-10	2.52E-09	2.22E-10	2.21E-09	5.72E-10
T17L-5HIA	2.60E-09	1.63E-09	2.15E-09	3.64E-10	1.89E-09	6.33E-10	T8L-5RAS	2.12E-09	8.74E-10	2.60E-09	2.42E-10	1.89E-09	7.79E-10
T8L-5RAS	1.78E-09	8.10E-10	2.21E-09	2.92E-10	1.54E-09	7.06E-10	T13L-5AVG-5	2.76E-09	1.17E-09	3.03E-09	2.85E-10	2.41E-09	1.00E-09
T18L2-7.8RAS	2.24E-09	1.02E-09	2.21E-09	4.16E-10	1.83E-09	5.00E-10	T24H-5HIA	3.49E-09	1.81E-09	3.70E-09	5.11E-10	2.85E-09	1.20E-09
T16H-7.8RAS	2.46E-09	1.23E-09	2.65E-09	3.51E-10	1.98E-09	7.75E-10	T10H-5HIS	3.88E-09	1.30E-09	4.09E-09	3.97E-10	3.53E-09	1.20E-09
T5H-7.8AVG	2.63E-09	1.40E-09	2.77E-09	3.23E-10	2.07E-09	8.28E-10	T17L-5HIA	4.01E-09	2.10E-09	4.38E-09	4.13E-10	3.25E-09	1.53E-09
T10H-5HIS	2.78E-09	1.24E-09	3.14E-09	2.48E-10	2.34E-09	9.13E-10	T5H-7.8AVG	3.96E-09	1.45E-09	4.45E-09	5.15E-10	3.56E-09	1.34E-09
T24H-5HIA	2.82E-09	1.97E-09	3.14E-09	5.13E-10	2.06E-09	1.16E-09	T16H-7.8RAS	4.21E-09	1.32E-09	4.74E-09	5.76E-10	3.90E-09	1.27E-09
T15H-7.8HIS	2.78E-09	1.39E-09	3.19E-09	3.32E-10	2.30E-09	1.01E-09	T15H-7.8HIS	4.30E-09	1.52E-09	4.94E-09	7.51E-10	3.92E-09	1.44E-09
T3H2-7.8AVG	3.75E-09	2.14E-09	4.37E-09	4.96E-10	3.00E-09	1.47E-09	T3H2-7.8AVG	5.16E-09	2.37E-09	5.52E-09	5.89E-10	4.50E-09	2.04E-09
T21L-7.8HIS	4.12E-09	2.69E-09	4.92E-09	1.07E-09	3.18E-09	1.97E-09	T21L-7.8HIS	6.47E-09	4.63E-09	8.15E-09	1.08E-09	4.91E-09	3.40E-09
	NO_2							NO_2					
T18L2-7.8RAS	2.24E-09	1.08E-09	2.04E-09	4.32E-10	1.91E-09	9.08E-10	T18L2-7.8RAS	2.28E-09	6.45E-10	2.40E-09	2.91E-10	2.06E-09	4.40E-10
T8L-5RAS	1.82E-09	9.82E-10	2.14E-09	3.10E-10	1.66E-09	1.02E-09	T8L-5RAS	2.05E-09	7.90E-10	2.47E-09	3.17E-10	1.82E-09	6.75E-10
T13L-5AVG-5	3.12E-09	3.77E-09	2.26E-09	4.43E-10	2.94E-09	4.22E-09	T24H-5HIA	2.51E-09	1.23E-09	2.76E-09	3.26E-10	2.07E-09	7.72E-10
T24H-5HIA	2.10E-09	1.28E-09	2.28E-09	4.40E-10	1.60E-09	7.91E-10	T10H-5HIS	2.62E-09	5.24E-10	2.85E-09	1.65E-10	2.47E-09	4.71E-10
T10H-5HIS	2.10E-09	8.45E-10	2.32E-09	3.55E-10	1.94E-09	8.79E-10	T14L-7.8HIS	2.94E-09	1.19E-09	3.15E-09	4.14E-10	2.50E-09	7.09E-10
T16H-7.8RAS	2.25E-09	9.05E-10	2.47E-09	1.82E-10	1.98E-09	7.91E-10	T13L-5AVG-5	2.83E-09	1.16E-09	3.26E-09	3.54E-10	2.45E-09	9.37E-10
T17L-5HIA	2.90E-09	1.59E-09	2.52E-09	6.26E-10	2.21E-09	7.58E-10	T16H-7.8RAS	4.08E-09	8.22E-10	4.33E-09	2.88E-10	3.41E-09	1.16E-09
T14L-7.8HIS	2.54E-09	1.10E-09	2.69E-09	6.30E-10	2.26E-09	9.90E-10	T17L-5HIA	4.06E-09	2.17E-09	4.53E-09	6.85E-10	3.25E-09	1.51E-09

	Eluent = VZP							Eluent = DIW					
	D_{obs} 7d up to 861d Avg.	σ	D_{obs} 28d to 63d Avg.	σ	D_{obs} 28d up to 861d Avg.	σ		D_{obs} 7d up to 861d Avg.	σ	D_{obs} 28d to 63d Avg.	σ	D_{obs} 28d up to 861d Avg.	σ
	NO_2							NO_2					
T15H-7.8HIS	2.47E-09	1.11E-09	2.90E-09	4.69E-10	2.16E-09	9.98E-10	T5H-7.8AVG	3.79E-09	1.58E-09	4.78E-09	7.30E-10	3.43E-09	1.57E-09
T5H-7.8AVG	2.57E-09	1.12E-09	2.97E-09	6.21E-10	2.24E-09	9.95E-10	T15H-7.8HIS	3.78E-09	1.54E-09	4.82E-09	8.56E-10	3.51E-09	1.62E-09
T3H2-7.8AVG	3.57E-09	1.84E-09	4.03E-09	3.44E-10	2.91E-09	1.23E-09	T3H2-7.8AVG	4.80E-09	1.70E-09	5.39E-09	3.63E-10	4.30E-09	1.37E-09
T21L-7.8HIS	7.06E-09	4.81E-09	8.79E-09	1.96E-09	5.45E-09	3.69E-09	T21L-7.8HIS	8.90E-09	6.95E-09	1.16E-08	2.56E-09	6.62E-09	5.30E-09
	NO_3							NO_3					
T8L-5RAS	2.25E-09	1.05E-09	2.80E-09	5.84E-10	1.95E-09	9.39E-10	T8L-5RAS	2.86E-09	1.15E-09	3.41E-09	7.14E-10	2.52E-09	9.74E-10
T10H-5HIS	2.73E-09	1.18E-09	3.05E-09	5.10E-10	2.34E-09	9.65E-10	T18L2-7.8RAS	3.67E-09	9.36E-10	3.63E-09	8.39E-10	3.34E-09	6.03E-10
T24H-5HIA	2.85E-09	2.03E-09	3.06E-09	8.85E-10	2.07E-09	1.21E-09	T10H-5HIS	3.70E-09	6.45E-10	3.80E-09	5.03E-10	3.50E-09	5.39E-10
T13L-5AVG-5	3.49E-09	1.76E-09	3.19E-09	9.05E-10	2.75E-09	9.00E-10	T24H-5HIA	3.60E-09	1.78E-09	3.85E-09	1.02E-09	2.99E-09	1.17E-09
T18L2-7.8RAS	3.42E-09	1.63E-09	3.26E-09	1.11E-09	2.77E-09	9.11E-10	T14L-7.8HIS	4.32E-09	1.51E-09	4.27E-09	8.13E-10	3.76E-09	8.38E-10
T5H-7.8AVG	3.23E-09	1.70E-09	3.66E-09	7.03E-10	2.63E-09	1.28E-09	T13L-5AVG-5	4.13E-09	1.54E-09	4.47E-09	7.64E-10	3.59E-09	1.10E-09
T16H-7.8RAS	3.34E-09	1.60E-09	3.69E-09	8.07E-10	2.78E-09	1.22E-09	T5H-7.8AVG	4.80E-09	1.87E-09	5.64E-09	1.01E-09	4.27E-09	1.69E-09
T14L-7.8HIS	3.56E-09	1.49E-09	3.79E-09	9.83E-10	3.02E-09	1.03E-09	T16H-7.8RAS	5.37E-09	1.57E-09	6.09E-09	8.56E-10	5.05E-09	1.58E-09
T15H-7.8HIS	3.36E-09	1.83E-09	3.97E-09	8.21E-10	2.75E-09	1.48E-09	T3H2-7.8AVG	6.13E-09	1.88E-09	6.32E-09	1.29E-09	5.58E-09	1.49E-09
T17L-5HIA	4.80E-09	2.93E-09	4.02E-09	1.35E-09	3.54E-09	1.33E-09	T15H-7.8HIS	5.38E-09	2.15E-09	6.38E-09	1.78E-09	4.90E-09	2.15E-09
T3H2-7.8AVG	4.22E-09	2.31E-09	4.62E-09	7.49E-10	3.42E-09	1.56E-09	T17L-5HIA	7.34E-09	5.11E-09	7.03E-09	2.13E-09	5.35E-09	2.39E-09
T21L-7.8HIS	8.87E-09	6.24E-09	1.09E-08	3.53E-09	6.81E-09	4.85E-09	T21L-7.8HIS	1.20E-08	9.60E-09	1.49E-08	5.40E-09	8.82E-09	7.14E-09
	Cr							Cr					
T8L-5RAS	*<3.58E-15*	6.99E-15	*<2.79E-15*	1.03E-15	*2.71E-14*	2.66E-14	T14L-7.8HIS	*<5.20E-15*	5.71E-15	*<5.70E-15*	4.02E-15	*<3.39E-15*	3.40E-15
T3H2-7.8AVG	*<1.37E-15*	2.25E-15	*<2.95E-15*	3.16E-15	*<1.42E-15*	2.48E-15	T8L-5RAS	2.71E-14	2.66E-14	1.84E-14	7.98E-15	1.80E-14	1.01E-14
T21L-7.8HIS	*<1.44E-15*	2.29E-15	*<3.06E-15*	3.07E-15	*<1.55E-15*	2.51E-15	T18L2-7.8RAS	*<1.73E-14*	1.07E-14	*<2.06E-14*	1.50E-14	*<1.75E-14*	1.15E-14
T15H-7.8HIS	*<2.34E-15*	3.86E-15	*<3.45E-15*	4.28E-15	*<1.88E-15*	3.35E-15	T21L-7.8HIS	2.97E-14	1.60E-14	3.06E-14	1.43E-14	3.29E-14	1.59E-14
T5H-7.8AVG	*<1.74E-15*	3.12E-15	*<4.07E-15*	4.10E-15	*<2.16E-15*	3.39E-15	T5H-7.8AVG	4.21E-14	4.85E-14	3.36E-14	1.65E-14	4.71E-14	5.36E-14
T14L-7.8HIS	*<1.96E-15*	3.02E-15	*<4.21E-15*	3.93E-15	*<2.13E-15*	3.30E-15	T24H-5HIA	5.26E-14	2.28E-14	5.58E-14	1.14E-14	4.66E-14	1.98E-14
T17L-5HIA	*<3.51E-15*	5.75E-15	*<6.54E-15*	8.26E-15	*<3.54E-15*	6.45E-15	T17L-5HIA	6.79E-14	3.09E-14	6.20E-14	2.47E-14	6.90E-14	3.47E-14

	Eluent = VZP							Eluent = DIW					
	D_{obs} 7d up to 861d Avg.	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d up to 861d Avg.**	**σ**		**D_{obs} 7d up to 861d Avg.**	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d up to 861d Avg.**	**σ**
	Cr							**Cr**					
T24H-5HIA	*<4.57E-15*	6.69E-15	*<6.66E-15*	7.40E-15	*<3.39E-15*	5.85E-15	T3H2-7.8AVG	2.74E-13	6.82E-13	7.98E-14	2.62E-14	3.15E-13	7.56E-13
T13L-5AVG-5	*<1.07E-14*	1.75E-14	*<7.75E-15*	8.14E-15	*<4.17E-15*	6.70E-15	T16H-7.8RAS	1.16E-13	7.39E-14	1.07E-13	3.06E-14	1.06E-13	7.79E-14
T10H-5HIS	*<3.56E-14*	6.94E-14	*<1.09E-14*	1.36E-14	*<5.63E-15*	1.08E-14	T10H-5HIS	2.18E-13	2.61E-13	1.32E-13	1.02E-14	1.07E-13	4.40E-14
T16H-7.8RAS	*<6.69E-15*	1.00E-14	*<1.38E-14*	1.29E-14	*<7.43E-15*	1.11E-14	T15H-7.8HIS	1.51E-13	1.17E-13	1.62E-13	4.79E-14	1.07E-13	7.08E-14
T18L2-7.8RAS	*<7.02E-15*	1.05E-14	*<1.48E-14*	1.38E-14	*<7.65E-15*	1.15E-14	T13L-5AVG-5	1.34E-12	4.17E-12	3.31E-12	6.59E-12	1.67E-12	4.66E-12

Italicized numbers = interval D_{obs} values included eluates where Cr concentrations were less than detection values. Note that the monoliths shaded gray in Table 4.3 were leached through 590 days and then stored for solid phase characterization. Values in red type suggest that the D_{obs} values for the cumulative leach intervals 590 and 861 are showing increases compared to the averaged interval D_{obs} value from 7 or 28 days through 427 days. **Note that none of the interval averaged D_{obs} values have been corrected for inventory depletion.**

Cr leach data in DIW shows one Cast Stone mix (T14L-7.8HIS) with much better performance than all the others. At this moment, we have no explanation for this observation. Table 4.4 shows that the best and worst performing Cast Stone monoliths when leached in VZP have moderate correlation with the best and worst performing monoliths when leached in DIW for each constituent of interest. For ^{99}Tc, Cast Stone Mixes 24, 15, and 14 are the better-performing mixes and Mixes 21, 17, and 16 are among the worst performing regardless which eluent is used. For iodide, regardless of eluent used, Cast Stone Mixes 10 and 24 are the best performing and Mixes 21 and 3 are the worst performing. For Na, Cast Stone Mixes 13, 14, and 18 leach less than other mixes and Mixes 21 and 3 leach the most regardless of eluent used. For nitrate and nitrite, Cast Stone Mixes 18 and 8 leach less and Mixes 21 and 3 leach the most regardless of eluent used. As found for the Screening Leach Tests –extended results in the previous subsection, the Archive Leach Test Suite D_{obs} values for the anions iodide, nitrate, and nitrite, and the cation sodium, are very similar for each of the Cast Stone mixes. As mentioned, the range in 28- to 63-day averaged interval D_{obs} values for these four constituents varies by only a factor of three to six across all the LAW simulants used regardless of the overall Na concentration 5 or 7.8 M and the dry blend solids to free water ratio used (0.4 or 0.6) or the source of the fly ash or blast furnace slag.

For uranium present in the Archived monoliths, when leached in VZP, almost all the eluates had no detectable uranium. Using the detection limit for uranium in the eluates and the starting inventory of uranium, the calculated D_{obs} values range from $<1 \times 10^{-16}$ to $<3 \times 10^{-16}$ cm^2/s. For two Mixes (8 and 16) leached in DIW, the eluate uranium concentrations were detectable yielding D_{obs} values of 1×10^{-15} and 4×10^{-16} cm^2/s, respectively.

Uranium and Cr leach the slowest, especially when the VZP eluent is used, followed by ^{99}Tc, and then the four more mobile constituents. The reason that Cr leaches the so slowly suggests that it takes longer for the blast furnace induced reduced Cr to re-oxidize than it takes reduced Tc to re-oxidize. Then we would expect both chromate and pertechnetate to diffuse out of the monoliths interior at about the same rate, given their similar size. As mentioned, uranium leaches very slowly because it likely is sequestered as very insoluble calcium uranate.

For most of the Archive Leach Test Suite monoliths leached in VZP, the mobile constituents (I, NO_3, NO_2, and Na) do not release much more than 20% of their starting inventories until 49- to 63-days of leaching has occurred and corrections for inventory depletion are not significant until after 93 days of leaching. Thus, for future IDF PA predictive modeling, the highlighted values for the 28- to 63-day averaged interval D_{obs} values for the VZP eluent are recommended for time extrapolation for intact waste forms, if an empirical diffusivity conceptual model is chosen for the release from LAW Cast Stone. The D_{obs} values for the COCs from earlier leaching periods (0.08 to 14 days) and the D_{obs} values for tests that used DIW as the eluent could be used to create a range of D_{obs} values that could be used in sensitivity analyses. The early leaching data for 0.08 to 7 days for all Cast Stone and most cementitious waste forms described in literature in general always exhibit high interval D_{obs} values; often an order of magnitude and in a few cases up to two orders of magnitude higher than interval D_{obs} values calculated after 28-days and longer leaching times. We don't generally recommend using the 0.08 to 7 day data because when leach testing small monoliths the "surface wash off" and release from "surface pores" that directly intercept the monolith's outer surface dominate the early release data. The wash off effect is believed to be caused by salts that evaporate on the monolith surface during curing. These early interval D_{obs} values are considered to be biased high for COCs, especially mobile ones such as iodide, NO_3, NO_2, and Na. In the actual IDF disposal setting the cementitious waste forms will be much larger (have a much smaller SA/V ratio) so the wash off and "surface pores" will have a much lower impact on the very early release of COCs. Regardless, if there is a need to select maximum (worst case) D_{obs} values to obtain a range to perform sensitivity analyses, the early leaching data and D_{obs} values obtained in DIW found in Appendix B could be used. However, all the data in this report is for LAW waste simulants solidified in one Cast Stone dry blend mix and are not necessarily relevant to the solidification of liquid secondary waste simulants in a different dry blend mix, which is work underway (see Um et al. 2016) for the first report on secondary liquid wastes solidified in Cast stone and a hydrated lime-based dry blend mix. It is the solidified secondary waste simulants that will be addressed in the 2017 IDF PA. Detailed results for individual interval D_{obs} values, cumulative fractions leached, eluate pH and EC values for the Archive Leach Test Suite tests are found in tables within Appendix B.2.

We recommend using the 28- to 63-day averaged interval D_{obs} values from using VZP eluent as the most probable values to use in future solidified LAW waste forms because they 1) better represent water that will percolate through the IDF facility; and 2) when the cumulative mass of a constituent released from the monoliths starts to exceed 20% of their starting inventories for mobile constituents (around day 93 of leaching in our data), the mass used to calculate the observed diffusion coefficient become less accurate. The accuracy issue arises because the leaching conditions no longer satisfy the assumed boundary conditions. Further, the D_{obs} values for the early leach periods (0.08 days up to 14 days) are biased high likely because of monolith surface salt wash-off and diffusion from pores directly connected to the relatively small monoliths' outer surface. That is, the leach results from the early times are also not good indications of diffusion from full-sized Cast Stone monoliths destined for burial in IDF. Thus, we chose the 28- to 63-day averaged interval D_{obs} values as most representative of IDF burial conditions.

One possible reason for the observed lower interval D_{obs} values for Cast Stone monoliths from the Archive Leach Test Suite that were leached in VZP compared to monoliths leached in DIW is the fact that significant quantities of secondary minerals precipitated on the surfaces of the VZP-leached monoliths (see Figure 4.3). We cannot confirm that there is enough surface precipitate on these monoliths leached in VZP to state objectively that the lower interval D_{obs} values consistently calculated for all constituents of interest from these monoliths in comparison to monoliths leached in DIW is caused by the physical "armoring" of the monoliths by the white precipitate. However, literature exists that shows performing leach tests on cement paste discs in nearly CO_2 –free ($pCO_2 = -5.5$ atm) conditions versus normal CO_2 conditions ($pCO_2 = -3.5$ atm) yield very different results. Chloride and iodide D_{obs} values drop significantly in the normal carbonated condition compared to the disks (1-cm thick) subjected to a carbon dioxide free condition (Sarott et al. 1992). Figure 4.4 (from Sarott et al. 1992) shows the cumulative activity of radioactive ^{125}I that diffused through a cement paste disc placed between two reservoirs of simulated cement paste pore water (one containing the tracer and one free of tracer). The carbonate-free reservoirs and through-diffusion test were performed in an anoxic glove box with no CO_2 present while the carbonate containing reservoirs and through-diffusion tests were open to the atmosphere. Figure 4.4 shows the cumulative activity of ^{125}I that passed through the cement paste disc as a function of time. The higher activity in the carbonate-free condition equates to a larger D_{obs} value compared to the similar test in the presence of carbon dioxide/carbonate. In our Cast Stone monolith leach tests, those performed in VZP eluent contained larger amounts of bicarbonate/carbonate than the tests performed in DIW; the measured D_{obs} values follow the same trend as shown in Figure 4.4. Further, monolith weight measurements taken after each sampling and before placing the monoliths in the next batch of fresh eluent show that over time, a consistent mass increase for those monoliths leached in VZP and a consistent mass decrease in monoliths leached in DIW. The former net mass increase is likely caused by the white precipitate observed on the VZP-contacted monoliths, while the loss of mass in the DIW-contacted monoliths suggests a net dissolution of the monoliths contacted with DIW. Figure 4.15 in the Tc-Gluconate section shows an example of the monolith mass changes with time for one set of monoliths.

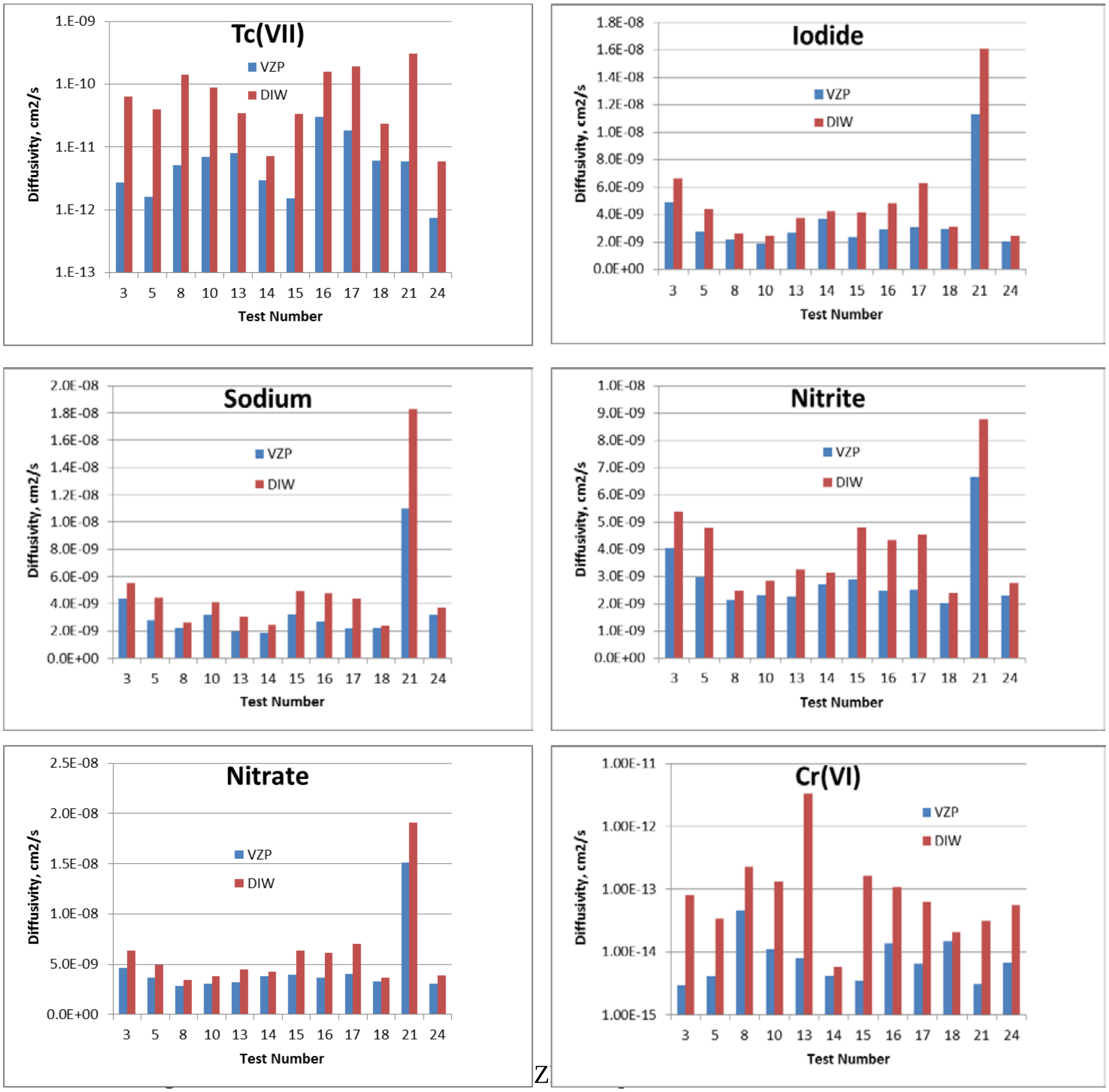
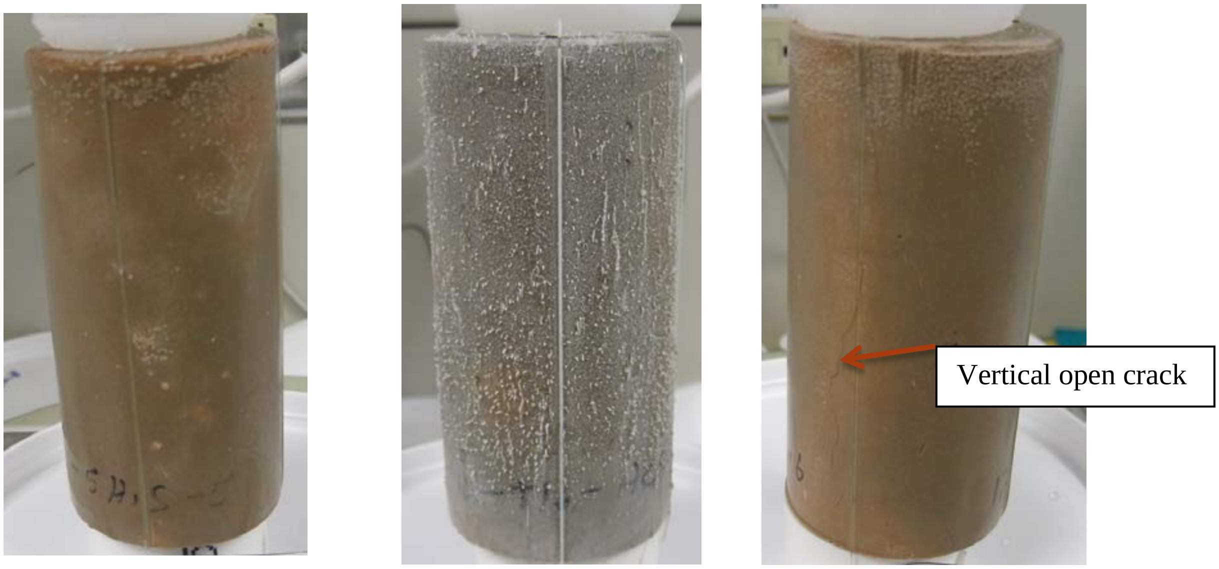


Figure 4.2. Archive Interval D_{obs} Values (cm^2/s) for 28-day to 63-day Samplings in DIW vs VZP Eluent

CS-T5HCS1-3 in VZP

CS-T5HCS1-6 in DIW
(note vertical crack)

CS-T10HCS1-3 in VZP



CS-T10HCS1-5 in DIW

CS-T16HCS1-4 in VZP

CS-T16HCS1-6 in DIW
(note vertical crack)

Figure 4.3. Comparison of Archived Monoliths for Three Mixes Leached in VZP vs. DIW

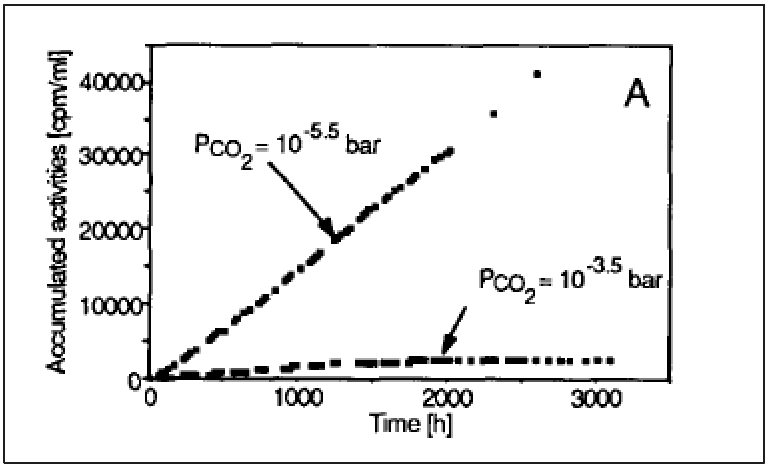


Figure 4.4. Cumulative Fraction of ^{125}I Passing through Cement Paste Disk under Carbonate-Free vs Carbonate Containing Conditions (Figure 3A in Sarott et al. (1992))

Another parameter that is consistently different for the VZP versus DIW eluates is pH. The VZP eluates' pH values are consistently lower than the DIW eluates' pH values. An example is shown in Figure 4.5 for Mix 5 monoliths from the Archive Suite of leach tests. There is an increase in pH between the 93- and 380- day samplings for both eluates because of the long interval between samplings. Essentially, over this 287-day interval, more material diffused out of the monoliths from both eluates. Because the simulants themselves and the Cast Stone internal pore water are inherently caustic, as the time between changing out the eluent increased the eluates' pH increases. Increased pH in the eluates surrounding the monoliths may cause more dissolution of the monolith, which leads to more COC release. At this time, we do not know if this pH difference could also be causing the lower leach rates when monoliths are immersed in VZP. All the other Archive Leach Test Suite monoliths' eluates show a similar pH pattern as Mix 5, (see Appendix B.1 Table B.13 for eluate pH values for all the Extended Suite Eluates, Appendix B.2 Table B.27 for Archive Suite eluate pH values, Appendix B.3 Table B.37 for Iodide Loading Suite eluate pH values and Appendix B.4 Table B.49 for Tc-Gluconate Suite eluate pH values).

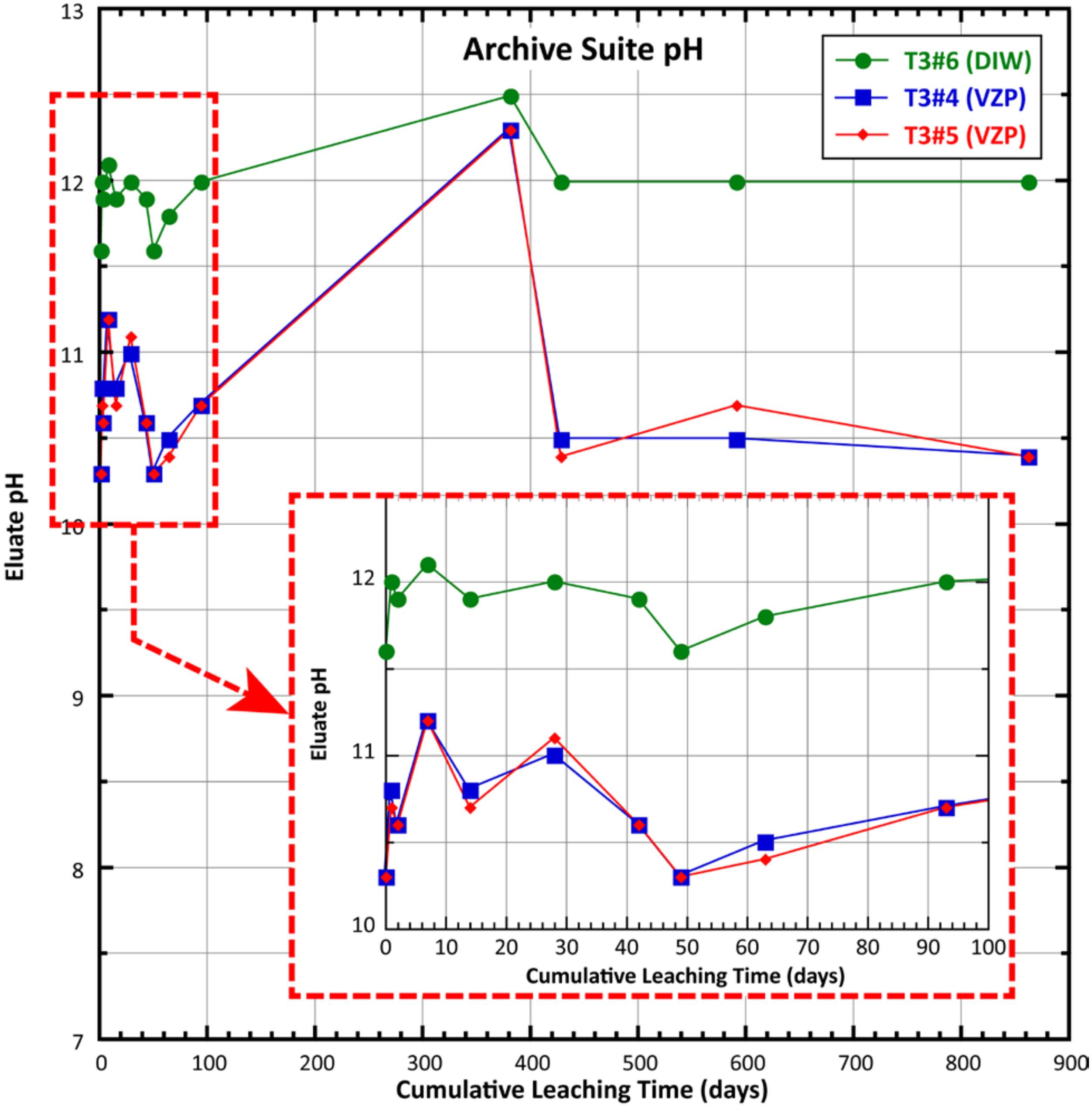


Figure 4.5. Typical pH Trend for Archive Suite Eluates vs. Leach Time

The white precipitate on the VZP monolith surfaces has been identified by XRD as predominantly aragonite (a polymorph of calcium carbonate) with some brucite ($Mg(OH)_2$) and perhaps some calcite (another calcium carbonate polymorph), although not all calcite peaks are present in any of the precipitates that were characterized. The fact that aragonite appears to form rather than the more stable calcite has recently been discussed (see Sun et al. 2015) as being caused by magnesium being present in oversaturated solutions in which the carbonate minerals are nucleating/precipitating. There is adequate Mg in the starting VZP pore water simulant and most of the Cast Stone monolith eluates after contacting monoliths with the VZP. Thus, this recent journal article and references within support our finding of aragonite as the dominant carbonate mineral in the white precipitates on the surfaces of the monoliths. To date, we have not collected adequate masses of the white precipitate off monoliths leached in DIW to get useful XRD patterns, but we feel confident that the white precipitate is likely some polymorph of calcium carbonate, and brucite.

Of particular interest is the vertical cracks found on the surfaces of some of the Archive Leach Test Suite monoliths leached in DIW (see Figure 4.3) and also significant “rubbelization” of monoliths from two mixes leached in VZP (Figure 4.6). The surface micro-cracks in monoliths leached in DIW are not covered by white precipitate, suggesting that they are newly formed. We wonder if this observation is the first sign of some internal degradation of Cast Stone monoliths between 427 and 590 to 861 days of leaching. At least two of the Archive Leach Test Suite monoliths leached in VZP show significant cracking and pieces close to spalling off at their bottoms as shown in Figure 4.6. Interestingly, these two LAW mixes have been the worst performing (T21 mix) and among the worst performing (T17 mix) for both the Extended Suite and Archive Suite since the beginning of leach testing. The significant “rubbelization” of these two mixes appeared between 427 and 590 days of leaching; however the high leaching of mobile constituents occurred from the very beginning of leach testing. Thus the observed “rubbelization” might not be the direct cause of the high leaching for these two mixes. Figure 4.7 shows a plot of interval D_{obs} values for monoliths (including the two mixes showing the “rubbelization”) from four mixes that in Revision 0 of this report hinted at an uptick in D_{obs} between the 380- and 427-day sampling events. With two more samplings (at 590 and for some 861 cumulative days) it now appears that the uptake in interval D_{obs} is muted for most of the mobile constituents (e.g., Na and NO_3) with the T17 monoliths (showing “rubbelization”) perhaps maintaining a slight uptick. Of interest is the occurrence of upticks in long-term interval D_{obs} values for nitrite from several monolith mixes (e.g., T10, T17, T21, and T24) leached in VZP between 380 and 590 days of leaching. Many of the monoliths leached in VZP show black spots developing on their outer surfaces that might be indications of microbiological growth. For one batch of VZP simulant that sat in two large barrels for up to nine months during the time the Extended Leach task was unfunded the chemical composition of the VZP eluent changed with a significant loss of nitrate and a concomitant increase in nitrite. A plausible explanation for the conversion of nitrate to nitrite is biological growth of specific microbes that use nitrate as an oxygen source. We speculate that the black spots on some of the Archive Suite monoliths continually leached in VZP monoliths for up to 861 days may be converting a few tens of mg/L of the nitrate that is leaching out of the monoliths to nitrite. The nitrate concentration in VZP eluates generated in the 590 and 861 day leachates range from 380 to 1520 mg/L and the nitrite eluates range from 60 to 180 mg/L. Thus the biological conversion of a few tens of ppm nitrate to nitrite would not be readily noticed as something unexpected. The black spots on Archived Suite monoliths leached in VZP are being further investigated in the solid phase characterization activities underway, which will be documented in a PNNL technical report by the end of FY2016.

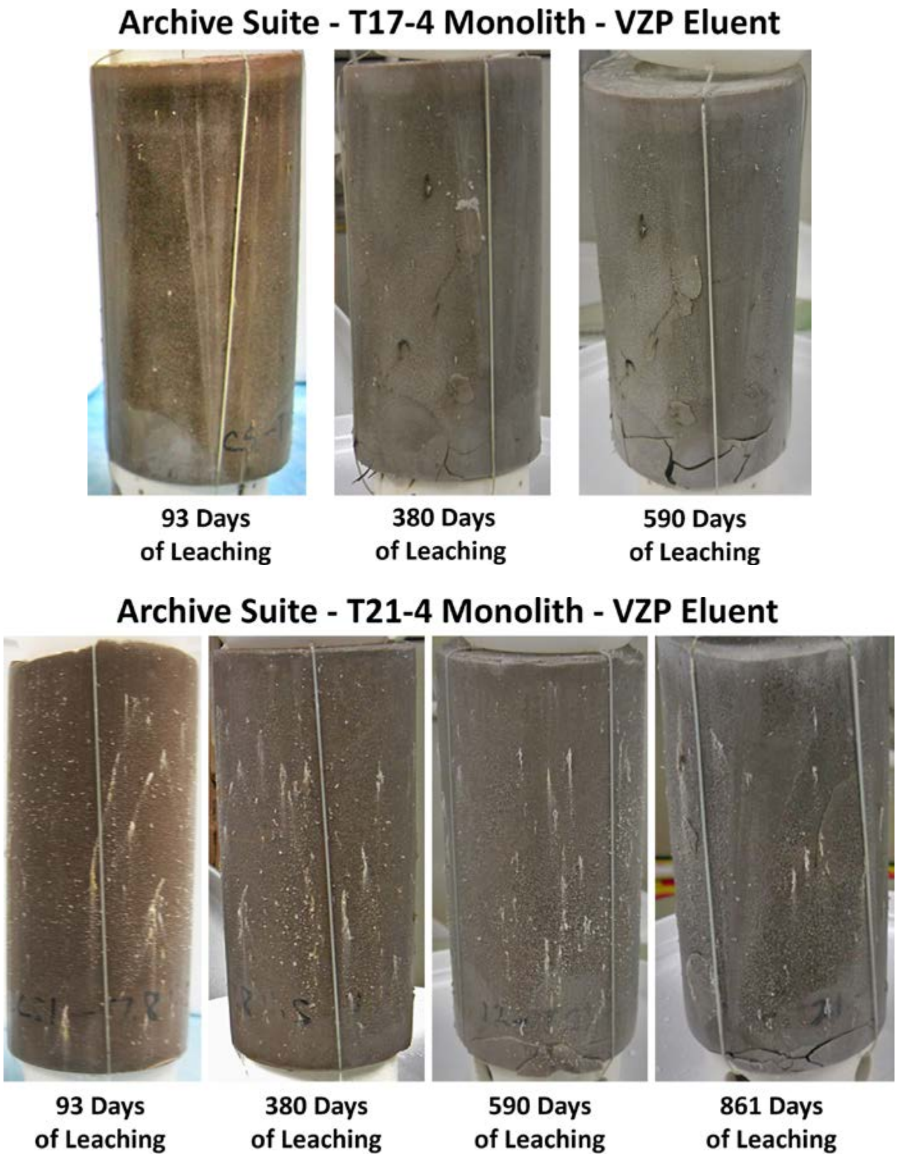
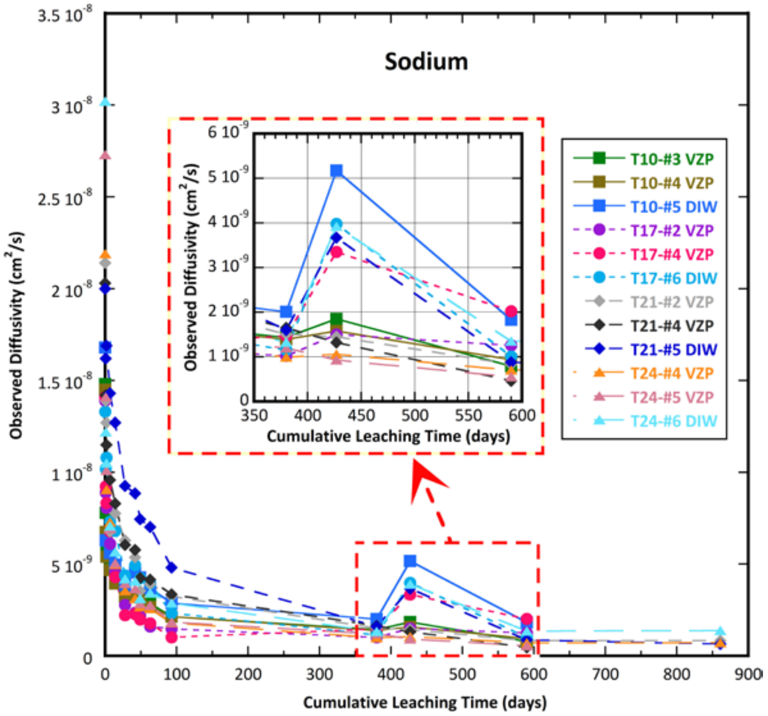
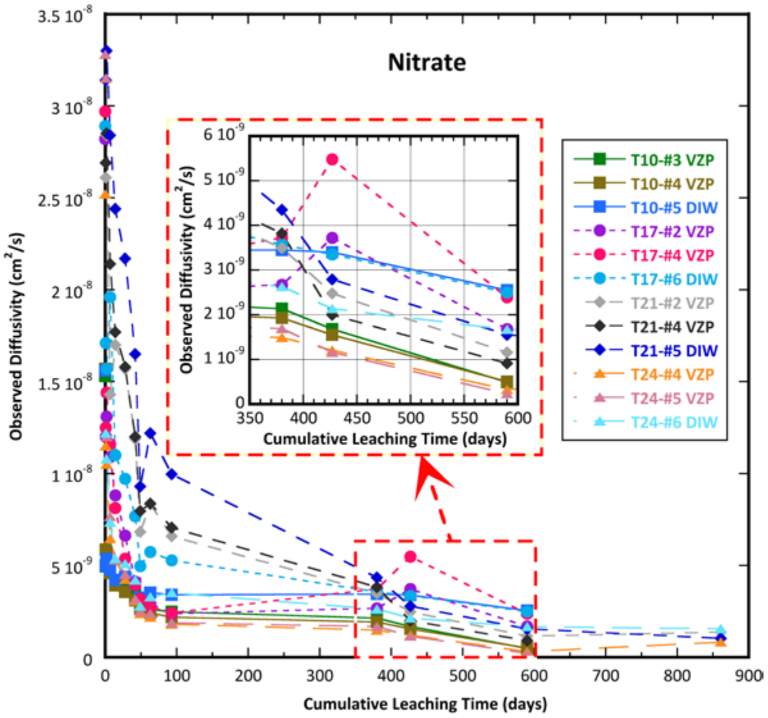


Figure 4.6. Two Archive Monoliths Leached in VZP Showing Rubbelization

Observing surface cracks and the" rubbelization" in monoliths submerged in water could lead to increased leaching of constituents from within the monoliths. However, in the IDF burial environment once the metal container is compromised and recharge water has the opportunity to contact Cast Stone solid waste forms, the presence of surface cracks may not lead to increased leaching because under unsaturated moisture conditions cracks are often not filled with water. The 2003 risk assessment for supplemental waste forms (Mann et al. 2003) remark that cracks in the Cast Stone blocks are not expected to fill with water in a vadose zone environment. However, cracks in Cast Stone can become a short-circuit pathway for gaseous oxygen diffusion into the interior of the Cast Stone. Thus Cast Stone cracking during weathering merits consideration in the upcoming 2017 IDF PA activities. Yabusaki et al. (2015) discuss cracking in more detail including both conceptual and numerical methods for performing predictive modeling of the impacts of cracked cementitious waste on contaminant release. Also the recent Solid Secondary Waste Form data package (see Flach et al. 2016) has some useful discussion on cementitious waste form degradation mechanisms that result in cracks and under what moisture saturation conditions might the cracks fill with water versus under what moisture conditions that cracks might be dry and available for oxygen entry.





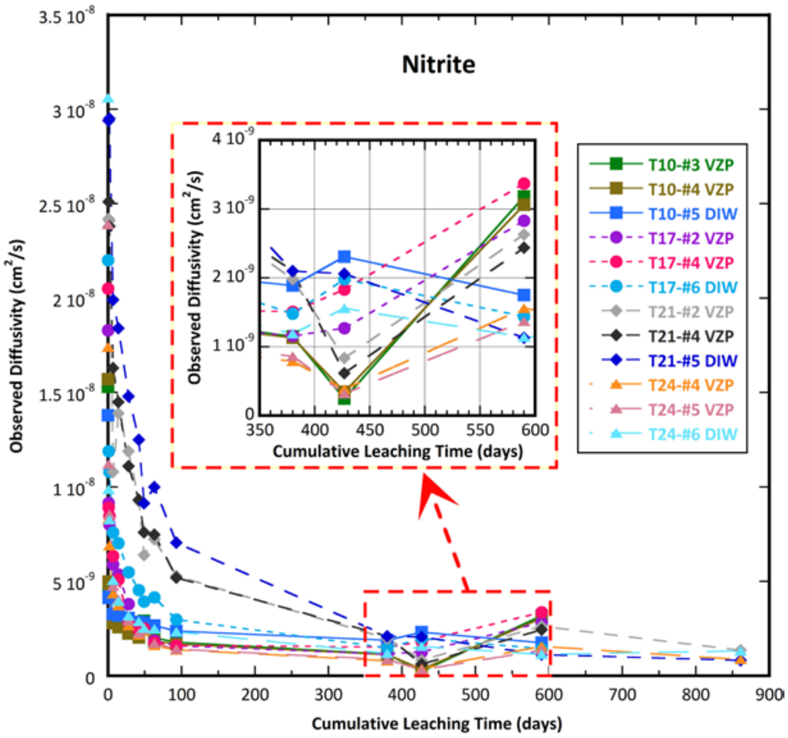


Figure 4.7. Uptick in D_{obs} between 380 and 590 Days for Archived Suite Monoliths

4.3 I-Loading Suite Results

The objective for testing the Iodide Suite of monoliths was to evaluate the impacts of varying the iodide loading (starting iodide concentrations in one LAW simulant, the 7.8 M Na average simulant) by manufacturing additional Cast Stone monoliths and repeating the EPA-1315 leach tests using DIW and the IDF VZP as eluents. The mass of stable iodide, ^{127}I, added to the original Screening Test Cast Stone monoliths was ~245 times larger than the average projected ^{129}I concentration (when converted from activity to mass) and ~45 times larger than the maximum projected ^{129}I mass concentration in LAW wastes (see Westsik et al. 2013a; pg 34). This large excess beyond the average and maximum projected ^{129}I concentrations in LAW waste streams was the result of wanting to be certain that there would be enough iodide in eluates to facilitate calculation of interval D_{obs} values.

Literature exists (see Pierce et al. 2004, Section 6 and Serne and Westsik 2011 for more detailed discussion and references) that suggests that diffusivity of iodine species leaching out of cement/grout waste forms depends on both the total concentration and speciation of iodine present in the waste form. However, most of the cement/grout studies in the cited literature reviews that had iodine loaded into the waste forms used high starting iodine concentrations, generally above 1 to 5 wt%. One study performed at Hanford, Lockrem (2005), did load Cast Stone, made with a liquid waste simulant similar to Hanford secondary wastes that are processed through the ETF, with less extreme concentrations of iodide concentrations. Figure 4.8 shows that the iodide leach index (see Eq. 3.4 for definition) in the Lockrem (2005) study does in fact decrease (i.e., D_{obs} increase by a factor of 10 between loadings of 0.14 to 1.4 wt% iodide.

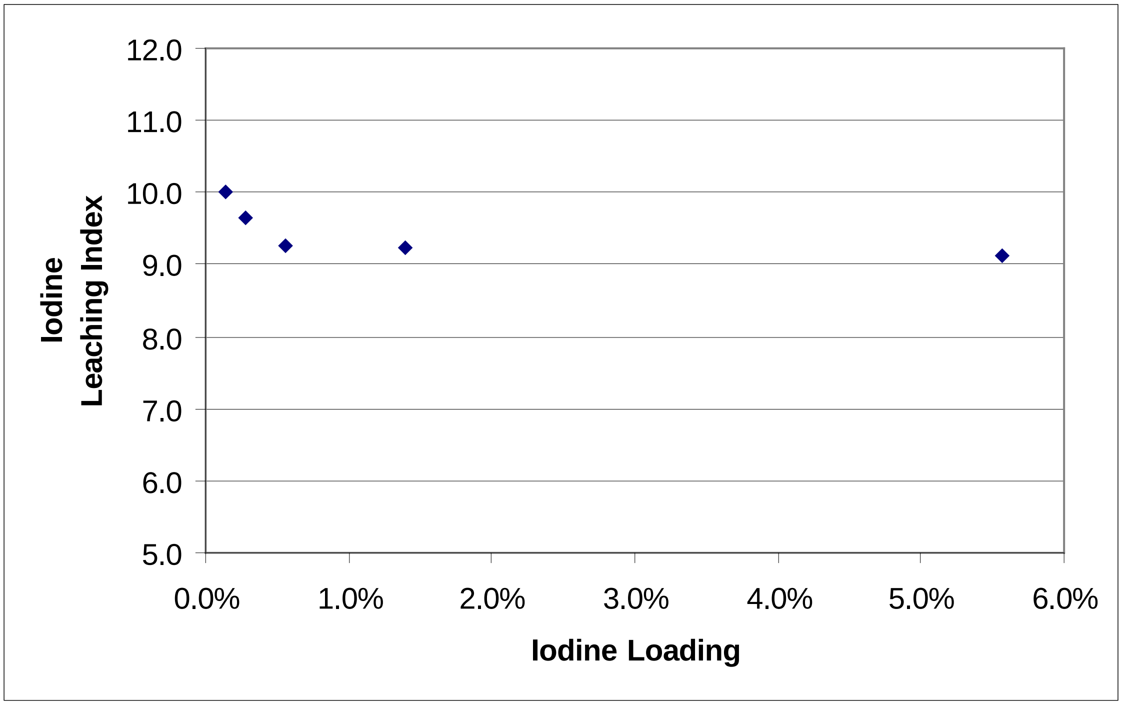


Figure 4.8. Leach Index of Iodide vs. Iodide Loading in Cast Stone (data from Lockrem 2005)

Given the observed iodide leach sensitivity to loading and the Screening Test results that showed relatively high iodide release rates (see Westsik et al. 2013a), we elected to perform the I-Loading Suite of tests with lower starting inventories of iodide in hopes of finding improved performance (lower D_{obs} values or higher LI values).

Table 4.5 lists the Iodide Suite of monoliths sample IDs, the iodide loading values, and the eluent used in the EPA Method 1315 leach tests. Recall that these Cast Stone monoliths were made with the 7.8 M Na Average simulant spiked with small quantities of reagent grade NaI. The iodide-spiked simulant was then solidified with the standard Cast Stone dry blend at a weight ratio of 0.6 free-water-to-dry blend. The Iodide Suite of monoliths is the same as the duplicated original Screening Test Mix 3 and 22, with the exception of the spiking levels of NaI. One key point is that the iodide loading for all the monoliths shown in Table 4.5 are at least a factor of 10 lower than the lowest iodide loading used by Lockrem (2005), and more than 100 times below 1 wt% loading.

The leach results for the I-Loading Suite of monoliths, along with comparable results from the Screening Test Mix #3 and #22 and the Archive Suite #3 (all representing tests on the 7.8M Na Average simulant mixed with the Cast Stone dry blend at a free-water-to-dry-blend ratio of 0.6), follows.
Table 4.6 provides the average interval D_{obs} values for the same three sampling time intervals as used previously, when available. Data from Table 4.6 for the iodide average interval D_{obs} values for the sampling period from 28- through 63- days are plotted as a bar chart in Figure 4.9. Combined, the table and figure suggest that there is little difference in the leach rates for the Iodide Suite of monoliths with low, medium, and high loadings, especially when leached in DIW. The original Screening Test monoliths (Mix 3 and Mix 22) leached in DIW show the largest difference in iodide averaged interval D_{obs} values, despite being replicates. When bar charts similar to Figure 4.9 are prepared for the other constituent averaged interval D_{obs} values, the same trend (that Mix 3 shows the largest averaged interval D_{obs} value and Mix 22 shows a lower averaged interval D_{obs} value) is found (see Figure 4.10). That is, the loading of all other constituents (nitrate, nitrite, and sodium) in the Iodide Suite and Mixes 3 and 22 are identical.

Thus, all the averaged interval D_{obs} values for a given constituent among this group of three should be identical for all five monolith types. The fact that there is the same trend in differences in averaged values for iodide, sodium, nitrate, and nitrite suggests that there are inherent differences in the monoliths themselves due to various physical properties. Possible differences in physical properties in the cured monoliths include slight differences in surface micro-cracks after curing and de-molding, and perhaps differential settling of the wet slurries during curing of each mix batch, leading to different cured porosities and tortuosity. On the other hand, the observed differences in leach properties may be caused by subtle differences in the processing of the wet slurry in each mix.

Table 4.5. Details on Iodide Suite of Monoliths

Sample ID	Iodide Spiking Level	Iodide Loading (mg/kg-dry)	Iodide Loading wt%	Eluent
CS-T1-VZ-1	Low	0.16	1.6×10^{-5}	VZP
CS-T1-VZ-2	Low	0.16	1.6×10^{-5}	VZP
CS-T1-DI-3	Low	0.16	1.6×10^{-5}	DIW
CS-T1-DI-4	Low	0.16	1.6×10^{-5}	DIW
CS-T2-VZ-1	Medium	1.6	1.6×10^{-4}	VZP
CS-T2-VZ-2	Medium	1.6	1.6×10^{-4}	VZP
CS-T2-DI-3	Medium	1.6	1.6×10^{-4}	DIW
CS-T2-DI-4	Medium	1.6	1.6×10^{-4}	DIW
CS-T3-VZ-1	High	3.6	3.6×10^{-4}	VZP
CS-T3-VZ-2	High	3.6	3.6×10^{-4}	VZP
CS-T3-DI-3	High	3.6	3.6×10^{-4}	DIW
CS-T3-DI-4	High	3.6	3.6×10^{-4}	DIW
Mix #3 & #22	Screening Test	77	7.7×10^{-3}	DIW

All Iodide Suite Monoliths leach tests were stopped after 569 days of leaching.

Table 4.6. Averaged Interval D_{obs} Values (cm2/s) for Iodide Suite of Monoliths and Similar Screening Test Monoliths

	Eluent = VZP							**Eluent = DIW**					
	D_{obs} 7d to 569d Avg.	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d to 569d Avg.**	**σ**		**D_{obs} 7d to 569d Avg.**	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d to 569d Avg.**	**σ**
	I							**I**					
T3HCS2-7.8AVG	4.69E-09	2.50E-09	4.90E-09	4.66E-10	3.76E-09	1.37E-09	T3HCS2-7.8AVG	6.42E-09	2.10E-09	6.63E-09	1.03E-09	5.76E-09	1.50E-09
Low I	8.46E-09	9.56E-09	4.59E-09	1.04E-09	8.29E-09	1.06E-08	Low I	6.01E-09	2.29E-09	4.69E-09	4.58E-10	5.16E-09	7.27E-10
Med I	4.18E-09	2.68E-09	3.26E-09	1.91E-10	3.11E-09	4.06E-10	Med I	5.85E-09	2.78E-09	4.66E-09	2.57E-10	4.77E-09	5.03E-10
High I	4.31E-09	2.86E-09	3.41E-09	2.98E-10	3.08E-09	5.66E-10	High I	5.79E-09	3.07E-09	5.09E-09	7.78E-10	4.94E-09	6.17E-10
T22	No Mix #22 monoliths were leached in VZP						T22HCS2-7.8AVG	too short		4.42E-09	8.32E-10	too short	
	Na							**Na**					
T3HCS2-7.8AVG	3.75E-09	2.14E-09	4.37E-09	4.96E-10	3.00E-09	1.47E-09	T3HCS2-7.8AVG	5.16E-09	2.37E-09	5.52E-09	5.89E-10	4.50E-09	2.04E-09
Low I	3.01E-09	2.59E-09	2.80E-09	9.78E-10	1.97E-09	1.14E-09	Low I	4.14E-09	2.31E-09	4.12E-09	5.06E-10	3.25E-09	1.24E-09
Med I	3.28E-09	2.82E-09	2.94E-09	9.16E-10	2.08E-09	1.14E-09	Med I	4.25E-09	2.81E-09	3.83E-09	6.29E-10	3.11E-09	1.17E-09
High I	3.30E-09	3.01E-09	2.83E-09	1.01E-09	2.04E-09	1.15E-09	High I	3.92E-09	2.57E-09	4.07E-09	8.95E-10	3.16E-09	1.31E-09
T22	No Mix #22 monoliths were leached in VZP						T22HCS2-7.8AVG	too short		4.84E-09	6.41E-10	too short	
	NO_2							**NO_2**					
T3HCS2-7.8AVG	3.57E-09	1.84E-09	4.03E-09	3.44E-10	2.91E-09	1.23E-09	T3HCS2-7.8AVG	4.80E-09	1.70E-09	5.39E-09	3.63E-10	4.30E-09	1.37E-09
Low I	1.94E-09	1.41E-09	1.82E-09	2.38E-10	1.37E-09	5.64E-10	Low I	2.66E-09	1.27E-09	2.48E-09	2.48E-10	2.14E-09	5.17E-10
Med I	1.92E-09	1.32E-09	1.76E-09	1.26E-10	1.38E-09	4.95E-10	Med I	2.61E-09	1.28E-09	2.32E-09	1.08E-10	2.07E-09	3.44E-10
High I	1.88E-09	1.34E-09	1.66E-09	1.47E-10	1.31E-09	4.70E-10	High I	2.43E-09	1.32E-09	2.36E-09	3.76E-10	2.05E-09	5.05E-10
T22	No Mix #22 monoliths were leached in VZP						T22HCS2-7.8AVG	too short		4.14E-09	4.75E-10	too short	
	NO_3							**NO_3**					
T3HCS2-7.8AVG	4.22E-09	2.31E-09	4.62E-09	7.49E-10	3.42E-09	1.56E-09	T3HCS2-7.8AVG	6.13E-09	1.88E-09	6.32E-09	1.29E-09	5.58E-09	1.49E-09
Low I	2.96E-09	2.32E-09	2.63E-09	4.84E-10	2.03E-09	8.41E-10	Low I	4.21E-09	1.90E-09	3.77E-09	4.10E-10	3.44E-09	6.54E-10
Med I	2.90E-09	2.06E-09	2.40E-09	2.58E-10	2.08E-09	7.48E-10	Med I	4.24E-09	1.94E-09	3.49E-09	1.22E-10	3.45E-09	4.38E-10
High I	2.77E-09	2.09E-09	2.27E-09	2.55E-10	1.90E-09	6.00E-10	High I	3.86E-09	2.04E-09	3.55E-09	5.54E-10	3.29E-09	6.30E-10
T22	No Mix #22 monoliths were leached in VZP						T22HCS2-7.8AVG	too short		4.15E-09	7.36E-10	too short	
	Cr							**Cr**					
	Cr was not measured in Iodide Suite eluates							Cr was not measured in Iodide Suite eluates					

Note that none of the interval averaged D_{obs} values have been corrected for inventory depletion.

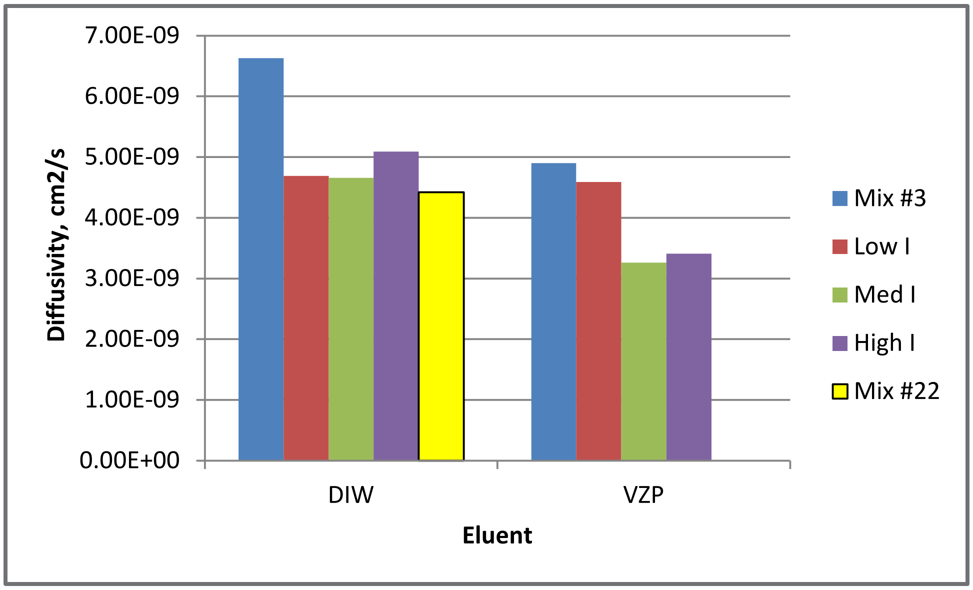


Figure 4.9. Comparison of the Iodide Averaged Interval D_{obs} Values (28 to 63-D) for Cast Stone Loaded with Varying Iodide Concentrations and Eluent Type

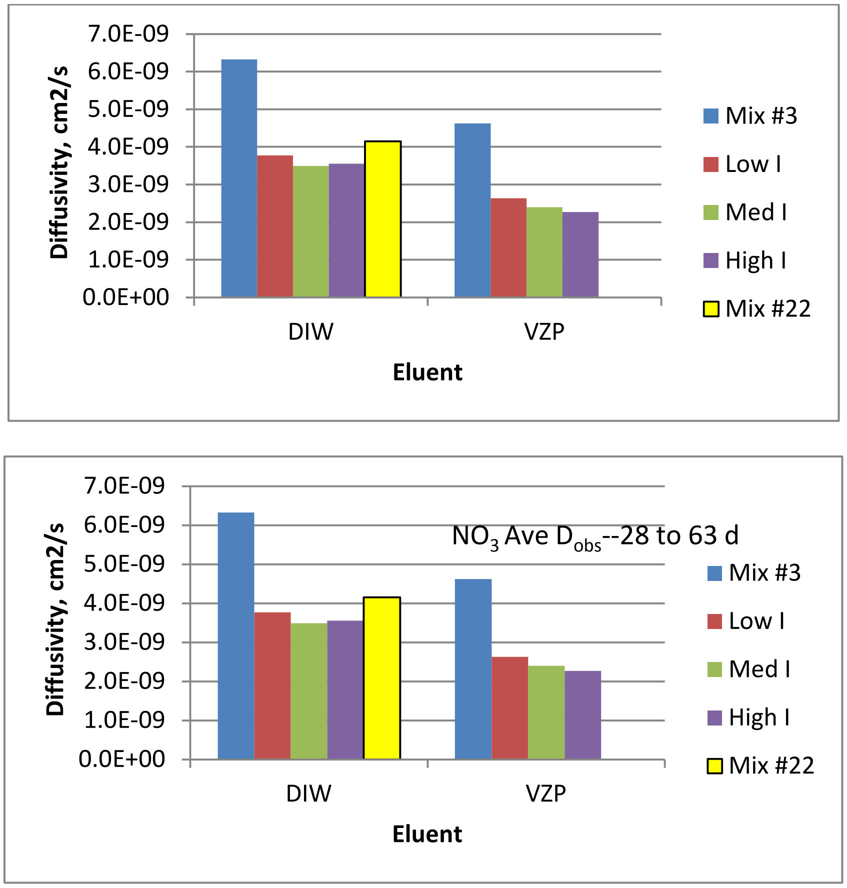


Figure 4.10. Averaged Interval D_{obs} Values for Na and NO_3 from Iodide Suite and Mix 3and Mix 22 Monoliths

Further, the standard deviations shown in Table 4.6 are large enough to preclude showing statistical differences in the averaged interval D_{obs} values for different iodide loadings (range from 1.6×10^{-5} to 7.7×10^{-3} wt%). The same can be said for the standard deviations of the averaged interval D_{obs} values for other constituents that leach from the Iodide Suite monoliths.

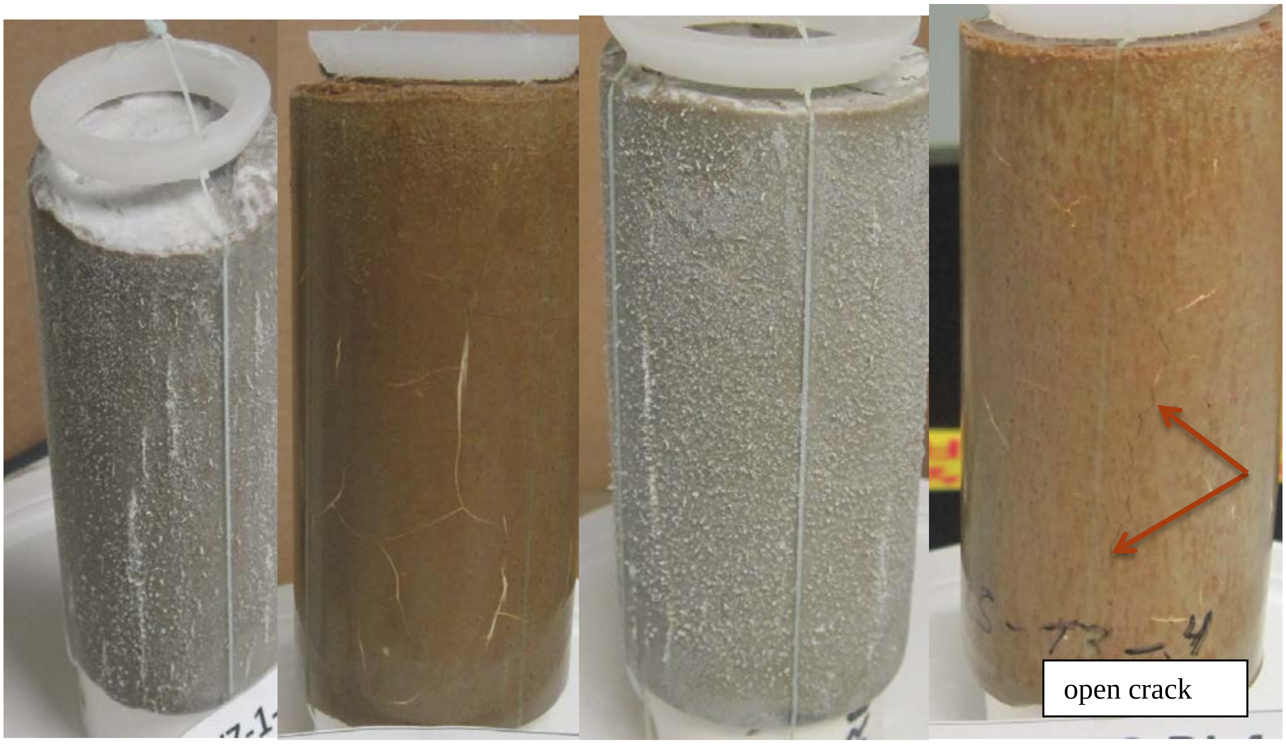
Thus, we conclude that at iodide loading well below 0.14% (the lowest loading used by Lockrem (2005)—as would be expected for all future Hanford waste streams based on HTWOS predictions—it should not matter what iodide loading is used in future simulants that are solidified into Cast Stone or grouts as long as the iodide loading is **below ~0.01 wt%**. The earlier literature suggesting that iodide loading does impact cement/grout leach tendencies was performed at total iodine loadings much higher than will ever occur at Hanford using actual liquid waste streams. Although we have not spiked Cast Stone with other possible forms of iodine (e.g., iodate), we do not expect loading impacts for other iodine species as long as low total iodine loadings used in future Cast Stone formulations remain **below ~0.01 wt%.** Atkins and Glasser (1990) state that iodide should be the iodine species that dominates in hardened cement pastes and blast furnace slag-cement blends. Only in highly oxidized cement/grout mixes could iodate be present and they hypothesize that this is a very unlikely possibility.

Bonhoure et al. (2002) also suggest that iodine is expected to exist predominantly as iodide (I^-) in cement systems except under oxic conditions, where iodate (IO_3^-) may be the dominant species. They cite several studies that indicated the potential of individual cement minerals to immobilize I^- and IO_3^- by sorption processes. Atkins and Glasser (1990) found that AFm (monosulfate: $3CaO \cdot Al_2O_3 \cdot CaSO_4 \cdot 12H_2O$) exhibited the highest potential for I^- sorption. Substitution of SO_4^{2-} by I^- is considered to be the predominant uptake process where $CaSO_4$ is replaced by CaI_2. The greater affinity of CSH phases for IO_3^- over I^- was observed and interpreted as being due to direct bonding between CSH and IO_3^- or to the precipitation of a Ca-IO_3^- containing solid precipitates. If the iodine speciation in leachates from Cast Stone, hydrated lime based grout, or cement/grout solidified solid secondary wastes transform to the highly oxidized iodate species, then iodate has been shown to have a larger adsorption K_d value than iodide (see discussion in Flach et al. 2016) for interactions with subsurface sediments.

Bonhoure et al. (2002) studied iodine speciation in iodine-laden cement minerals using XAS. Solid samples were prepared by mixing 1 g of solid material (hydrated cement paste (HCP) and calcium silicate hydrogels ($CaO*SiO_2*H_2O$ gel, CSH) with 40 mL, pre-equilibrating the suspensions simulated cement paste pore water. The pre-equilibration times were (HCP: 28 days and CSH: 14 days). Then individual slurries were spiked with either I^- or IO_3^- to create slurry concentrations of ($[I]_{tot} = 10^{-3}$ M and 10^{-2} M), respectively. The iodine spiked suspensions were equilibrated again for 28 more days. After centrifugation and decanting the simulated cement paste pore water, the residual wet paste was used for the XAS measurements. A key finding of the XAS characterization of the cement mineral phases was that redox reactions did not influence sorption processes. That is, there was no change of oxidation states observed upon uptake of the either of the iodine species (either I^- or IO_3^-) by HCP or CSH. The HCP and CSH contacted with I^- bearing simulated cement paste pore water showed XANES spectra for only I^- and likewise HCP and CSH contacted with IO_3^- spiked simulated cement paste pore water showed only IO_3^- in the residual solids. Thus there were no changes in iodine speciation over the 14- to 28-d contact times for the two solids immersed in iodine-spiked simulated cement paste pore water.

As observed for the Archive Leach Test Suite of monoliths, the averaged interval D_{obs} values for iodide from the Iodide Suite of monoliths suggests slightly lower leaching when VZP is the eluent in comparison to DIW. The same white precipitate found on the Archive Leach Test Suite monoliths' surfaces is observed on the I-Loading Suite monoliths after several weeks of VZP leaching. Photographs of four of the I-Loading Suite monoliths after 569 days of leaching are shown in Figure 4.11. The left photograph is I-Loading Suite monolith T1-VZP-2, and it shows white precipitate coating the whole surface and copious amounts associated with the top of the monolith. The second photograph (to the right) is I-

Loading Suite monolith T1-DIW-4, and it shows that the surface is relatively devoid of white precipitate, but that white precipitate is associated with micro-cracks that likely were present after the monolith was removed from its mold. The third photograph is monolith T2-VZP-2, and it shows the white precipitate over the entire surface as well as some linear-shaped precipitate along vertical surface cracks that were likely present when the monolith was removed from its mold. The fourth photograph (on the right) is Iodide Suite monolith T3-DIW-4, and it shows little white precipitate on the monolith surface as expected for monoliths leached in DIW. Further, the T3-DIW-4 monolith exhibits a long vertical crack that is not filled with white precipitate (see the red arrow). We speculate that this crack might be recently formed, and thus has not had time to infill with calcium carbonate.



T1-VZP-2 T1-DIW-4 T2-VZP-2 T3-DIW-4

Figure 4.11. Photographs of Iodide Suite Monoliths After 569 Days of Leaching

Similar to the Archive Leach Test Suite eluates, the I-Loading Suite eluates from DIW show higher pH values than the VZP eluates. A plot for the Iodide Suite eluates' pH is shown in Figure 4.12. Figure 4.12 is very similar to Figure 4.5 and in fact, all Cast Stone monoliths of a particular type that have been leached in both DIW and VZP show remarkably similar pH differences at each sampling period (see eluate pH data for all Suites in Appendix B). All I-Loading Suite eluates show a quick rise in pH for the first seven days and then a drop until the 47 and 63 day samplings, followed by a second rise in pH as the interval between samplings increases. When project funding was shut down, there was a large sampling interval between 100 and 369 cumulative days, causing a concomitant rise in pH in eluates. Between the 369-day and 414-day samplings, the pH of all I-Loading Suite eluates dropped.

One other key conclusion that results from all the Cast Stone leach testing performed to date at Hanford is that iodide leaches more rapidly than pertechnetate and chromate, but at about the same rate as nitrate, nitrite, and sodium. As a result of this finding, there are studies, funded by this WRPS Cast Stone program, in which iodide-specific getters are being used to improve the retention of iodide within Cast Stone (see Neeway et al. 2014 and Qafoku et al. 2015 for descriptions of early work).

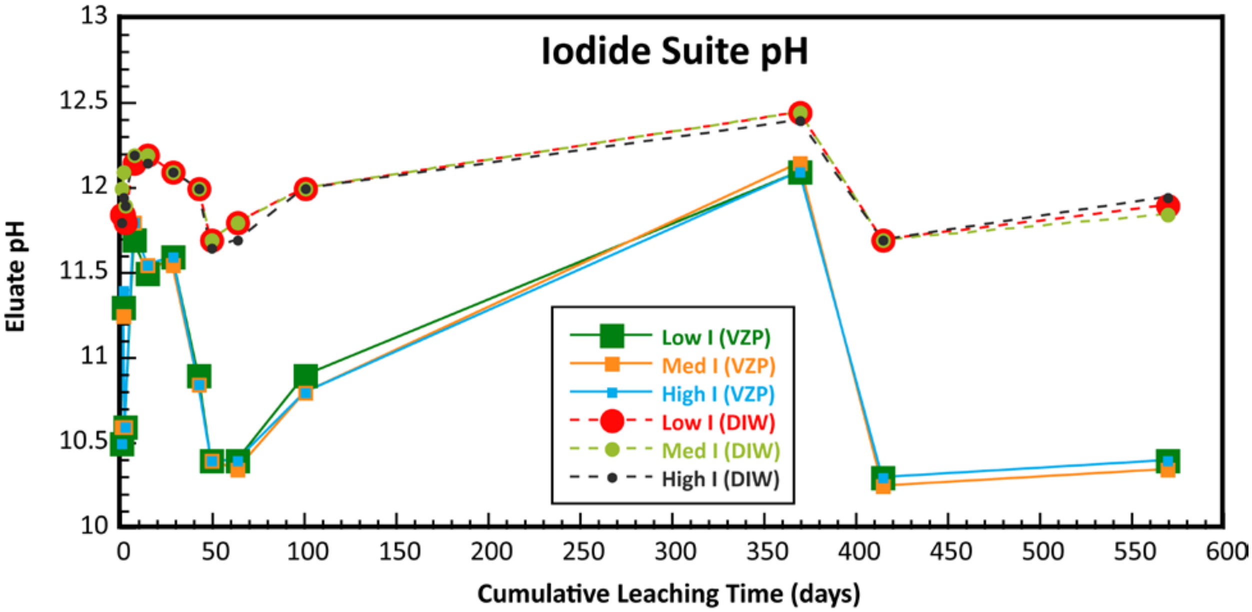


Figure 4.12. pH in Iodide Suite Eluates

4.4 Tc-Gluconate Spiked Suite Results

The objective of the Tc-Gluconate Suite of monolith leach testing was to evaluate the impacts of using a non-pertechnetate form of Tc that appears to be present in Hanford double-shell tanks (DSTs) (see Rapko et al. 2013a, 2013b, Rapko 2014 and Serne et al. 2014 for more details). In this activity, the LAW 7.8 M Na Average simulant was spiked with a Tc(I)-gluconate species then the spiked simulant was solidified into Cast Stone monoliths and subsequently leached using the EPA-1315 leach protocol with DIW and VZP eluents. The leach results for Cast Stone monoliths using the Tc(I)-gluconate spiked LAW simulant are compared to Cast Stone monoliths made at the same time with the same LAW simulant but spiked with pertechnetate, which has been the Tc form used in all previous and more recent simulants under study.

Table 4.7 lists the Tc(I)-Gluconate Suite monolith IDs (and other relevant monoliths made with the LAW 7.8 M Na Average simulant with Tc), details on Tc loading, and which eluent was used in the EPA Method 1315 leach tests. Table 4.8 shows the averaged interval D_{obs} values for three sampling intervals, when applicable, that summarize the ^{99}Tc leach properties from Cast Stone monoliths made using the 7.8 M Na Average simulant with emphasis on differences between the eluents used and speciation of the Tc-spike. The averaged interval D_{obs} values in each cell of Table 4.8 represent averages of the results from two replicate monoliths over the designated time intervals, with the exception of the monolith Archive T3HCS2-7.8AVG leached in DIW that had no replicate monolith. Recall that only the Tc4-Gluc monoliths contain the non-pertechnetate spike, and all the other monoliths contain pertechnetate. Figure 4.13, a bar chart of the 28- to 63-d averaged interval D_{obs}, suggests that the non-pertechnetate species in the T4 monoliths leach more readily than does pertechnetate in all the other pertechnetate-bearing monoliths regardless of eluent (DIW or VZP) used. Further, for the T4 (non-pertechnetate), T5 (pertechnetate), and archived T3 monoliths, considerably more Tc leaches in DIW than in VZP. Observing more leaching to occur when monoliths are leached in DIW in comparison to leaching in VZP is a consistent trend found for all constituents that we are tracking. Figure 4.14 shows the comparable averaged 28- to 63-day interval D_{obs} values for nitrate and sodium from these Tc-bearing monoliths. Based on the fact that all the monoliths should have identical concentrations of nitrate and sodium, one might expect the averaged interval D_{obs} values for these two constituents to be similar for T4 (non-pertechnetate) and all monoliths spiked with pertechnetate. Despite the observation that both nitrate and sodium leach somewhat faster out of the T4 (Tc-gluconate containing) monoliths compared to the T5 (pertechnetate-containing) monoliths in both eluents, the differences are no larger than differences for other pertechnetate-containing monoliths (T3 Archive, and T3 and T22 from the original screening tests). The difference in Tc leaching from the T4 monoliths in comparison to T5 monoliths in both DIW and VZP is larger, thus suggesting that there may be a real difference in the leaching of the non-pertechnetate species compared to pertechnetate. The non-pertechnetate species (identified prior to spiking into the LAW simulant to be Tc(I) species dominated by Tc-tricarbonyl gluconate) leaches faster from the T4 monoliths than Tc(VII)-pertechnetate species present in the T5 monoliths. Whether the Tc in eluates from the T4 monoliths or within the inside of the leached T4 monoliths remains in a non-pertechnetate form has not been determined. Three of the T4 eluates with the highest total Tc concentration were analyzed using nuclear magnetic resonance (NMR) spectroscopy (the technique used to identify the Tc speciation in the original spike solution), but no Tc signal (for any oxidation state or species) was found, even after two attempts of 3-day spectra collection using two different state-of-the-art NMR instruments. The highest concentration of total Tc found in eluates from T4 monoliths ranged from ~12 to ~20 µg/L, well below the detection limit of the NMR (see Levitskaia et al. 2014 for discussion on synthesis, stability, and NMR identification of the Tc(I) species). Thus, at this time, we can only speculate that if the Tc(I) species spiked into the 7.8 M Na Ave simulant and then solidified in the T4 Cast Stone monoliths remained in their Tc(I) forms throughout the leach test, it is possible that these Tc(I) species did not get impacted by the reductants within the BFS as did pertechnetate spiked into the 7.8 M Na Ave simulant that was solidified into T5 monoliths. Significant portions of pertechnetate in the T5 monoliths do get reduced to a low solubility form that only leaches upon being re-oxidized as oxygen in the eluent penetrates into the monolith matrix. Thus, one can hypothesize that the reduced pertechnetate re-oxidizes and diffuses out of the T5 monoliths slower than the Tc(I) species diffuses out of the T4 monoliths even though the Tc(I) species have a larger molecular size. That is, the larger a species is, the slower its diffusion properties through the water-filled Cast Stone matrix would be. However, because the BFS can reduce pertechnetate to a low solubility precipitated solid phase that is slowly re-oxidized back to the soluble (and relatively lower molecular sized) pertechnetate ion, the net result is the observed lower D_{obs} value for pertechnetate. If this hypothesis is accurate the fact that the D_{obs} values for the Tc in T4 monoliths are almost two orders of magnitude lower than for other (assumed non-sorbing) species such as nitrate, one can conclude the Tc(I) species such as Tc(I)-tricarbonyl gluconate in the T4 monoliths is much more chemically reactive than the nitrate, nitrite, and sodium. Or alternatively if the Tc(I) species within the T4 monoliths truly are stable, soluble and not chemically reactive (K_d ~0 ml/g) then the large size of the soluble Tc(I) may be physically hindered more so than smaller molecules, which would suggest that the constrictivity (δ) and/or the tortuosity (τ), are not inherent (independent of species within the pore water) properties of the porous media physical structure. We favor the assumption that the physical parameters of the porous Cast Stone are constant for the porous media regardless of the size of the COC species within the pore water. Thus the Tc(I) species in the T4 monoliths are likely chemically interacting and not present as non-sorbing soluble species. We proffer that the Tc(I) species are not inert/stable and that they are being partially re-oxidized to Tc(VII) soluble species at a rate slightly faster than the rate that pertechnetate within the T5 monoliths is being re-oxidized.

Table 4.7. List of All Monoliths Containing ^{99}Tc Relevant to this Report

Sample ID	**Type of Tc-Spike**	**^{99}Tc Loading (mg/kg)**	**^{99}Tc Loading wt%**	**Eluent**	**Duration of Leach Test (d)**
Tc-Gluconate Suite					
CS-T4-VZ-1	Tc-gluconate	3.29	3.3×10^{-4}	VZP	841
CS-T4-VZ-2	Tc-gluconate	3.29	3.3×10^{-4}	VZP	569
CS-T4-DI-3	Tc-gluconate	3.29	3.3×10^{-4}	DIW	841
CS-T4-DI-4	Tc-gluconate	3.29	3.3×10^{-4}	DIW	569
CS-T5-VZ-1	Pertechnetate	7.915	7.9×10^{-4}	VZP	841
CS-T5-VZ-2	Pertechnetate	7.915	7.9×10^{-4}	VZP	569
CS-T5-DI-3	Pertechnetate	7.915	7.9×10^{-4}	DIW	841
CS-T5-DI-4	Pertechnetate	7.915	7.9×10^{-4}	DIW	569
Archive Suite					
T3HCS2-7.8AVG-3	Pertechnetate	9.32	9.3×10^{-4}	VZP	861
T3HCS2-7.8AVG-4	Pertechnetate	9.32	9.3×10^{-4}	VZP	861
T3HCS2-7.8AVG-5	Pertechnetate	9.32	9.3×10^{-4}	DIW	861
Original Screening Test					
T3HCS2-7.8AVG-2	Pertechnetate	9.32	9.3×10^{-4}	DIW	91
T3HCS2-7.8AVG-3	Pertechnetate	9.32	9.3×10^{-4}	DIW	91
T22HCS2-7.8AVG-3	Pertechnetate	9.42	9.4×10^{-4}	DIW	91
T22HCS2-7.8AVG-4	Pertechnetate	9.42	9.4×10^{-4}	DIW	91

Note: Monoliths with cumulative leach times of 841 or 861 remain actively in on-going tests. Monoliths with cumulative leach times of 569 days are stored for solid phase characterization. Monoliths with cumulative leach times of 91 days have been sent to waste.

Table 4.8. Averaged Interval Dobs Values (cm2/s) for Tc-Gluconate Suite and Related Archive and Screening Test Monoliths

	Eluent = VZP						**Eluent = DIW**						
	D_{obs} 7d up to 841d Avg.	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d up to 841d Avg.**	**σ**		**D_{obs} 7d up to 841d Avg.**	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d up to 841d Avg.**	**σ**
	Tc							**Tc**					
T4 (Tc-Gluconate)	2.17E-11	1.02E-11	2.77E-11	9.55E-12	2.33E-11	1.07E-11	T4 (Tc-Gluconate)	1.81E-10	7.66E-11	2.56E-10	2.73E-11	1.84E-10	8.44E-11
T5 (Pertech)	1.25E-12	4.13E-13	1.30E-12	2.17E-13	1.19E-12	3.93E-13	T5 (Pertech)	2.18E-11	7.61E-12	2.66E-11	5.20E-12	2.15E-11	7.38E-12
Archive T3HCS2-7.8AVG	2.17E-12	1.04E-12	2.70E-12	5.94E-13	2.11E-12	1.13E-12	Archive T3HCS2-7.8AVG	9.59E-11	7.48E-11	5.10E-11	1.43E-11	1.12E-10	7.39E-11
Screening T3	No Mix #22 screening monoliths were leached in VZP						Screening T3HCS2-7.8AVG	too short		3.47E-11	2.75E-12	too short	
Screening T22	No Mix #22 monoliths were leached in VZP						Screening T22HCS2-7.8AVG	too short		2.48E-11	7.54E-12	too short	
	Na							**Na**					
T4 (Tc-Gluconate)	3.25E-09	2.48E-09	3.08E-09	5.21E-10	2.25E-09	1.04E-09	T4 (Tc-Gluconate)	3.94E-09	2.26E-09	3.72E-09	3.91E-10	3.01E-09	1.09E-09
T5 (Pertech)	2.58E-09	2.36E-09	2.25E-09	5.19E-10	1.58E-09	8.03E-10	T5 (Pertech)	3.44E-09	2.30E-09	3.06E-09	3.84E-10	2.53E-09	9.57E-10
Archive T3HCS2-7.8AVG	3.75E-09	2.14E-09	4.37E-09	4.96E-10	3.00E-09	1.47E-09	Archive T3HCS2-7.8AVG	5.16E-09	2.37E-09	5.52E-09	5.89E-10	4.50E-09	2.04E-09
Screening T3	No Mix #22 screening monoliths were leached in VZP						Screening T3HCS2-7.8AVG	too short		5.32E-09	1.10E-09	too short	
Screening T22	No Mix #22 monoliths were leached in VZP						Screening T22HCS2-7.8AVG	too short		4.84E-09	6.41E-10	too short	
	NO_2							**NO_2**					
T4 (Tc-Gluconate)	1.67E-09	1.05E-09	1.60E-09	1.47E-10	1.27E-09	4.42E-10	T4 (Tc-Gluconate)	2.18E-09	9.44E-10	2.25E-09	9.79E-11	1.81E-09	5.11E-10
T5 (Pertech)	1.36E-09	9.39E-10	1.24E-09	1.53E-10	9.87E-10	3.31E-10	T5 (Pertech)	2.01E-09	8.00E-10	2.00E-09	2.28E-10	1.71E-09	3.93E-10
Archive T3HCS2-7.8AVG	3.57E-09	1.84E-09	4.03E-09	3.44E-10	2.91E-09	1.23E-09	Archive T3HCS2-7.8AVG	4.80E-09	1.70E-09	5.39E-09	3.63E-10	4.30E-09	1.37E-09

	Eluent = VZP							**Eluent = DIW**					
	D_{obs} 7d up to 841d Avg.	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d up to 841d Avg.**	**σ**		**D_{obs} 7d up to 841d Avg.**	**σ**	**D_{obs} 28d to 63d Avg.**	**σ**	**D_{obs} 28d up to 841d Avg.**	**σ**
Screening T3	No Mix #22 screening monoliths were leached in VZP						Screening T3HCS2-7.8AVG	too short		5.44E-09	1.209E-09	too short	
Screening T22	No Mix #22 monoliths were leached in VZP						Screening T22HCS2-7.8AVG	too short		4.14E-09	4.74E-10	too short	
	NO_3							**NO_3**					
T4 (Tc-Gluconate)	2.92E-09	2.05E-09	2.56E-09	2.31E-10	2.10E-09	7.50E-10	T4 (Tc-Gluconate)	3.96E-09	1.85E-09	3.57E-09	2.15E-10	3.19E-09	6.50E-10
T5 (Pertech)	2.23E-09	1.72E-09	1.86E-09	3.19E-10	1.51E-09	5.05E-10	T5 (Pertech)	3.50E-09	1.41E-09	3.07E-09	4.15E-10	2.94E-09	5.39E-10
Archive T3HCS2-7.8AVG	4.22E-09	2.31E-09	4.62E-09	7.49E-10	3.42E-09	1.56E-09	Archive T3HCS2-7.8AVG	6.13E-09	1.88E-09	6.32E-09	1.29E-09	5.58E-09	1.49E-09
Screening T3	No Mix #22 screening monoliths were leached in VZP						Screening T3HCS2-7.8AVG	too short		4.41E-09	6.22E-10	too short	
Screening T22	No Mix #22 monoliths were leached in VZP						Screening T22HCS2-7.8AVG	too short		4.31E-09	7.65E-10	too short	
	Cr							**Cr**					
T4 (Tc-Gluconate)	*<1.38E-15*	2.26E-15	*<2.89E-15*	3.10E-15	*<1.46E-15*	2.48E-15	T4 (Tc-Gluconate)	9.83E-14	7.09E-14	9.57E-14	2.59E-14	7.27E-14	5.11E-14
T5 (Pertech)	*<3.07E-15*	6.76E-15	*<3.01E-15*	3.22E-15	*<1.55E-15*	2.56E-15	T5 (Pertech)	7.33E-14	5.43E-14	6.73E-14	2.07E-14	5.48E-14	4.20E-14
Archive T3HCS2-7.8AVG	*<1.37E-15*	2.25E-15	*<2.95E-15*	3.16E-15	*<1.42E-15*	2.48E-15	Archive T3HCS2-7.8AVG	2.74E-13	6.82E-13	7.98E-14	2.62E-14	3.15E-13	7.56E-13
Screening T3	No Mix #22 screening monoliths were leached in VZP						Screening T3HCS2-7.8AVG	too short		1.64E-13	4.26E-14	too short	
Screening T22	No Mix #22 monoliths were leached in VZP						Screening T22HCS2-7.8AVG	too short		1.79E-13	5.63E-14	too short	

Too short = test duration was only 91 days. Archive T3HCS2-7.8AVG monoliths data up to 861 days leaching, while T4 and T5 monoliths data averaged to 569 or 841 days as shown in Table 4.7.

Italicized numbers for Cr interval D_{obs} values included eluates where Cr concentrations were less than detection values.

Note that none of the interval averaged D_{obs} values have been corrected for inventory depletion

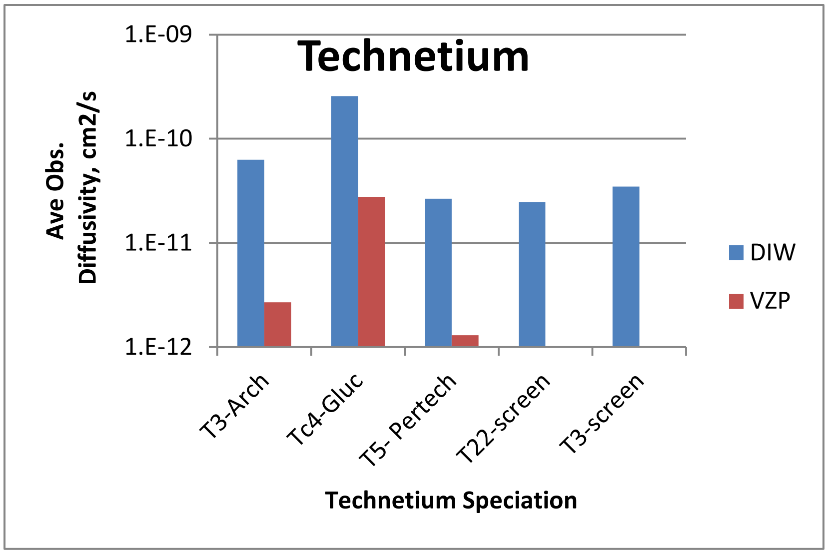


Figure 4.13. Averaged Interval 28-d to 63-d D_{obs} Values for Tc from Monoliths Leached in DIW vs. VZP

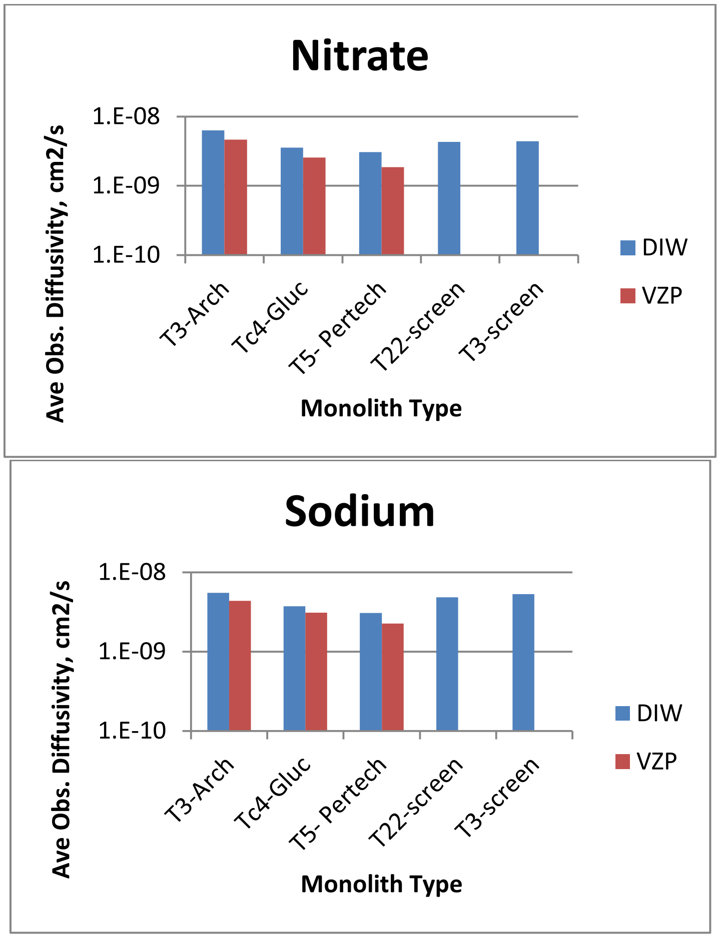


Figure 4.14. Averaged Interval 28-d to 63-d D_{obs} Values for Nitrate and Sodium from Tc-Monoliths Leached in DIW vs. VZP

T4-VZP-2 (Tc-gluconate) T4-DIW-3 (Tc-gluconate) T5-VZP-1 (Pertech) T5-DIW-4 (Pertech

Figure 4.15. Photographs of Tc-Gluconate and Pertechnetate Monoliths Leached in Two Eluents for 361 Days

Table 4.9. Tc and Cr Cumulative Fraction (%) Leached After 841 Days Leaching for T4 and T5 Monoliths

Eluent	T4 (Tc)	T5 (Tc)	T4(Cr)	T5 (Cr)
VZP	4.3	1.2	<0.03	<0.07
DIW	11.2	3.9	~0.20	0.18

Figure 4.15 shows photographs of the T4 (containing Tc-gluconate) and T5 monoliths (containing pertechnetate) after 361 days of leaching in either VZP or DIW. Photographs of T4 and T5 monoliths that have been leached for 569 and 861 days look similar to the photographs shown in Figure 4.15. As found for all the other Suites of monoliths studied, the monoliths leached in VZP show significant amounts of secondary mineral formation on their surfaces, while the monoliths leached in DIW show clean surfaces with a few linear traces of white precipitate along hairline surface cracks. Figure 4.16 plots the change in weight of T4 and T5 monoliths leached in VZP and DIW as a function of time. In general, there is a slight weight gain for the monoliths leached in VZP that increases with leach time, especially for T5 monoliths that contained the pertechnetate spiked simulant. In contrast, there is a general decrease in weight of the monoliths leached in DIW, especially for the T4 monoliths that contained the Tc-gluconate spiked simulant. Based on the appearance of white precipitate on the surfaces of the monoliths leached in VZP, it makes sense that there is an increase in the monolith mass as leaching continues. We note that there is less weight gain for the T4 monoliths containing the Tc-gluconate spike in which we are not completely sure what the overall composition of the concentrated small volume (~7 mL) of stock solution was that was added to 493 mL of simulant. The Tc-gluconate stock solution contained 0.75 M sodium hydroxide and likely an undefined amount of residual gluconate and other precursor species used to synthesize the Tc(I)-tricarbonyl gluconate. The weight loss from the T4 monoliths leached in DIW exceeded the loss from the T5 monoliths leached in DIW. We know that only 0.76 mL of stock pertechnetate solution in dilute ammonium hydroxide solution was used to spike 500 mL of the simulant. The weight gain or loss of the T4 monoliths over time suggests that there was more dissolution/leaching of the T4 monoliths that reduced the weight of the monoliths and conversely reduced the weight gain from secondary precipitates form on T4 monoliths leached in VZP compared to the T5 monoliths for which we have full knowledge of the composition of the spiked simulant. We do not know whether the small volume (7 mL) addition of somewhat uncharacterized Tc-gluconate spike to the 493 mL of 7.8M Na Ave simulant could have made the final cured Cast Stone T4 monoliths less resistant to overall dissolution/leaching than the T5 monoliths that contained well characterized pertechnetate spiked simulant.

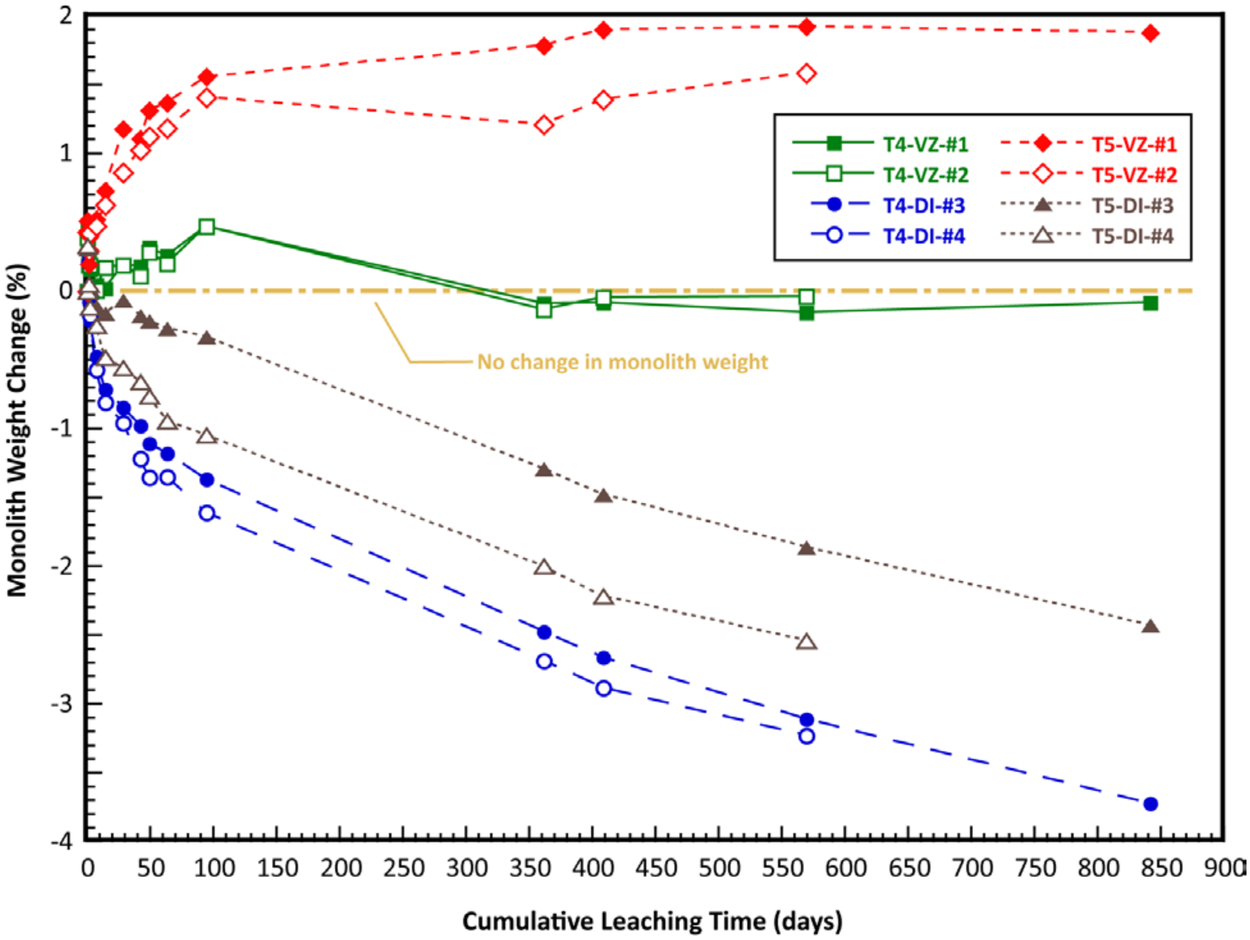


Figure 4.16. Change in Monolith Weights as a Function of Time and Eluent Used

As found for all the 2-in diameter by 4-in long Cast Stone monoliths regardless of test suite, the cumulative fraction leached for mobile contaminants (iodide, nitrate, nitrite, and sodium) from the T4 monoliths exceeded 20% of initial value generally between the 63- to 94-day sampling when leached in VZP, and near the 63-day sampling when leached in DIW. For the T5 monoliths, mobile constituents reached or exceeded 20% cumulative leached close to the 94-day sampling when leached in VZP and between the 63- to 93-day samplings when leached in DIW. Per the discussion in ANS16.1 methodology, when the cumulative fraction leached exceeds 20%, the monoliths no longer follow the semi-infinite source term assumption, and observed diffusion coefficient calculations based on Eq. 3.1 are no longer accurate. Thus, we have been recommending the use of the averaged interval D_{obs} values for sampling times between 28 and 63 days as the best values to use in simple diffusion release conceptual models, if such conceptual models are chosen for long-term Cast Stone release predictions. The cumulative release of Cr and Tc from the T4 and T5 monoliths never comes close to this 20% limit by the end of 841 days of leaching. Table 4.9 shows the cumulative percentage leached for these two constituents after 841 days of testing. Of note for Tc is the difference in % released between VZP and DIW eluents wherein more Tc is released into DIW eluate and the increased Tc release % for the T4 (Tc-gluconate) versus the T5 (pertechnetate) monoliths. The latter again suggests that there may be a demonstrable difference in Tc leach tendencies dependent on Tc speciation. The Cr release data again shows less release of Cr into VZP eluate than DIW but there is no difference between T4 and T5 monoliths as would be expected because the Cr speciation and total mass solidified in T4 and T5 monoliths were identical.

5.0 Conclusions and Recommendations for Future Activities

In this section we summarize the key conclusions from the extended leach testing activity and present recommendations on activities that could be continued on the existing leach tests as well as potentially useful characterization studies on some of the leached monoliths. Then we discuss new studies and activities that could potentially improve the long-term understanding of constituent release from Cast Stone/grout waste forms containing Hanford Site liquid waste streams. Gaining a more mechanistic technical understanding of the long-term contaminant release performance of Cast Stone/grout would make the development of the required performance assessment for the IDF more defensible. The recommendations suggested below should be vetted (and perhaps expanded) by discussions among U.S. Department of Energy Office of River Protection (DOE-ORP) management, WRPS and their subcontractors, Hanford Site regulators, and stakeholders.

5.1 Conclusions from the EPA Method 1315 Leach Testing of Cast Stone Monoliths made with LAW Simulants

Despite finding a two order of magnitude range in ^{99}Tc and Cr observed diffusivities and almost an order of magnitude range in observed diffusivities for the more leachable anions (nitrate, nitrite, iodide) and sodium, the results of the screening tests (see Westsik et al. 2013a) that leached 26 different Cast Stone mixes made with 4 different LAW simulants for 91 days found that there were no statistically significant correlations between the leach properties of any of these key constituents with the type of waste simulant, the source of the dry blend, or wet slurry properties. There does seem to be a weak correlation with the free-water-to-dry blend mix ratio.

Thus, monolith leach studies continue with the intention of identifying correlations that shed light on the controlling mechanism(s) that lead to the variation in leach properties. Conclusions derived after leaching four different suites of Cast Stone monoliths for almost three years follow. One universal observation from these extended leach studies on LAW Cast Stone monoliths, when leached in VZP eluent, is that the leach rates of the five constituents studied are lower than when leached in DIW. One potential cause for the lower leach rates is the formation of significant quantities of secondary precipitates on the surfaces (and perhaps deeper into the interior) of the monoliths contacted with VZP, while the monoliths contacted with DIW show only trace quantities of secondary precipitates that are generally associated with surface hairline micro-cracks present after curing. In general monoliths leached in VZP gain mass over time while monoliths leached in DIW lose mass. Gaining mass reflects precipitation of solids from the VZP eluent's dissolved salt content. Losing mass suggests net dissolution of material from the cured monoliths.

The leach rates of the mobile constituents (nitrate, nitrite, iodide, and sodium), through ~34 months of leaching in DIW, showed a continual decrease; a faster decrease in the first seven days followed by a continual slow decrease over longer times. For monoliths leached in VZP there may be a very slight "late stage" increase in leach rates for nitrite after about 427 days of leaching. This slight increase in interval D_{obs} values for nitrite may be associated with biological interactions associated with surface black spots on selected monoliths. The VZP eluent used for the Archived Suite of monoliths after the 380 day solution exchange was found to have biological activity that reduced much of the starting nitrate (~220 mg/L nitrate in simulant dropped to ~54 mg/L nitrate) present in the simulant to nitrite. This represents ~75% reduction of the starting VZP nitrate. The increase in nitrite in the blank VZP eluates was 122 mg/L which is equivalent to the ~166 mg/L of nitrate lost given the stripping of one oxygen from each mole of nitrate to form nitrite (166 mg/L nitrate becomes 123 mg/L of nitrite upon loss of an oxygen by reduction). It is possible that the "biological infected" VZP eluent facilitated biological growth/attachment to Archived Suite monoliths immersed in VZP during and after the 427 sampling event. Future solid phase characterization of Archived Suite monoliths removed from leach testing at the 590-d sampling event is ongoing. Some of the black spots have been submitted to microbiologists for characterization. Aside from very slight increases in nitrite D_{obs} values beyond the 380-d sampling all the other constituents of interest show the slow decrease in D_{obs} versus time as leaching in VZP eluent continues. The observed coating of secondary precipitates on the VZP-contacted monoliths continues to slowly increase and for at least two of the original Cast Stone Mixes (T17 and T21) show cracking and near spalling (see Figure 4.6) of pieces from the bottoms of the monoliths. These two mixes have consistently shown high leach rates for the mobile constituents regardless of which eluent was used. Figure 5.1 shows the weight status of Archived Suite monoliths T17 and T21 leached in both VZP and DIW. Based on the monoliths weights after each leach interval there is no indication that pieces with significant weight have spalled off the monoliths leached in VZP as could have occurred in the later sampling intervals based on Figure 4.6. It is interesting to note that monoliths from Mix 21, the mix with the highest leaching for almost all constituents shows the largest mass loss. Even the T21 monoliths leached in VZP show consistent mass loss whereas most all other monoliths leached in VZP show some weight gain as leaching progressed (see also Figure 4.16). We speculate that Mix 21, which was made with the 7.8 M Na high sulfate simulant at the 0.6 free water to dry blend weight ratio is inherently a difficult liquid waste to solidify and its cured monoliths leach COCs and dissolve its matrix more than other mixes.

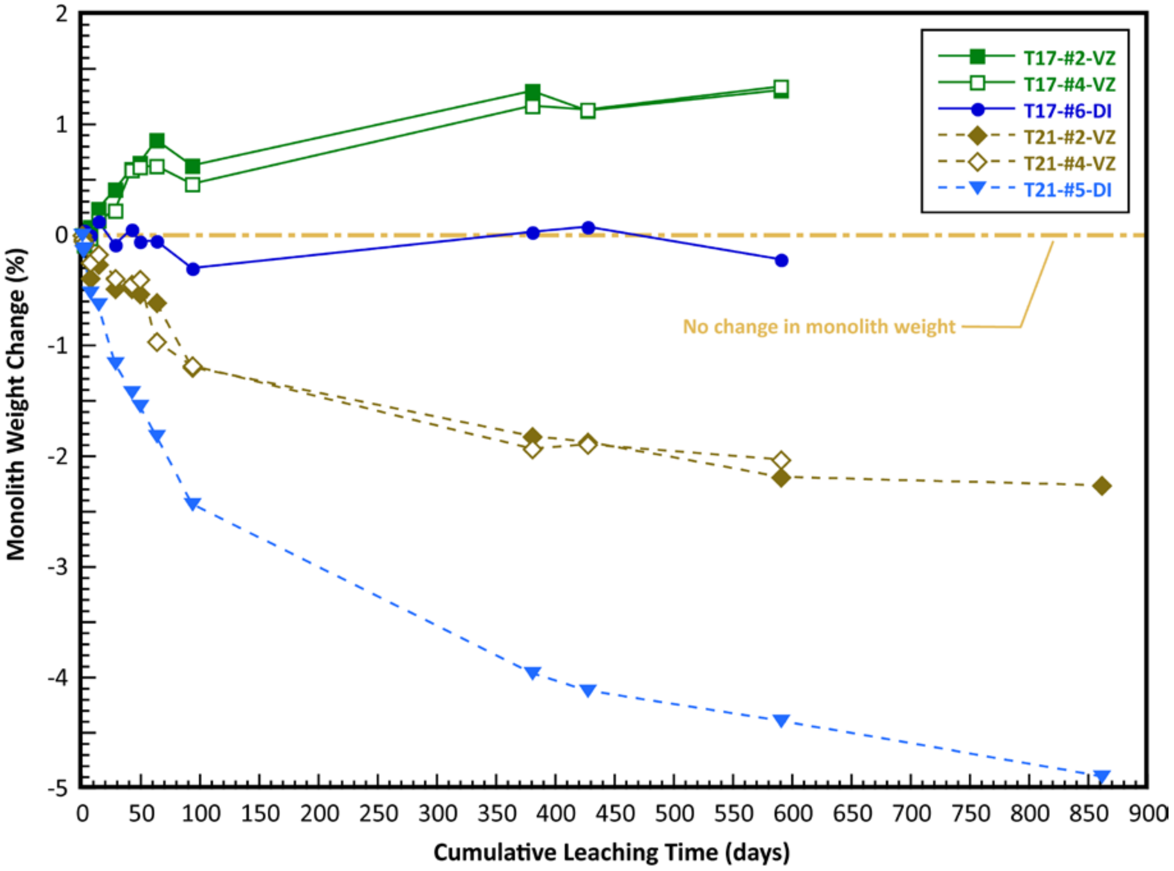


Figure 5.1. Changes in T17 and T21 Archive Suite Monoliths as a Function of Cumulative Leach Time and Eluent

In general, all monoliths leached in VZP have a significant white precipitate coating on their surfaces such that it is difficult to readily evaluate whether there are new micro-cracks developing. There do appear to be some “new” micro-cracks developing on monoliths leached in DIW after a year or more but there is no increase in interval D_{obs} values at long leaching times; however, the new micro-cracks might slow down the observed continual slow decrease in D_{obs} values as the original source inventories deplete well beyond 20%.

For all the monoliths studied herein, tables of interval averaged D_{obs} values (a way of quantifying leach rates) for ^{99}Tc show a factor of about 50 difference between the best performing Cast Stone mix to the worst performing mix, regardless of which eluent is used. The same range of interval averaged D_{obs} values for best to worst Cast Stone mix for Na, I, and nitrate differ by a factor of about 6, and for nitrite by a factor of 3. Chromium interval averaged D_{obs} values for VZP eluents are generally not quantifiable because eluate Cr concentrations were below detection limits. There was one exception—archived monoliths from mix 10 (5 M Na high sulfate LAW waste solidified with NW sources of fly ash and BFS at a water-to-dry blend ratio of 0.6) had measureable Cr in all VZP eluates as well as DIW eluates. The average 28 to 63 day interval D_{obs} values for this mix 10 monoliths leached in VZP was 4.0×10^{-14} cm^2/s and 2.4×10^{-13} cm^2/s when leached in DIW. Cr leach rates (quantified by D_{obs}) from Cast Stone leached in DIW are also low, but quantifiable, and ranged from 3.3×10^{-12} to 5.8×10^{-15} cm^2/s. Again, lower Cr leach rates are found when monoliths are leached in VZP compared to monoliths leached in DIW. These low values suggest that Cr in Cast Stone, which contains BFS, is likely sequestered as reduced Cr(III) in very low solubility phases, perhaps $Cr(OH)_3$ or solid solutions of $(Fe,Cr)(OH)_3$. In summary, Cr leach rates from Cast Stone are very low in either eluent, such that potential risk to groundwater from released Cr is unlikely.

The best and worst performing Cast Stone monolith mixes leached in VZP have moderate correlation with the best and worst performing monoliths when leached in DIW for each constituent of interest. Based on the Screening Leach Tests –Extended Suite, where 11 of the original 26 Screening Test mixes were leached for up to 1042 d in DIW only, Cast Stone Mixes 14 and 18 are the best performing mix for restricting the release of ^{99}Tc, and Mix 8 is the worst performing for retaining ^{99}Tc. For iodide, sodium, nitrate and nitrite leaching, Cast Stone Mixes 8 and 18 are the best performing and Mix 21 is the worst performing. When Archived Suite monoliths from these same mixes were leached for up to 861 d in VZP the best performing mix for retaining Tc was mix 24 and the worst performing mixes were 16 and 17. For iodide retention Mixes 10 and 24 were best and Mix 21 was the worst. For Na Mixes 13 and 14 were best performing and mix 21 worst. For nitrate and nitrite Mixes 8, 13, 18 and 24 had the lowest release rates and mix 21 the highest release rate.

A key finding for the Archived Suite of monoliths is that (as shown in Figure 4.2) is that for all five COCs D_{obs} values for monoliths leached in DIW when compared to “duplicate” monoliths leached in VZP are higher. The difference in D_{obs} values is most striking for Tc where the 28-d to 63-d averaged D_{obs} values show an average difference of a factor of ~16 larger Tc observed diffusivities when leached in DIW in comparison to values leached in VZP. The range in the ratio of 28-d to 63-d Tc D_{obs} values in DIW to D_{obs} values in VZP is 2.4 to 53, with Mix 14 having the lowest ratio and Mix 21 having the highest ratio. The difference in Cr D_{obs} values between DIW and VZP leached monoliths is also large but because Cr eluate concentrations were usually below detection limits accurate Cr D_{obs} values for the VZP leached monoliths were calculated using the eluate Cr detection limit. The range in the DIW-to-VZP D_{obs} ratio for the more mobile constituents is much lower but consistently shows iodide, sodium, nitrite and nitrate leases slightly faster in DIW than in VZP. The range in the DIW-to-VZP D_{obs} ratio for these four mobile constituents ranges from 1.06 to 2.03 and consistently is highest for Mix 17 and lowest for Mix 18 and/or Mix 8.

For uranium, when leached in VZP, almost all the eluates had no detectable uranium. Using the detection limit for uranium in the eluates and the starting inventory of uranium, the calculated D_{obs} values range from <1×10^{-16} to <3×10^{-16} cm^2/s. For two Mixes (8 and 16) leached in DIW, the eluate uranium concentrations were detectable yielding D_{obs} values of 1×10^{-15} and 4×10^{-16} cm^2/s, respectively.

Results reported by Pabalan et al. (2012) suggest that uranium is immobilized within SRS saltstone as a $CaUO_4$ (calcium uranate) phase, which is highly insoluble. Note that this uranium compound has U in the +6 oxidation state, thus uranium does not appear to be reduced by the BFS. In Wellman et al. (2007), Portland cement specimens were prepared with a U(VI) nitrate spike and the specimens were aged for various time spans ranging from 2 weeks to 1 year. SEM-energy dispersive spectroscopy (SEM-EDS) and XRD were used to identify the uranium-bearing compounds formed in the specimens. These investigations demonstrated the formation of uranium minerals, after only 2 weeks, within the hardened cement that was under saturated with respect to uranium solid phases and where sorption/co-precipitation was believed to be the mechanism of retention. Cement specimens spiked with large amounts of uranium that resulted in over saturation with respect to uranium mineral phases exhibited extensive formation of discrete uranium oxyhydroxide phases within the first two weeks. The significance of the uranium paragenetic sequence was clearly demonstrated during the subsequent 2-month time frame. Uranyl oxyhydroxide phases were followed by the formation of mixed uranyl-oxyhydroxide/silicates, with further formation of uranyl-silicates, then mixed uranyl-silicate/phosphate phases formed, and finally uranyl-phosphate phases formed. Wellman et al. (2007) identified the uranium minerals sequence to be soddyite, becquerelite, uranophane, and autunite. This finding is consistent with that of Ewart et al (1992), Krupka and Serne (1996), and Criscenti et al. (1996) that uranium phases other than schoepite could be the controlling phase in cement.

Golovich et al. (2011) and Wellman et al. (2008) performed uranium mineral solubility tests in five simulated cement/grout waste form pore waters (taken from work by Ewart et al. (1992), including one simulant of the pore water generated by a 90% BFS:10% OPC mix. The aqueous uranium concentrations in these solubility tests were measured for up to 405 days of reaction. After this time period the uranium in the solids were characterized using SEM-EDS and XRD to look for secondary mineral formation (conversion of the starting uranium mineral to another uranium phase). The starting uranium phases were autunite [$Ca(UO_2)_2(PO_4)_2 \cdot 12H_2O$], soddyite [$(UO_2)_2SiO_4 \cdot 2H_2O$], becquerelite [$Ca(UO_2)_6O_4(OH_6) \cdot 8H_2O$], and uranophane [$Ca(UO_2)_2(SiO_3OH)_2 \cdot 5H_2O$]. All of these minerals have uranium present in its U(VI) valence state. After the long exposure of each mineral in the five simulated grout waste form pore waters steady-state aqueous uranium concentrations were observed except for soddyite and becquerelite in some of the waste form pore-water simulants. The final solids, in general, did not show significant transformations to other uranium phases in most of the five simulants, suggesting that each of these uranyl based minerals are relatively stable in cement based waste forms. The observed aqueous uranium concentrations generally were inversely correlated with the simulant's Ca concentration. That is, as Ca concentration decreased uranium solution concentrations increased. These studies do not preclude BFS as being capable of reducing uranyl species in liquid wastes that are solidified, rather the studies suggest uranium solidified in Cast Stone does not have to be reduced to U(IV) species in order to be readily sequestered and to exhibit extremely low diffusivities as observed in all the Cast Stone leach tests described herein.

Regardless of the Cast Stone mix composition, the D_{obs} values for the anions iodide, nitrate, and nitrite and the cation sodium are very similar (as mentioned the range in averaged interval D_{obs} values for these four constituents varies by only a factor of three to six). Cr leaches the slowest especially when VZP eluent is used, followed by ^{99}Tc, and then the four more mobile constituents.

Because the cumulative mass of mobile constituents released from the monoliths starts to exceed 20% of their starting inventories when VZP is used as the eluent generally from 49 to 63 days of leaching, we are recommending that the VZP leached averaged 28- to 63-day interval D_{obs} values be used to represent the most probable values for any future IDF PA predictive modeling for LAW solidified Cast Stone, if an empirical diffusivity conceptual model is chosen for describing release from LAW Cast Stone. The D_{obs} values for the COCs from earlier leaching periods (0.08 to 14 days) and the D_{obs} values for tests that used DIW as the eluent could be used to create a range of D_{obs} values that could be used in sensitivity analyses. The early leaching data for 0.08 to 7 days for all Cast Stone and most cementitious waste forms described in literature in general always exhibit high interval D_{obs} values; often an order of magnitude and in a few cases up to two orders of magnitude higher than interval D_{obs} values calculated after 28-days and longer. We don't generally recommend using the very earliest data for 0.08 to 7 days because when leach testing small monoliths the "surface wash off" and release from "surface pores" that directly intercept the monolith outer surface dominate the early COC release data. The wash off effect is believed to be caused by salts that evaporate on the monolith surface during curing. These early interval D_{obs} values are considered to be biased high for COCs, especially mobile ones such as iodide, NO_3, NO_2, and Na. In the actual disposal setting, the cementitious waste forms will be much larger (have a much smaller SA/V ratio) so the wash off and "surface pores" will have a much lower impact on the very early release of COCs.

The observed diffusion coefficients, calculated from the EPA Method 1315 tests extended to long leach times, tabulated in this Rev. 1 report are for Cast Stone monoliths that used LAW simulants. The two recent IDF waste form data packages, Cantrell et al. (2016) and Flach et al. (2016), focus on cementitious waste forms that solidify liquid and solid secondary wastes, respectively, generated during operation of the WTP. The LAW waste stream simulants solidified in Cast Stone are 5–7.5 M sodium solutions (25–35 wt% solids, pH >14) with nitrate and hydroxide as the major anions. The current liquid secondary waste (LSW) waste stream simulants are primarily ammonium sulfate (10–20% solids) and are near neutral pH. Therefore, the LSW simulants are being solidified in a different mix of dry ingredients than Cast Stone, and the subsequent hydration, weathering, and leaching reactions that occur may vary. The observed diffusion coefficient values tabulated in this Rev. 1 report are similar to those found in the last columns in Table 3.1 in Cantrell et al. (2016), which lists the geometric mean and low to high range values for the 26 Cast Stone mixes made with four different LAW simulants. However, given the difference in both the liquid waste simulants and the modified dry blend composition, the D_{obs} reported in this Rev. 1 report should not be expected to be quantitatively identical to those for hydrated lime based solidified secondary liquid wastes (values shown in the first several columns in Table 3.1 of Cantrell et al. The same is true for observed diffusion coefficients provided in tables found in Section 8 of Flach et al. (2016), which list D_{obs} values for COPCs present in solid secondary wastes solidified/encapsulated into cementitious grouts. On the other hand, in general, the numerical values for observed diffusion coefficients for the five COPCs most often of interest (Tc, I, Cr, nitrate, and nitrite) do show qualitatively the same trends amongst the three waste forms (Cast Stone solidified LAW liquid simulants, hydrated lime-based grout solidified ETF-LSW simulants, and cement/grout solidified secondary solid wastes) and in general, each recommended D_{obs} value for a specific COPC agree within an order of magnitude amongst the three waste forms. If there is a need to select maximum (worst case) D_{obs} values to obtain a range to perform sensitivity analyses, the range provided in Table 3.1 (right hand column) in the recent cementitious secondary waste data package (Cantrell et al. 2016) should be used.

When the cumulative mass of a constituent released from the monoliths starts to exceed 20% of their starting inventories, use of Eq. 3.1 or Eq. 3.2 to calculate the observed diffusion coefficient is not accurate because the leaching conditions no longer satisfy the assumed semi-infinite source condition. Based on Eq. 3.1 and/or Eq. 3.2, plots of cumulative fraction released versus the square root of cumulative leach time should yield a straight line for each constituent of interest to satisfy the simple diffusion-controlled release. Figure 5.2A shows an example plot for monolith T8LCS-5RAS-4 from the Extended Suite that was leached in DIW. As shown, the data for the more mobile constituents (nitrate, nitrite, iodide, and sodium) do not create a straight line, but the data for Tc and Cr do form a straight line from the 0-0 axis all the way to the end of testing, suggesting that a diffusion mechanism with a semi-infinite source term is in fact capable of explaining the data. However when the Tc and Cr fractions leached are plotted on an expanded scale as shown in Figure 5.2B, the Tc data shows some deviation from a linear line. At this time, we can only note that the cumulative fraction released for Tc and Cr from the Cast Stone monoliths studied herein is very low and well below the 20% value wherein inventory depletion starts to impact simple data analysis using Eq. 3.1 and Eq. 3.2. One should not imply from this observation that the mechanism controlling the release of Tc and Cr from these monoliths is merely diffusion of the TcO_4^- and CrO_4^{2-} that were present in the liquid waste simulants through the tortuous Cast Stone internal matrix. Rather, the controlling mechanism is likely oxygen diffusion into the Cast Stone matrix with concomitant re-oxidation of reduced Tc and Cr species, followed by their diffusion as their oxyanions from the Cast Stone matrix. We suggest that readers look at recent reports authored by SRNL scientists that describe studies directed at oxygen ingress into Cast Stone monoliths and the subsequent fate of Cr and Tc (Almond et al. 2012a; Langton and Almond 2013; Langton 2014). Studies of this nature will be needed to unravel the controlling mechanisms for the leach properties of redox sensitive constituents, such as Tc and Cr, from Cast Stone and other grouts using BFS as one of the dry blend components. Such studies will be especially useful for simulating the IDF water unsaturated burial environment. One of the key observations from the cited SRNL studies is that oxygen ingress into Cast Stone, saltstone, and other grout waste forms may be more rapid under water unsaturated conditions than in the typical water saturated tests such as EPA 1315. Such a hypothesis has merit, given the relatively larger concentration of oxygen in air (~20 wt%) versus the oxygen content of air saturated water (~8 mg O_2/L of water). Diffusion coefficients for gases through porous media are ~10^{-3} cm^2/s while the diffusion coefficients of dissolved species in water are ~10^{-5} cm^2/s.

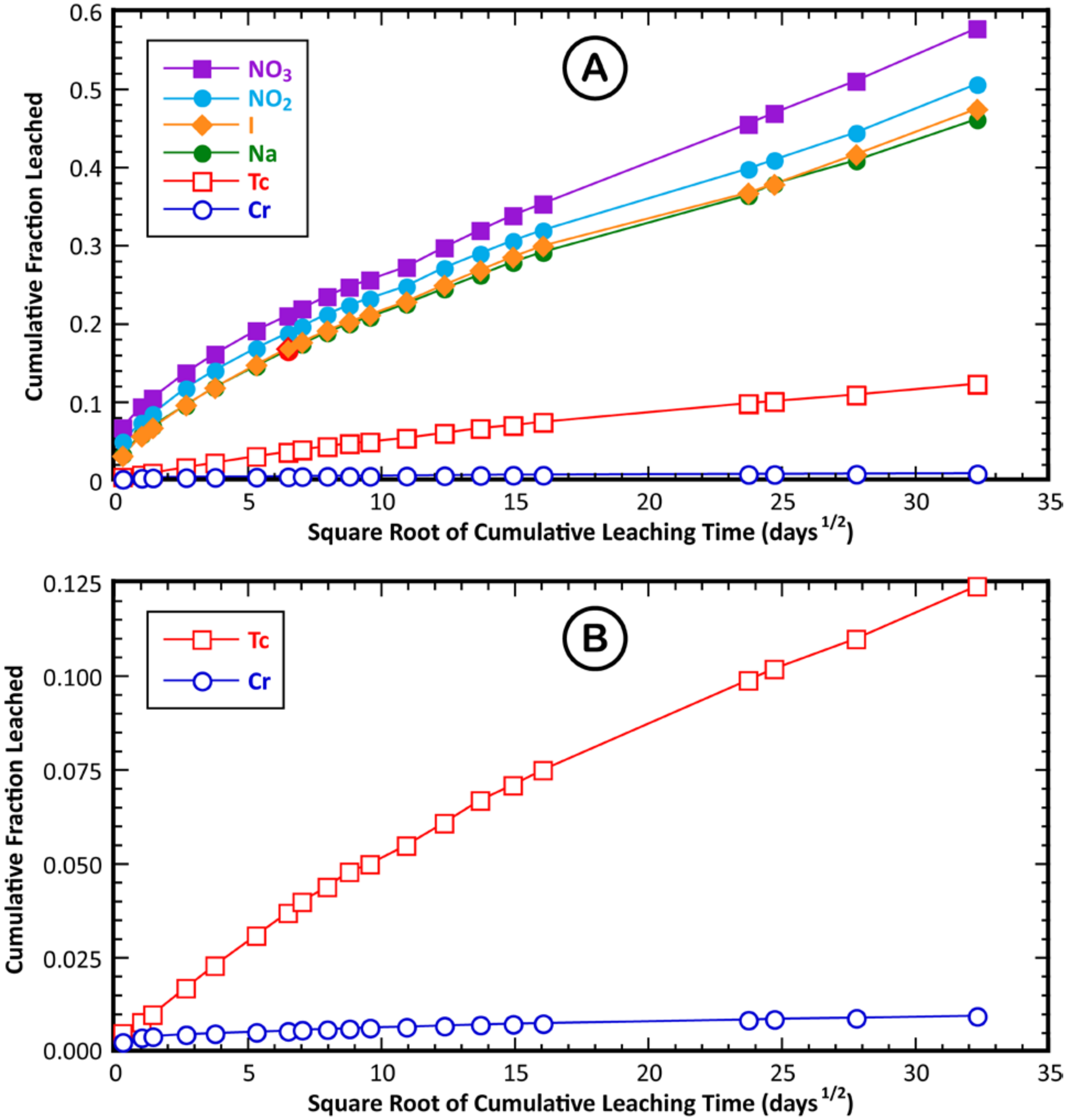


Figure 5.2. T8LCS-5RAS-4 Leached in DIW for 1042 d; (A) all COCs (B) Tc and Cr

The data for the mobile constituents start to deviate significantly from the straight line starting at the (0,0) axes intercept when cumulative fraction leached reaches the 20% value. Among other potential causes, the depletion of the starting inventory is likely the dominant cause for the deviation shown in Figure 5.2. There are other mathematical relationships available in the literature (Thomas 1987; Pescatore 1990, 1991) that could be used to calculate observed diffusion coefficients (D_{obs} values) for experiments where inventory depletion is occurring (such as ours). If more accurate D_{obs} values are required in future predictive modeling these more complicated data analysis approaches could be used. The two Pescatore references explicitly address finite inventory and finite dimensional waste forms with rectangular shapes, which at the moment is the baseline configuration for LAW solidified Cast Stone, should Cast Stone be chosen as a supplemental waste form for LAW. The current baseline for ETF-processed solid secondary waste forms destined for burial at IDF is solidification in a grout by a commercial off-site waste treatment vendor.

The white precipitate on the VZP monolith surfaces (and perhaps within the monoliths interior)[1] has been identified by XRD as predominantly aragonite (a polymorph of calcium carbonate) that has a slightly higher solubility product, K_{sp}, than the K_{sp} for calcite[2]. The white precipitate also contains some brucite [$Mg(OH)_2$]), and perhaps minor amounts of calcite. The same minerals are likely covering some of the surface micro-cracks on monoliths leached in DIW, but we have not recovered adequate precipitate mass off the DIW leached monoliths to get a good XRD spectrum.

The Iodide Suite leach results for monoliths with four different starting iodide inventories (the three inventories Low, Med, and High in the Iodide Suite and the original Screening Test inventory found in the Extended and Archive Suites) show no significant differences in the interval D_{obs} values. Thus, we conclude that at iodide loading well below 0.14% (the lowest loading used in previous studies that showed differing iodide leach rates as a function of iodine loading), there should not be loading sensitivity. HTWOS predictions of ^{129}I masses (converted from activity) in future Hanford waste streams are several orders of magnitude below this threshold value. The iodide loading values used in our Iodide Suite studies ranged from 1.6×10^{-5} to 3.6×10^{-4} wt% and past screening tests used 7.7×10^{-3} wt%. The lower limit, 1.6×10^{-5} wt%, resulted in iodide concentrations in eluates that were nearing detection limits (using ICP-MS analysis of stable ^{127}I as a surrogate for ^{129}I). Thus, in future Cast Stone testing with simulants, it should not matter what iodide loading is used as long as the value is kept below ~0.01 wt% and above ~5×10^{-5} wt%. Earlier iodine loading studies found in literature that suggest that iodide loading does impact cement/grout leach tendencies were performed at total iodine loadings much higher (some up to 9 wt%) than will ever occur at Hanford using actual liquid waste streams. Although we have not spiked Cast Stone with other possible forms of iodine (e.g., iodate) we do not expect loading impacts for other iodine species as long as low total iodine loadings are used in future Cast Stone formulations. Both Atkins and Glasser (1990) and Bonhoure et al. (2002) state that iodide should be the dominant iodine species in almost all cements and grouts including those containing BFS. Further, Bonhoure et al. (2002) found no interconversions of iodide or iodate contacting hydrated cement paste or hydrated calcium silicate hydrogel over 28 days of contact using XANES spectroscopy. If the iodine speciation in grout leachate does transform to the highly oxidized iodate species, then iodate has been shown to have a larger adsorption K_d value than iodide (see discussion in Flach et al. 2016) for interactions with subsurface sediments. Thus, iodate's migration away from the weathered waste forms should be retarded more than iodide.

One other key conclusion that results from all the Cast Stone leach testing performed to date on Hanford Cast Stone waste forms is that iodide leaches more rapidly than pertechnetate and chromate, and at about the same rate as nitrate, nitrite, and sodium.

The leaching results for Tc-bearing monoliths with differing starting technetium species (T4 monoliths with Tc-tricarbonyl gluconate, referred to herein as Tc-Gluc, and T5 monoliths with pertechnetate [TcO_4^-]) suggest that T4 monoliths do in fact leach Tc faster than T5 monoliths and other pertechnetate-bearing

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[1] No post-leaching characterization of the leached monoliths have been performed but ample literature exists that shows carbonation reactions cause calcium carbonate precipitation within concrete and cement structures.

[2] The K_{sp} for aragonite is 6.0×10^{-9} while that of calcite is 3.36×10^{-9} based on reference http://www.solubilityofthings.com/water/ions_solubility/ksp_chart.php
Generally the mineral with the lower K_{SP} will precipitate first and control the solution concentration of the minerals' components; however there are kinetic hindrances for the formation of calcite (the carbonate with the lower K_{SP}) in the Cast Stone leachates so that the more soluble aragonite is observed and controlling the dissolved Ca^{2+} and CO_3^{2-} concentrations in the leachates. Over long times, aragonite will convert to calcite to satisfy thermodynamic driving forces that push geochemical systems to reach their lowest free energy states.

monoliths from the Archive and Extended Leach Suites. The faster Tc leaching from Tc-Gluc (T4) than from monoliths containing pertechnetate is observed in both VZP and DIW eluents. Whether the Tc in eluates from the T4 monoliths remains in a non-pertechnetate form has not been determined because the concentration of total Tc in the T4 monolith eluates is orders of magnitude below concentrations needed for direct speciation determinations for Tc(I) species. The increase in Tc leaching from the T4 monoliths in comparison to pertechnetate leaching from companion T5 monoliths is almost a factor of 10 for DIW and over a factor of 20 for VZP (data from Table 4.8 visually shown in Figure 4.13). At this time, the most probable hypothesis that we have to explain these differences in the Tc leach tendencies is that the BFS converts a significant portion of the starting pertechnetate within the T5 monoliths to a very low solubility, solid Tc-bearing phase[1] that then slowly re-oxidizes with time allowing pertechnetate to slowly diffuse out of the monoliths. Conversely, assuming that the Tc-gluconate and other Tc(I) species present in the T4 monoliths remain soluble in the T4 monolith internal pore water, then these soluble species can diffuse out based on their molecular size with no need for some change in overall redox state within the monolith's interior. This hypothesis is compatible with the observed faster release of Tc from the T4 monoliths. The Tc(I) species in the spike supplied to us included $Tc(CO)_3(gluconate)^{2-}$ and $Tc(CO)_3(H_2O)_2(OH)^0$. Both species have larger sizes than common inorganic species but they are anionic or neutrally charged and if stable and non-sorbing should diffuse out of the porous matrix slightly slower than the mobile anions (nitrate, nitrite, and iodide) or mobile sodium because the Tc(I) species are larger molecules. However, the D_{obs} values for Tc in the T4 monoliths are in the range of 3×10^{-11} to 3×10^{-10} cm^2/s whereas the mobile anions and sodium in the T4 monoliths have D_{obs} values much larger in the range of 2×10^{-9} to 4×10^{-9} cm^2/s. Thus the Tc(I) species are either more reactive (sorb stronger) to the Cast Stone matrix or in fact are being oxidized to pertechnetate faster than the BFS-reduced pertechnetate and diffusing out of the porous matrix. Without the ability to measure the Tc(I) species in the eluates and differentiate them from pertechnetate we can't provide a definitive explanation for the differing Tc leach rates between T4 and T5 monoliths.

One observation that emanates from all the monolith leaching data presented in this report is that duplicate monoliths that come from one specific batch of wet slurry (recall from Section 2.2 that each full batch of Cast Stone wet slurry yielded six monoliths that were cured together under the room temperature high relative humidity) show remarkably good precision in leach tendencies for each constituent that we have been tracking. Figure 5.3 shows an example of the remarkably good precision for the leach tendencies for duplicates from each batch of monoliths. The data show the duplicate monoliths (T4 -1 and T4-2 for Tc-Gluc, T5-1 and T5-2 duplicates for the Tc-pertechnetate batch, and T3-1 and T3-2 duplicates for the Archive Suite T3). This very good precision is not as well manifested for different batches of Cast Stone made with the same composition but prepared in separate batches. For example, when comparing D_{obs} plots for the five replicates (see the Screening Test Matrix shown in Figure A.1) prepared for the screening tests in Westsik et al. (2013a) where the replicate batches were made on different days more scatter is manifested. This observation suggests that making the same Cast Stone mix or composition (at least in the laboratory setting) at different times, even when using the same equipment

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[1] Using the initial pertechnetate loading in Cast Stone monoliths (dry weight basis) shown in Table 4.7 and the wet density of cured monoliths and % solids for Mix 3 and 22 (replicates) in Tables D.4 and D.1, respectively, in Westsik et al. (2013a) and the measured density of the 7.8 M Na Ave simulant [1.336 g/mL], we calculated the concentration of Tc in the internal Cast Stone pore water. Assuming that all the pertechnetate in the simulant remains dissolved in the pore water, the pore water concentration would range between 3.1×10^{-4} and 3.8×10^{-4} mol/L. However, in the reducing environment created by BFS the measured solubility (see Kaplan et al. 2011 and Cantrell and Williams 2013) of various Tc(IV) oxides, the dissolved Tc concentration is limited to $\sim 1.0 \times 10^{-8}$ mol/L. Further this lower concentration agrees well with thermodynamic solubility predictions of various Tc oxides calculated for an assumed pore water composition (Li and Kaplan 2013, SRR CWDA 2014). Thus, in our Cast Stone monoliths, which contain BFS, the majority of the initial pertechnetate in the LAW simulants should be reduced, precipitated, and its release controlled by the re-oxidation of the Tc(IV) oxide precipitates to the soluble pertechnetate species that subsequently diffuses out of the porous matrix.

and mixing procedure, does in fact lead to small differences in the leach properties of the ostensibly same final cured Cast Stone monoliths. More discussion on this topic is found in Westsik et al. (2013a) in Section 8.5. Whether these small differences have any relevancy to full-scale production of large monoliths, or have any potential impact on the contaminant release data selected for future use in IDF predictive modeling, cannot be judged at this time.

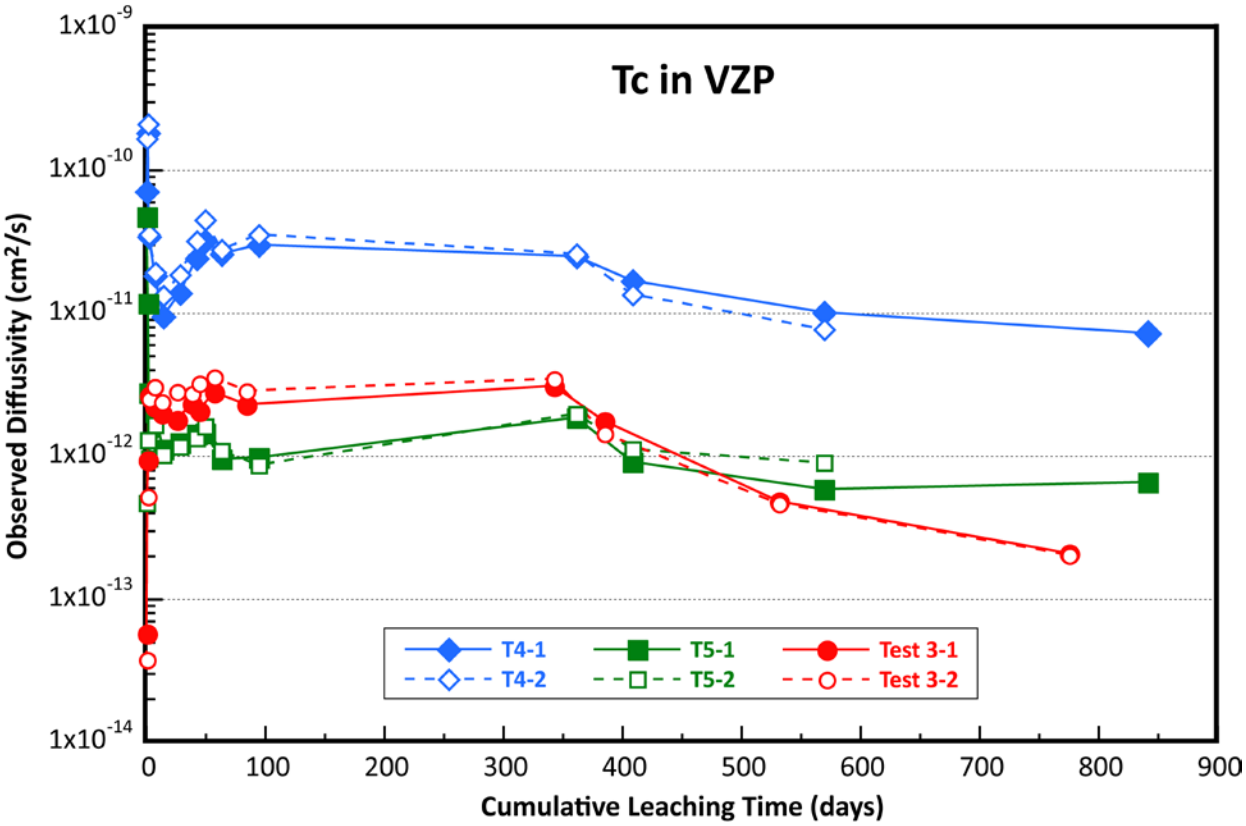


Figure 5.3. Shows Excellent Precision in Tc Leach Tendencies for Duplicate Monoliths from the Same Batches

In February 2015, about half the monoliths in the Extended, Archive and Tc-Gluconate Suites of leach tests were put into “wet” storage in a small volume of their final eluate and many are currently undergoing detailed solid phase (post-leaching) characterization to gain information on the mineralogical changes that occur during leaching, and to estimate the rate of oxygen and carbon dioxide ingress into the monoliths’ interiors, which quantifies the re-oxidation and carbonation processes that impact the release of the key groundwater risk driving contaminants. The post-leached monoliths selected for the first detailed characterization were carefully sectioned into small pieces with known locations as a function of distance from the outer surface using a diamond bladed saw. The sectioning was performed in an anoxic glove bag to minimize exposure to air (re-oxidation concerns) and cutting was performed without water cooling the blade to minimize water leach artifacts. Finally state-of-the-art solid phase characterization instrumentation is being used on pieces with known location to ascertain valence state information on redox sensitive elements (e.g, Tc and Cr) as a function of distance from the monoliths outer surfaces. The goal of all the solid phase characterization activities and eluate analyses will be used to elucidate the weathering reactions caused by the leaching process. All the information is important to improve the technical defensibility of waste form performance in the upcoming 2017 IDF PA.

5.2 Rationale and Resources for Potentially Useful Tests to Study the Leach Properties of Key Constituents

To evaluate contaminant release rates and controlling mechanisms from Cast Stone, or other grouts containing reductants such as BFS, the following testing ideas could prove useful. One key issue that influences the amount of study required to better understand controlling mechanisms for contaminant release from Cast Stone/grout is how certain and detailed the conceptual release model must be to perform technically defensible long-term predictions. That is, if the inventory of contaminants solidified in the Cast Stone/grout and/or the release rates of key risk driving contaminants is low, such that impacts to groundwater are slight, then available data and simple release conceptual models, such as the empirical observed diffusion coefficient, might be sufficient. However, if inventories in the Cast Stone and release rates for key contaminants are larger such that predicted impacts to groundwater, the accessible environment, and human health are estimated to be close to or above acceptable limits then more studies, especially those that elucidate the controlling release mechanism are necessary.

Several documents have been published in the last five years that:

1. Identify technical risks and uncertainties associated with the Cast Stone waste immobilization process at Hanford. Namely, science and technology gaps were identified, by a joint team of PNNL and SRNL scientists (see Brown et al. 2014), in areas such as conducting PAs and risk assessments of Cast Stone waste form and disposal system performance.

2. Present an iterative hierarchical total system performance assessment (TSPA) modeling strategy that can be illustrated as a pyramid with a high-level, stochastic TSPA modeling for decision-making at the top, supported by more mechanistically detailed deterministic “process” models below, which in turn are supported on a base of previous knowledge, chemical-physical theories, and new information from characterization and experimental studies (see Yabusaki et al. 2015).

3. Delineate detailed plans on the types of laboratory and field experiments that should be considered to provide the technical basis for understanding the long-term performance of Cast Stone/grout waste forms in the IDF disposal environment (see Westsik and Serne 2012 for test plans for Cast Stone solidified secondary wastes and Westsik et al. 2013b for test plans for using Cast Stone as a supplemental waste form for LAW liquid wastes). Both of these plans describe testing to show Cast Stone/grout waste forms can comply with existing or likely IDF waste form specifications and acceptance criteria. The proposed experiments/tests also include some activities to demonstrate that the Cast Stone immobilization process can be controlled to consistently provide an acceptable waste form product. All the proposed activities in these two plans would provide data on waste form performance that potentially will be needed to support the IDF PA analyses of the long-term environmental impact of Cast Stone/grout disposed of in the IDF. Further, Yabusaki et al. (2015) presents pertinent test methodologies in its Section 5.

As mentioned, the amount of additional studies required to better understand controlling mechanisms for contaminant release from Cast Stone/grout is dependent on how certain and detailed the conceptual release model must be to perform technically defensible long-term predictions. If the inventory of key risk driving contaminants and their release are deemed problematic, then a more robust understanding of contaminant release will be required. A key fact is that there is a large contrast in the physical and chemical properties of the Cast Stone waste form versus the IDF backfill and surrounding sediments. Freshly made Cast Stone exhibits low permeability, high tortuosity, low carbonate content, high pH, and low Eh attributes, whereas the backfill and native sediments have high permeability, low tortuosity, high carbonate content, circumneutral pH, and high Eh. These contrasts have important implications for flow, transport, and reactions across the Cast Stone – backfill interface. Over time, with transport across the interface and subsequent reactions, the sharp geochemical contrast will blur and there will be a range of spatially-distributed local equilibrium conditions. In general, the COC's mobility and transport will be sensitive to these geochemical variations, which also include physical changes in porosity and permeability from mineral dissolution and precipitation reactions. Therefore, the effectiveness of Cast Stone as a barrier to COC release is expected to evolve over the lifetime of the IDF. The technical approach to determining PA modeling parameters should therefore consider processes, properties, and conditions that alter the physical and chemical controls on COC transport in Cast Stone and the IDF subsurface environment over long time frames (1,000 to >10,000 years) and large length scales (~100s of meters). One imposing fact is that current data and understanding of Cast Stone behavior come from relatively short-term, small-scale experiments. Thus, an important role and challenge for IDF PA modeling is to provide a mechanistic link between these short-term and small-scale laboratory and field tests, and the prediction of Cast Stone performance over the cited repository time frames and length scales. An important goal of the modeling strategy is to demonstrate that performance is acceptable when the relevant uncertainties affecting performance have been appropriately accounted for.

As mentioned, most organizations responsible for disposal facility operation and their regulators support an iterative hierarchical TSPA modeling strategy. The strategy is conceptually represented as a pyramid (see Figure 5.4) with the high-level, stochastic TSPA modeling for decision-making at the top, supported by more mechanistically detailed deterministic "process" models below, which in turn are supported by a base of chemical and physical theories and information from site-specific- characterization and experimental studies. The chemistry of contaminant release and physics of different phases (water, gas, and solute) flow in high-level TSPA-type modeling is often abstracted to a set of COC-specific empirical coefficients (e.g., desorption K_ds, observed diffusion coefficients, and/or solubility limits). These simpler lumped parameter models are computationally efficient, allowing comprehensive coverage of the system features, as well as stochastic treatment of uncertainties in parameters, process models, and conditions. However, TSPA models are not mechanistic. This means that these lumped, single-parameter models are not sufficiently robust to address the spatial and temporal variation in geochemical processes, properties, and conditions that control COC and macro components behavior.

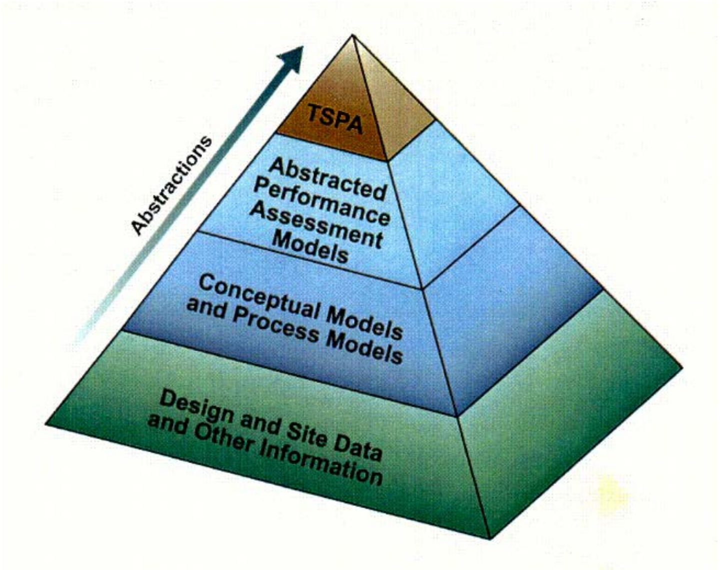


Figure 5.4. Total System Performance Assessment Information Pyramid (from TRW 2000)

The hierarchical modeling strategy is intended to address limitations in the simplified (abstracted) models used in the high-level TSPA simulator. In this strategy, detailed property distributions and mechanistic process models are used to provide a technically defensible basis for assessing the level of rigor that is ultimately included in the PA modeling. In this way, the sensitivity of the estimated risk to simplifications and/or omissions of specific process models can be directly evaluated.

If decision makers at Hanford decide that a more mechanistic understanding of the release of COCs would be beneficial to improve technical defensibility and lessen uncertainties, experiments described in Westsik and Serne (2012), Westsik et al. (2013b), and Yabusaki et al. (2015) are recommended. Some of these test methods are briefly described in the next subsection.

Other investigations that consider 1) gas-phase flow and transport, 2) fracture/matrix flow for cracked Cast Stone, 3) multi-component solubility, redox, aqueous and surface complexation reactions for contaminants' release, and 4) feedback to physical properties from mineral precipitation and dissolution should also be considered.

5.3 Thoughts to Consider Before Selecting Test Methodologies

First, it is important to remember that there needs to be continual interaction between the experimentalists and the modeling team that will use the data in future IDF PA activities. If the experiments produce data that cannot be readily used to develop conceptual or mathematical models that directly support the IDF system PA simulators (computer codes used to calculate COC release and subsequent transport to regulatory points of compliance), little is gained. This guiding principle, the need for continual interaction between the experimentalists and PA modeling team, is often acknowledged but much less often put into practice. We therefore stress that attention and diligence is required by all participants in the 2017 IDF PA effort to keep the experimentalists and modelers "on the same page."

The following discussion addresses some of the potentially useful experimental methodologies that could be used to improve the data base for contaminant release from Cast Stone/grout and to improve our knowledge of controlling mechanisms for such processes as Cast Stone weathering in the IDF subsurface environment. Some of the key controlling processes likely include:

1. Re-oxidation of redox sensitive COCs such as Tc and Cr in Cast Stone via O_2 ingress.

2. Carbonation of Cast Stone surfaces and interior by CO_2 in air and dissolved bicarbonate in pore water.

3. Weathering induced changes in porosity and permeability that lead to cracking or alternatively waste form surface armoring and reduction in internal porosity/permeability.

4. Solubility/dissolution, or adsorption/desorption, or paragenetic mineral transformation instead of diffusion as process controlling COC sequestration/release.

These four potential controlling COC release mechanisms are influenced by the materials that surround the Cast Stone waste form, such as the infiltrating recharge water and air, metal container, backfill, proximal Hanford formation vadose zone sediments, and potentially eluates from other nearby waste forms, such as immobilized low-activity waste (ILAW) glass. Thus, some experimental tests should either include other materials besides the Cast Stone/grout itself or include fluids that have been equilibrated with the other materials. If the former approach is used, these tests are called multi-component tests. Generally, Cast Stone/grout contaminant release testing emphasizes detailed time-dependent characterization of eluates that have contacted the solid waste form (e.g., all the EPA Method 1315 eluates discussed in the bulk of this report). However, detailed characterization of the pre-leached and post-leached Cast Stone solids is also necessary to elucidate controlling processes. Solid-phase characterization is required to evaluate changes in mineralogy, porosity, permeability, pore size distribution and shape, tortuosity, and distribution and oxidation state of key COCs. Many of these solid-phase attributes need to be measured as a function of distance from the outer surface of the monolith, as well as time of exposure to leaching fluids (both aqueous and gaseous).

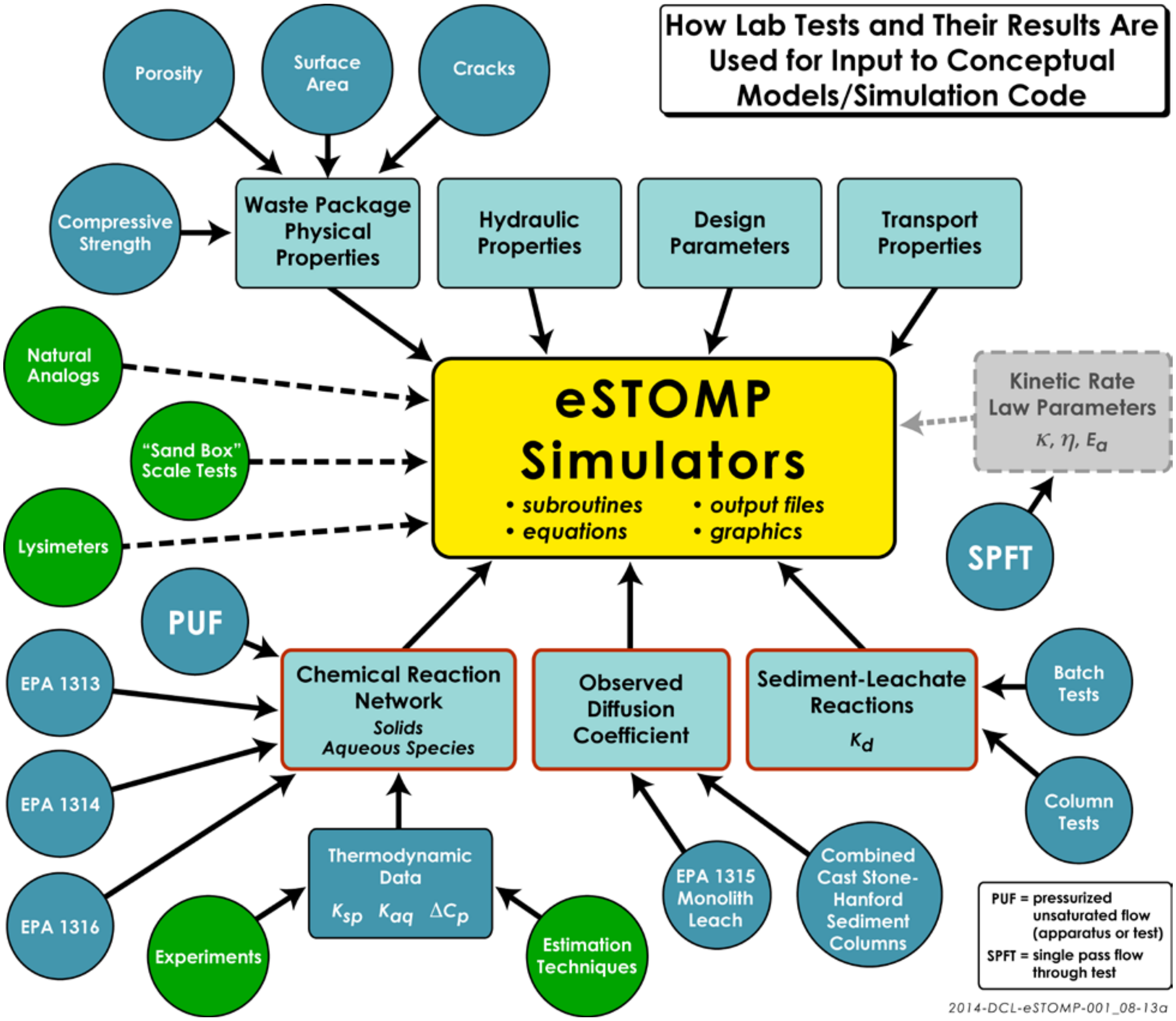
A list of potentially useful solid-phase characterization techniques (again as a function of position from the monolith surface and versus weathering/leaching time) follows, and more details are available as Appendix C in Westsik et al. (2013a):

- mineralogy using bulk powder XRD and micro-XRD of selected particles,
- mineralogy and particle size/shape using scanning and transmission electron microscopy (SEM and TEM) equipped with chemical microprobe energy-dispersive X-ray fluorescence,
- porosity and pore structure (size distribution and pore throat shapes and sizes) and crack numbers and sizes using X-ray and neutron micro-tomography, Hg intrusion porosimetry, N_2 and/or Ar gas adsorption using Brunauer-Emmett-Teller (BET) methodology, and SEM,
- valence state and nearest neighbor atoms determination for COCs using synchrotron X-ray absorption spectroscopy (XAS) (X-ray absorption near edge structure [XANES] and extended X-ray absorption fine structure [EXAFS]) spectroscopy. X-ray photoelectron spectroscopy [XPS] is another surface technique that can provide valence state information for some elements but preliminary attempts to characterize the surfaces of Cast Stone for Tc, have been unsuccessful because of the large Na signal that swamps out the region where Tc spectra would be found.
- redox capacity of individual dry materials and pre- and post-leached Cast Stone waste forms,
- specific surface area using N2 and/or Ar gas adsorption (BET), and
- bulk chemical composition of pre- and post-leached Cast Stone using various solid digestion techniques followed by digestate analysis by ICP, ICP-MS, IC, and perhaps radio-counting.

A key observation from all the Cast Stone solid characterization studies reviewed to date that address mineralogy changes, valence state changes, and identification of redox and carbonation front penetration distances (e.g., Um et al. 2011a, 2013; Langton and Almond 2013; Langton 2014) is that the sample preparation for analysis cannot unequivocally prove that preparation steps did not alter the state of the Cast Stone prior to analysis. For the oxidation front and valence state determinations, the ability to identify the exact location (in relationship to the monolith outer surface) of the disaggregated subsamples (necessary to perform the characterization) needs to be improved. That is, many of the measurements have been made on disaggregated particles (generally further crushed to powders) for which meticulous and consistent efforts were **not** made to identify their exact location in relationship to the monolith's original surface. This comment is not a condemnation, but rather, an honest evaluation of the difficulty of manipulating monolithic solid samples into disaggregated forms amendable for characterization without causing alterations in desired properties or losing accurate knowledge of location. Careful sectioning of Cast Stone monoliths into circular thin disks and further perpendicular cutting of the disks into smaller pieces all within an anoxic glove bag has been performed in early CY2016. The sawing was performed by "dry" cutting techniques to minimize leaching of constituents. The carefully cut pieces with known locations from the monolith's outer surface are currently undergoing solid phase characterization using techniques described above. Solid phase characterization results will be published in a report by the end of CY2016. The key point is that more effort must be used to minimize these problems in future solid-phase characterization activities. If successful, the solid-phase characterization information as a function of location from the monolith outer surfaces can better address re-oxidation and carbonation front penetration rates. An understanding of these two processes and their rates is needed to better understand Cast Stone long-term weathering to improve the technical defensibility of future IDF PAs.

5.4 Potential Methodologies/Experiments to Measure COC Release from Cast Stone/Grouts

This section briefly describes test methodologies and experiments that could improve our understanding of the release of contaminants from Cast Stone/grouts. Many of the previous cited plans and the approach document provide more details on the test methods and additional discussion on how the data generated can be used to improve the technical robustness of the IDF PA. Figure 5.5 is a logic/flow diagram that attempts to show how data from the various laboratory and field tests/experiments can be used to directly feed input values or information into both conceptual and numerical process models that constitute a TSPA. This figure, from Westsik et al. (2013b), assumes a TSPA based on eSTOMP codes/simulators, although QA/QC validated a second suite of simulators/codes available from the Cementitious Barriers Partnership (CBP) that focuses specifically on cementitious waste as an alternative. The simulator Subsurface Transport Over Multiple Phases (eSTOMP) is a very robust detailed process code that can accommodate many algorithms that can calculate flow of various fluids, complex chemical reaction networks, and transport of COCs through subsurface media. eSTOMP capabilities include 3-D, variably saturated liquid and gas flow, dual porosity modeling for fractures and the porous matrix, and multi-component reactive chemical transport with feedback to physical properties (e.g., porosity, permeability, tortuosity) caused by changes in mineral volume fractions (e.g., precipitation of secondary minerals and dissolution of primary minerals). Two current limitations of eSTOMP include that it is a continuum-based model that assumes all solid phase media are porous or can be modeled as a porous equivalent media and it is not specifically constructed to address cementitious media and their complex physical, chemical, and structural properties or reactions. However, after adding cement-specific algorithms, input parameters for the algorithms, and appropriate boundary conditions, eSTOMP has recently been applied to predicting the release of contaminants from cracked cement that includes many processes, properties, and conditions relevant to Cast Stone weathering, such as evolution of cement mineralogy in contact with high pH pore fluids, cement degradation from decalcification reactions, and the impact of cracking on the degradation process. Freedman et al. (2016) discuss in greater detail how eSTOMP predictions of cement weathering compared with other simulators used (Perko et al. 2015). More detailed descriptions of eSTOMP attributes are found in Freedman et al. (2016) and Yabusaki et al. (2015) and references therein.



2014-DCL-eSTOMP-001_08-13a

Figure 5.5. Flow/Logic Diagram Showing How Data from Test Methods/Experiments Feed Computer Codes

In this subsection, we will focus our discussion on the blue and green circles in Figure 5.5 that directly address COC release from Cast Stone/grout. The figure shows traditional laboratory-scale leach tests such as the EPA Method 131X (where X = 3,4,5,6), wherein Cast Stone monoliths or crushed material is contacted with an aqueous solution under water saturated conditions, either in batch or flow-through mode. Dependent on which EPA test is used, the goal is to gather data on the release of key contaminants and major components of the Cast Stone waste form. Method 1315 directly provides observed diffusion coefficient values and constituent fluxes over specific sampling intervals or over cumulative time. This type of information can be used to parameterize empirical diffusion-controlled conceptual release models, and was used in all the testing described in this report. Additional extended leaching tests beyond the conventional 63-day leaching period using a VZP eluent are recommended for future Cast Stone/grouts studies. One variant of the EPA Method 1315 would be to slowly and continuously bubble moist air into the VZP eluent to maintain constant atmospheric concentrations of oxygen and carbon dioxide in the eluates. As long as this air flow-through approach does not cause eluate evaporation issues, the results of these tests could be compared with the results of the "standard" EPA Method 1315 tests to see if there are changes in leach tendencies of the redox sensitive Cr and Tc, and to see whether there is additional build-up of the white precipitate. Another improvement when using the EPA Method 1315 test methodology would be to use larger monoliths such that the cumulative fraction leached for the mobile constituents (I, nitrate, nitrite, and sodium) does not exceed 20% before the traditional 63-day test is completed. This would avoid the violation of the semi-infinite source boundary condition. However, using larger monoliths would require using larger volumes of eluent that after analysis becomes a radiological waste, and if one uses a VZP simulant, then large quantities of chemical reagents are consumed.

Traditional EPA Method 1315 leach testing with both DIW and VZP eluents have been started on Cast Stone or grout monoliths containing Tc and I getters. Comparison of leaching results for monoliths with and without getters for these two groundwater risk drivers will be informative. Preliminary studies of getters are documented in Um et al. (2011b), Neeway et al. (2014), and Qafoku et al. (2015).

EPA Method 1313 uses granular (disaggregated) Cast Stone and contacts the crushed material with DIW buffered to a range of specific pH values until equilibrium (more accurately steady-state) solute concentrations are observed. From the steady-state concentrations as a function of pH, one can address whether solubility-precipitation processes might be controlling contaminant release over some pH range. The EPA Method 1314 test uses granular (disaggregated) Cast Stone in a flow-through column configuration. The packed column is then contacted by a leaching solution at a low flow rate under water saturated conditions. Note however, with modifications, unsaturated flow columns could be prepared in addition to saturated flow. Effluent concentrations and cumulative mass releases of COCs are plotted as a function of cumulative liquid-to-solid (L/S) ratio (or time) to predict COC release directly in one dimension as a function of time. This would provide directly an empirical one-dimensional (1D) flux value as a function of time, or alternatively, pore volumes passing through or by the waste form. The EPA Method 1314 effluent concentrations can also be used along with geochemical speciation modeling to infer the mineral phases that control the release of key constituents from the Cast Stone waste form.

The EPA Method 1316 uses disaggregated Cast Stone in batch mode contacting an aqueous solution at the natural pH of the solid material. The method varies the L/S; generally, five values that range from 0.5 to 10 mL/g-dry solid are chosen, under conditions that approach chemical equilibrium. The results of this test are used to show changes in equilibrium eluate concentrations as L/S approaches that of field conditions and to estimate internal pore water concentrations at low L/S. Generally, experimentalists prefer to use the EPA Method 1314 over Method 1316 to gather the same type of data.

One key issue in using this suite of EPA Methods to study Cast Stone/ETF-processed secondary waste grout is the fact that the IDF burial environment exists in a highly water unsaturated state, while the EPA tests are conducted in fully saturated conditions. There is concern that contacting intact Cast Stone monoliths or disaggregated materials under water saturated conditions may not allow as much re-oxidation and carbonation reaction to take place in a given time period as would be available for unsaturated conditions where gaseous O_2 and CO_2 in air exist at relatively higher concentrations and can diffuse faster into low permeability monoliths than dissolved O_2 and CO_2 (bicarbonate-carbonate) in contacting solution. Langton (2014) suggests that Cr and Tc re-oxidation occurs faster in monoliths buried in Hanford sediment kept under unsaturated conditions than monoliths held in water saturated conditions. On the other hand, studies reviewed in Golovich et al. (2014) suggest that the observed diffusion coefficient (D_{obs}) for contaminants such as Tc(VII) and iodide leaching from spiked concrete into unsaturated Hanford sediments are several orders of magnitude lower than D_{obs} values for the same COCs present in other cementitious waste forms that are leached in the traditional water saturated conditions. In a similar fashion, Golovich et al. (2014) reviewed studies where COCs were spiked into Hanford sediments that were contacted with uncontaminated Hanford sediments in half-cell diffusion tests. In these tests, the diffusion of the COCs from the spiked sediment cell into the uncontaminated sediments was systematically lower as the moisture content in the two sediment filled cells was reduced from saturated conditions down to 7 wt% and then 4 wt%. These finding were quoted in Section 7.5.2.8 of the TC&WM EIS (DOE 2012) as follows:

> ‘Data suggest that grout surrounded by soil with a lower moisture content would lead to a corresponding decrease in the diffusivity of concrete for grouted waste forms, and thus a better-performing waste form with slower release rates (Mattigod et al. 2001). Figure 7–24 *(in TC&WM EIS)* reanalyzes the data for the grout sensitivity case (i.e., 7 wt% percent moisture content). The results suggest that the sensitivity grout would perform substantially better—almost two orders of magnitude better for all grouted waste forms—and thus would likely lead to much lower concentrations in groundwater at the Core Zone Boundary for onsite sources of waste disposed of in an IDF. At an infiltration rate of 3.5 millimeters per year, lowering the diffusivity for grout by two orders of magnitude (i.e., from 1.00×10^{-10} to 1.00×10^{-12} square centimeters per second) would decrease the contribution of ETF-generated secondary waste by a factor of 100, thus deleting this waste from the list of dominant contributors to risk. Similar results were predicted for simulations under Tank Closure Alternatives 3A, 3B, and 3C, as discussed in Appendix M, Section M.5.7.5.’

Besides the classical half-cell diffusion cell technique described in Golovich et al (2014) and the primary references cited within, one methodology/instrumentation that allows controlled unsaturated conditions to be maintained is the pressurized unsaturated flow (PUF) system (see Westsik et al. 2013b Figure 4.8 and Section 4.10.5 for more details). PUF tests on Cast Stone/ETF-processed grouts can use either intact monoliths or disaggregated material. In addition, PUF tests can be run at controlled higher temperatures that might be a method of accelerating the weathering reactions. Recall that the ASTM C-1308 monolith leach test methodology (very similar to EPA Method 1315) recommends leach testing at several elevated temperatures as a way of accelerating the diffusion process and quantifying the effects of temperature on effective (observed) diffusion coefficients. Also, recall that the steady-state ambient temperature of the IDF facility, once it is closed, is ~15 °C, a value lower than that used in all the lab-scale leach testing to date. Thus, a temperature correction could be made on all the available empirical observed diffusion coefficients, D_{obs}. There are some practical limitations to using the PUF method to study Cast Stone materials, such as cost of the apparatus, complicated operating details to maintain controlled and constant conditions, required long times to gather adequate data, and potential interactions of the pressure-plate frit (used to maintain the unsaturated conditions) with the effluent solution and any suspended solids (or precipitates) in the eluates. Interactions include adsorption of trace constituents in the eluates thus altering flux rates from their true values and complete frit plugging by solids that stop the collection of effluent.

Less expensive Wierenga-style unsaturated column devices (Wierenga et al. 1993; Horton et al. 1982; http://www.soilmeasurement.com/flow-cells.html) and the hanging column technique (Rod et al. 2010; Chang et al. 2011) are options for performing unsaturated soil column tests (see ASTM 2008b Method D6836) for the wetter end of unsaturation, but both of these test configurations also use a ceramic or plastic pressure plates at the bottom of the packed columns to minimize air entry that can interfere with maintaining the constant suction (controls the water saturation) on the packed column. The Wierenga-style and hanging water unsaturated column devices are also less suited for higher temperature experiments.

On the right side of Figure 5.5 is a blue circle (constituting experiments/test methodologies) labeled “Combined Cast Stone-Hanford Sediment Columns” that feeds data and information to a process model (see Figure 5.5) that is a detailed simulator that attempts to shed light on controlling mechanisms for contaminant release from Cast Stone/grout. Another more generic name for this blue circle is “multi-component tests,” wherein one or more of the materials surrounding the Cast Stone waste form are present, along with the Cast Stone, in an experiment that attempts to quantify constituent releases. Multi-component tests evaluate the interactions between Cast Stone waste packages (Cast Stone in its mild steel container) and the near-field backfill, surrounding sediment, and recharge pore water. As mentioned earlier, it is possible that the IDF burial environment might allow leachates from other types of waste forms, such as ILAW glass, to alter the chemical composition of the recharge pore water prior to interacting with the Cast Stone waste package, backfill, and near-field sediments. The key point of performing multi-component tests is to capture the interactions among the various components with the Cast Stone. That is, chemical reactions solely between Cast Stone and vadose zone recharge water may not give an accurate depiction of release from the entire near-field environment, whose release is transported through the remaining un-impacted vadose zone to the water table, and then to the "accessible" environment/regulatory points of compliance.[1] For example, Ojovan et al. (2011) compared the leach rates of ^{137}Cs out of a cement waste form made with a sodium nitrate dominated salt solution spiked with trace amounts of ^{137}Cs. Small cement monoliths (2.8 mm diameter by 2.8 mm length) were leached in DIW using a Russian leach test similar to ASTM-1308 at room temperature. In a companion field test, larger monoliths (24 cm diameter by 24 cm length) of the same cement waste form were buried under water saturated conditions in a shallow field lysimeter. Three of the larger monoliths were buried at a depth of 1.7 m (below the freeze zone) within a coarse sand backfill. The larger monoliths and backfill were contained in a stainless steel tray. An extraction pipe was situated in the coarse sand to extract leachate. The tray containing the cement waste forms and coarse sand backfill were covered by local loamy surface soil. About twice a month for 12 years, leachate was pumped out of the waste form tray. Meteorological observations on the site showed an average temperature of 6.5 ºC and 557 mm of precipitation per year. The normalized ^{137}Cs leach rate (g/cm^2 day) was calculated for both the laboratory leach test and field lysimeter. The results showed that the average annual ^{137}Cs leach rate in deionized water was about 35 times greater for the laboratory specimens compared to the first year leach rate for the field test. The cumulative fraction of ^{137}Cs leached from the lab monoliths (3.74%) was similar to values reported in the literature for similar lab experiments. However, the cumulative fraction of ^{137}Cs released in the first year of the field lysimeter test was only ~0.01%. Therefore, the authors concluded that to compare field and laboratory test results, several scaling factors are required in order to account for waste form surface to volume, temperature, and leachant type (DIW vs site groundwater differences). Although this study may not be directly relevant to Cast Stone/secondary waste grout disposal in IDF, it does show that multi-component testing under relevant subsurface burial conditions can yield significantly different calculated release rates for COCs than simple waste form leach tests such as those described herein.

Our report discusses "binary" leach tests between Cast Stone and two simple eluents, but other leach tests that contain other near-field materials would improve the available database and likely identify the significance of their interactions with Cast Stone/secondary waste grouts. Multi-component tests are most amenable to using the flow-through column approach, but batch tests with various components are not precluded. In the column set up, intact Cast Stone monoliths can be packed within backfill or Hanford formation sediments in a geometry that simulates actual IDF burial design, or a disaggregated layer of Cast Stone can be placed between layers of backfill and/or sediment in proportions that simulate the vertical dimensions of the IDF burial design. Figure 5.6 is a schematic of some multi-component testing performed in the past using Hanford Grout (Serne et al. 1987). One could also surround the Cast Stone with thin pieces of or crushed mild steel to represent the container. To date, inclusion of the steel container has not been done based on predictions that its corrosion occurs relatively quickly in comparison to disposal time frames, and that the mass and thickness of mild steel is insignificant in comparison to the Cast Stone, backfill, and surrounding sediments.

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[1] Note herein we assume that the key pathway dominating future risk for contaminants leached from Cast Stone/grout buried in IDF is groundwater, as shown by all past Hanford Site IDF PAs (see discussions in Section 3.1.1 in Yabusaki et al. 2015).

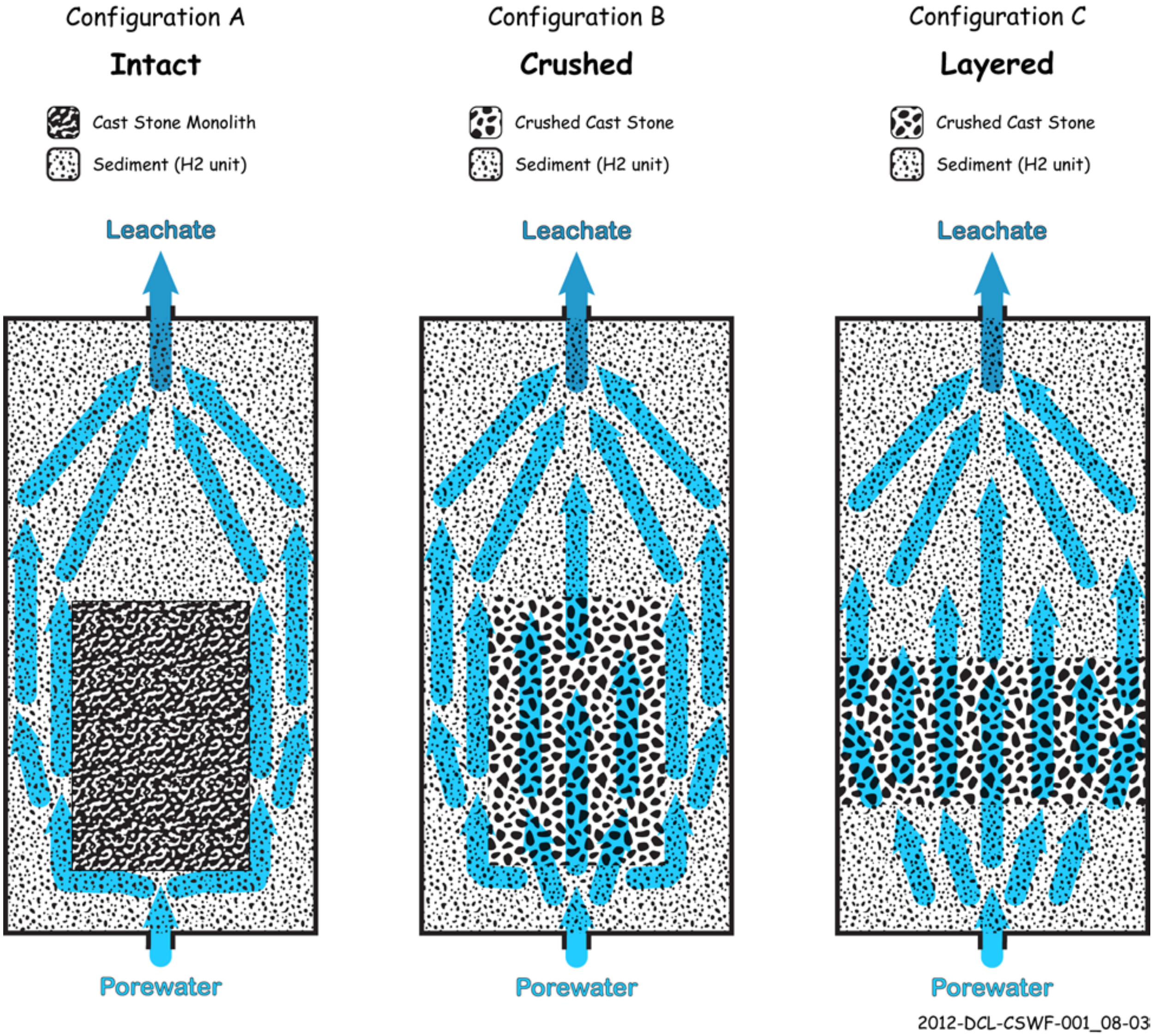

Figure 5.6. Schematic of Multi-Component Flow-Through Column Tests

Lab methods/tests that would be most practical for studying multi-component interactions for the IDF burial environment include unsaturated column tests (using the hanging water column technique or the PUF system previously mentioned).

On the left-hand side of Figure 5.5 is a green circle that represents experiments on single-phase minerals that might be needed to bolster the multi-component reaction networks used in the detailed process models. The reaction network is a system of relevant chemical reactions, stability constants, and associated rate laws that can be solved with a multi-component geochemical speciation model to calculate the compositions of solid phases consistent with their associated pore solution. For example, a multi-component reaction network for Cast Stone and its COCs will require stoichiometry and thermodynamics for the important mineral phases (and perhaps sorption sites) that describe Cast Stone solid phases, COCs, products from the oxidation and corrosion of the mild steel waste containers, and the backfill materials. In the reaction network approach, the behavior of the individual solution components is predicted using elementary reactions based on the law of mass action. Kinetics can be included through simplified but thermodynamically consistent rate laws. The benefit of the multi-component reaction network is that it can potentially predict the evolving geochemical controls that lead to the observed pore water chemistry and concentrations of both major and minor constituents for varying locations and times in Cast Stone near-field environment or the entire IDF subsurface. The challenge is to characterize not only the reactions, thermodynamics, and rates directly controlling COC mobility and transport, but also the auxiliary reaction processes not involving COCs that significantly affect the direct controls (e.g., pH, Eh, porosity, macro solute composition). Thermodynamic databases provide reference reactions, stoichiometry, and temperature-dependent stability constants that are used to build the reaction networks for predicting equilibrium species and mineral phase assemblages in Cast Stone even without experimental data support. The solid-phase assemblage is for the most part responsible for the mechanical, hydraulic transport, and chemical properties of the Cast Stone. Extensive thermodynamic databases exist, but they are not exhaustive; in particular, gaps in the thermodynamic data for materials like fly ash, BFS, and degradation-produced materials need to be determined or defensibly estimated and validated, as well as reactions involving the key COCs.

The experiments used to study single-phase minerals are generally batch solubility tests where pure single-phase minerals are dissolved in aqueous solutions with controlled pH-Eh and chemical composition until a steady-state (equilibrium) condition is reached that represents the solubility of the mineral, quantified by its stability constant or solubility product (K_{sp}). Similar batch adsorption-desorption tests can be performed with single pure phase adsorbents and trace COC-spiked solutions that use controlled pH-Eh and macro constituent concentrations with only the COC concentration varied and measured until equilibrium conditions are reached, at which time sorption-desorption K_d functions can be quantified.

These types of batch solubility or adsorption-desorption tests have recently also been performed on Cast Stone (not a pure single-phase mineral) to generate more empirical solubility or adsorption-desorption relationships for key COCs. The test methods and how the raw data are processed to get the empirical solubility and K_d values are found in Kaplan et al. (2011), Li and Kaplan (2013), Cantrell and Williams (2012), Estes et al. (2012), Almond et al. (2012b) and Um et al. (2016). These empirical solubility and sorption-desorption approaches have been used by the Savannah River Site (SRS) to perform Saltstone PAs. A brief summary of the SRS PA contaminant release modeling and these empirical conceptual model approaches is found in Yabusaki et al. (2015) in Section 3.1.2. One key point to consider is that the test methods described in the SRNL reports may need to be slightly modified to accommodate IDF site-specific conditions, and more certainly, the results of testing Hanford sediments for COC K_d values and/or empirical solubility values will likely differ from values measured at SRNL for SRS conditions (see for example, Almond et al. 2012b; Estes et al. 2013; Kaplan et al. 2011; and, Li and Kaplan (2013).

Also shown in Figure 5.5 are experiments/test methods to quantify the sorption-desorption of leached COCs (from the Cast Stone waste package) onto the underlying vadose zone and aquifer sediments, which is the dominant process that further retards the migration of some leached contaminants. The test methods used are the same type of batch and flow-through column tests already described for quantifying COC release from Cast Stone or the multi-component Cast Stone waste packages contacting near-field materials. Such sorption-desorption tests using simulated Cast Stone leachates, simulated LAW glass leachates, and native VZP that are all spiked with the key risk drivers (Tc, iodide, and Cr(VI)) were recently performed at PNNL under funding from WRPS (see Last et al. 2015).

Finally on the right-hand side of Figure 5.5 are three green circles labeled sand box scale, lysimeter tests, and natural analogs representing larger-scale experiments or long time field observations. The former two can be considered larger-scale multi-component tests, and the natural analogs are field-scale multi-component tests of existing natural occurrences. All three of these types of tests are used to “validate” the more common short-term lab-scale tests. All three can require large time and resource commitments, especially for the slowly reacting Hanford vadose zone where recharge rates through engineered surface covers can be < 1 mm/yr. Should such larger-scale tests be considered, they need to start as soon as possible, and sustained funding commitments need to be on the order of decades.

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Appendix A

Supporting Information

Appendix A

Supporting Information

This appendix contains supporting information, including the original Screening Test Matrix and XRD spectra of precipitates formed in VZP simulant and VZP eluates after contact with Cast Stone monoliths, curing times for all the monoliths used, and a comparison of the measured concentrations of the macro constituents in the VZP eluent with its theoretical composition based on chemical reagents added.

	Fly Ash Source / Blast Furnace Slag Source (Northwest or Southeast USA)							
Waste Composition	NW/NW	NW/SE	SE/NW	SE/SE	NW/NW	NW/SE	SE/NW	SE/SE
Average 5M		20						13 2
High SO_4 5M			1		10			
High Al 5M		4			24			17
SST Blend 5M			8			12		
Average 7.8M	5		6			3 22		
High SO_4 7.8M	15 25			14 7			26 21	
High Al 7.8M			19			9	11	
SST Blend 7.8M	16			18			23	
Mix Ratio (w/db)	0.4	0.4	0.4	0.4	0.6	0.6	0.6	0.6

🟦 **Mix # -no replicates**

🟩 **Mix # -Replicates**

Figure A.1. Screening Test Matrix

Table A.1. Preparation Notes on Making New 7.8 M Na Average Simulant (4.5 L)

Compound	Amt Added (g)	Observations
DIW	**2800 (mLs)**	Starting volume in large beaker; chemicals added in order listed
$Al(NO_3)_3 \bullet 9H_2O$	**807.93**	Dissolved OK and volume increased from 2800 to 3200 mL
KNO_3	**23.247**	Dissolved OK and volume increased from to 3250 mL
$NaNO_2$	**273.61**	Bubbled vigorously and soon brown nitrous oxide fumes were escaping
$NaNO_3$	**399.141**	Bubbled vigorously and soon brown nitrous oxide fumes were escaping but then settled down and solution cleared a lot with a faint yellow color
$Na_3PO_4 \bullet 12H_2O$	**131.292**	Solids not dissolving, cloudy appearance with some brown nitrous oxide fumes
Na_2SO_4	**85.266**	Solids not dissolving, cloudy appearance with some brown nitrous oxide fumes
Na_2CO_3	**203.98**	Solids not dissolving, cloudy appearance with some brown nitrous oxide fumes when solution stirred vigorously; definite white precipitate particles present
NaF	**9.316**	Solids not dissolving, cloudy appearance with some brown nitrous oxide fumes when solution stirred vigorously; definite white precipitate particles present
NaCl	**17.307**	Lots of white foam floating on surface, light crust formed; lots of white precipitate, no nitrous oxide fumes on stirring
NaOH (50% soln)	**1565.15**	Solutions cleared up; no white foam floating on surface; few precipitates noticeable
$NaC_2H_3O_2$	**22.084**	Seemed to dissolve OK
$Na_2Cr_2O_7 \bullet 2H_2O$	**22.317**	All dissolved and imparted yellow "urine" color
$Pb(NO_3)_2$	**0.595**	So little solid added that hard to tell if it totally dissolve; a few whitish specks/particles were observed in the stirred solution
$Ni(NO_3)_2 \bullet 6H_2O$	**6.7**	Obvious green particles not dissolving
$Cd(NO_3)_2 \bullet 4H_2O$	**0.341**	So little mass added could not tell if all Cd salt dissolved; slurry dominated by green nickel particles

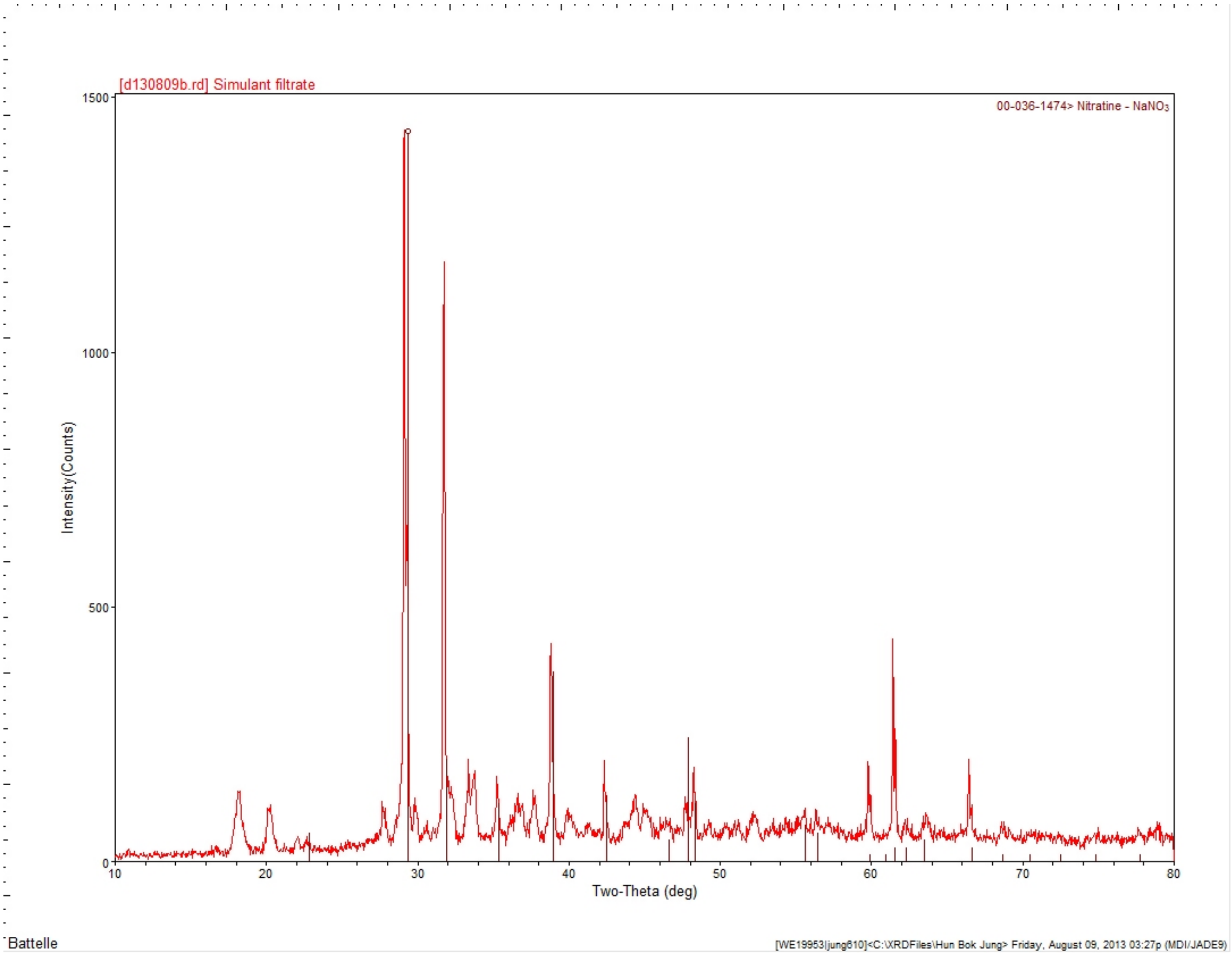


Figure A.2. XRD Spectra for Suspended Solids in 7.8 M Na Average Simulant-New Batch June 2013

Table A.2. Curing Times for all LAW Cast Stone Monoliths

Mix #	Made	Extended Leach Started	Cure Time (days)	Archived Leach Started	Cure Time (days)
1	12/11/2012	1/9/2013	29		
2	12/11/2012	1/9/2013	29		
3	12/7/2012	1/9/2013	33	7/9/2013	214
4	12/10/2012	1/9/2013	30		
5	12/11/2012	1/9/2013	29	7/9/2013	210
6	12/11/2012	1/9/2013	29		
7	12/11/2012	1/9/2013	29		
8	12/7/2012	1/9/2013	33	7/9/2013	214
9	12/11/2012	1/9/2013	29		
10	12/11/2012	1/9/2013	29	7/9/2013	210
11	12/11/2012	1/9/2013	29		
12	12/11/2012	1/9/2013	29		
13	12/10/2012	1/9/2013	30	7/9/2013	211
14	12/7/2012	1/9/2013	33	7/9/2013	214
15	12/12/2012	1/14/2013	33	7/9/2013	209
16	12/13/2012	1/14/2013	32	7/9/2013	208
17	12/14/2012	1/14/2013	31	7/9/2013	207
18	12/12/2012	1/14/2013	33	7/9/2013	209
19	12/12/2012	1/14/2013	33		
20	12/13/2012	1/14/2013	32		
21	12/13/2012	1/14/2013	32	7/9/2013	208
22	12/12/2012	1/14/2013	33		
23	12/12/2012	1/14/2013	33		
24	12/14/2012	1/14/2013	31	7/9/2013	207
25	12/13/2012	1/14/2013	32		
26	12/12/2012	1/14/2013	33		
Test Suite	Made	Leach Started	cure time (days)		
Tc-Gluconate	6/26/2013	7/29/2013	33		
Iodide	6/25/2013	7/24/2013	29		

Table A.3. Raw Data from C4124 UFA Squeezings Used to Generate VZP Simulant

Chemical Composition of Hanford H2 unit (sand) vadose pore water.

Based on UFA squeezings from borehole C4124

For the 200E area I had only 2 data sets from the same borehole 299-E27-22 (C4124. Two 1-ft long by 4-in diameter core liners for this borehole at 47.5 to 48.5 ft bgs and 81.5 to 82.5 ft bgs were removed from the core liners and aliquots of the sediment were quickly placed in centrifuge containers that were constructed with an inner bottom chamber that collected water that was ultra-centrifuged out of the sediment. The sediment was supported on a porous filter that allowed the extracted water to be collected below the filter. The UFA derived porewater data are found in the analytical template for this borehole PNNL in an excel file called C4124.xls located on the Geosciences group share drive "VZ-tank" in folder C Farm\C4124 (299-E27-22).
I have extracted the pertinent values from various worksheets in excel file C4124.xls and pasted the values below. The mid depth of the 1-ft long cores is used as the sample ID (48 and 82 ft). The F, Cl, NO3, Sulfate, Phosphate values come from work sheet **corrected anions.** The pH comes from work sheet **pH and conductivity**. The cation data are from worksheet **ICP-OES WE & AE see rows 179-180 and note conversion from ug/L to mg/L.** The alkalinity data are found in worksheet **alkalinity** as CaCO3. **Note that there was no phenolphthlein alkalinity so all the alkalinity is bicarbonate. Thus use bicarbonate chemical to make simulant. The average measured sulfate eemed suspiciously high compared to UFFA squeezings from uncontaminated boreholes in 200-W for which we had more measurements. Also geochemical modeling of the C4124 averaged measurements suggest that calcite could be oversaturated. Thus we elected to lower the sulfate and alkalinity some and after charge balancing there were slight increases in Ca, Na, and Cl. Our minor modifications resulted in the VZP pore water simulant shown in** Table 2.5**. Adjusted concentrations (mg/L) are compared to the averaged measured C4124 UFA data (uncharged balanced) below.**

	Actual Analytical Data		mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
		pH	Fluoride	Chloride	Nitrate	Alk as $CaCO_3$	Sulfate	Phosphate
299-E27-22	47.5 to 48.5'	7.18	<1.17	213.65	162.98	34.00	1323.68	<5.05
C4124	81.5 to 82.5'	6.93	<1.17	270.35	255.19	27.20	1675.81	<5.05
	Average	7.1	<1	242.00	209.09	30.6	1499.74	<5
	Values Used after adjustments and charge balancing	7.1	0	255.26	210.80	20.00	1152.00	0
		mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
		Ca	K	Mg	Na	Sr	Si	Al
299-E27-22	47.5 to 48.5'	475.2	29.2	152.7	110.4	1.8	23.1	<0.05
C4124	81.5 to 82.5'	448.5	23.8	101.2	118.3	1.6	22.8	<0.05
	Average	461.85	26.48	126.96	114.33	1.68	22.95	<0.05
	Values Used after adjustments and charge balancing	480.00	27.37	121.56	126.50	0	0	0

Table A.4. Measured Composition of VZP Simulant Compared to Theoretical

Constituent	**M**	**Theoretical ppm**	**Measured (Avg. of 8 Batches)**			**Difference (Theory - Measured)**
			Avg.	**Std Dev**	**% Std Dev**	
Ca	0.012	480	460.6	23.7	5.2%	4.1%
Mg	0.005	160.75	108.3	5.0	4.6%	39.0%
Na	0.0055	126.5	127.5	10.0	7.9%	-0.8%
K	0.0007	27.37	26.8	1.3	5.0%	2.0%
SO_4	0.0146	1401.6	1366.3	44.7	3.3%	2.6%
Cl	0.0072	255.26	260.9	7.5	2.9%	-2.2%
NO_3	0.0034	210.8	231	17.4	7.5%	-9.1%
HCO_3	0.0004	24.4	21.1	1.8	8.6%	14.5%

Appendix B

Observed Diffusion Coefficient, Fraction Leached, and Eluate pH and EC Worksheets

Appendix B

Observed Diffusion Coefficient, Fraction Leached, and Eluate pH and EC Worksheets

This appendix contains tables with the calculated observed diffusion coefficients, the fraction of each constituent's starting mass that has leached, the eluate pH and eluate EC all as a function of time from the EPA 1315 Leach Test on monoliths from the Extended, Archived, Iodide Spike, and Tc-Gluconate Suites of tests.

The tables include observed diffusion coefficient values for each Contaminant of Potential Concern (COPC) that leach from 4 different low-activity waste (LAW) waste simulants that had been solidified in Cast Stone (dry blend 8 wt% Portland cement, 45 wt% class F fly ash, and 47 wt% blast furnace slag). The mix ratio of liquid waste simulant to dry blend mix was performed at two weight ratios (0.4 and 0.6) of free water in waste simulant to dry blend.

The leach testing was performed on 2" diameter by 4" tall right cylinders, referred to as monoliths, which had been cured at room temperature and high (85 to 100%) relative humidity. The Archived Suite monoliths were cured/stored at room temperature and high relative humidity for 207 to 214 days prior to starting the Archive Suite leach tests. The Screening Test Extended Suite used monoliths cured for 29 to 33 days, a bit longer than the 28 days commonly used in grout leach testing. The leach testing used EPA Method 1315-January 2013 version. More details on the original Screening Tests are found in PNNL-22747. The Iodide Loading Suite and Tc-Gluconate Spiked Suites used monoliths cured for ~29 and 33 days, respectively at room temperature and high (85 to100%) relative humidity.

For all four Suites, the EPA 1315 leach testing was continued beyond the 63 day time period specified in EPA 1315. In the Screening Test Extended Suite, Cast Stone monoliths for 11 of the 26 original Cast Stone mixes, described in PNNL-22747, were leached well beyond the 63 day time period. A table (see Table 3.1 in main text) is provided for each of these mixes that had leaching "extended." For the Archived Suite, 12 of the original 26 Screening Test mixes had monoliths leached in both the DIW and VZP eluents after the prolonged curing period (see Table 4.2 in the main text). In the Iodide-Spiked Suite three iodide loadings were used and monoliths were leached in both DIW and VZP (see Table 4.5 in the main text). For the Tc-Gluconate Suite, 4 monoliths were spiked with a Tc(I) species and 4 monoliths were spiked with the more common Tc(VII) [pertechnetate] species (see Table 4.7 in the main text). Four of the Tc-Gluconate Suite monoliths were leached in DIW and four in VZP.

Those monoliths leached in a vadose zone pore water (VZP) simulant instead of deionized water require that their eluates be corrected for the eluent concentrations in the starting VZP. Because the VZP consists of a dilute salt solution of constituents found in Hanford sediments (mostly Na, Ca, Mg, sulfate, bicarbonate, chloride and nitrate), the starting eluent contains measureable concentrations of two of the COPCs for which observed diffusion coefficients are being calculated (Na, nitrate and in one instance nitrite). It is necessary to subtract the blank concentrations of these constituents from the measured eluates to obtain an accurate measure of the mass of these constituents that leached from the Cast Stone monoliths. Although no Cr was supposedly present in the chemical reagents used to make the VZP simulant, there were some detectable Cr concentrations measured in a few of the aliquots of the VZP eluent removed from the barrels prior to use and in a few of the VZP blanks. Thus, although there should not be Cr in the VZP simulant or the VZP blanks we honored the measurements and made Cr blank corrections to the monolith eluates. The measureable Cr in a few of the VZP simulant batches either represents trace contaminants in the chemical reagents used to make the simulant or analytical "vagaries". In either case vadose zone eluent corrections were also made on the Cr eluate data for tests using VZP eluent.

No day 93 blank samples were collected, analyzed, or reported by the analytical lab. To perform blank corrections for the DIW 93-d sampling it was assumed that the DIW 93-d blank analytical values were non-detects (ND) because all DIW blanks for other sampling periods were ND as one would expect. To perform blank corrections for the VZP 93-d sampling

- For Na and NO_3 that were known to be present the average values of blanks at all the earlier sampling times (0.08 through 63 days) were used.
- For Cr, the 93-d VZP blanks were assumed to be ND because the 63 and 380-d Cr blanks were ND and we did not feel that the early VZP Cr blanks that had detectable Cr values reported were fully justified despite analytical re-runs showing accurate results. We suspect some post-sampling Cr contamination issue occurred on the early interval blanks.
- For I-127, Tc-99, and U and NO_2, not present in the VZP simulant recipe, ND values from blanks were used in the calculation of D_{obs} values. This is a "conservative" approach that leads to a slightly larger D_{obs} calculated value, which is the protocol for use in performance assessment predictive modeling.

One oddity occurred in the batch of VZP eluent used to leach Cast Stone monoliths for the 427-d sampling. It appears that microbes reduced much of the VZP nitrate to nitrite prior to the eluent being added to the leach containers. This unexpected event was confirmed by triplicate analyses of the VZP blanks and residual solution left in the large barrel used to prepare VZP simulant. Thus the 427-d VZP blanks for nitrate and nitrite as measured were used in making blank corrections to the measured eluate concentrations. The microbe issue occurred because there was a many month hiatus in the testing program when funds were not provided to continue the leach tests. Apparently, microbes established themselves in sufficient quantities to significantly reduce nitrate to nitrite in the one barrel of VZP simulant that was not used for up to 10 months.

By calculation convention, when COPC analytical values fall below the Estimated Quantitation Limit (EQL) (i.e., a 'non-detect' with value signified by "ND"), the diffusivity and related expressions are calculated using the EQL value in place of "ND", and the values are formatted with ">" or "<" (as appropriate) to indicate that they are approximate values.

EPA Method 1315, Rev. 0, "Mass Transfer Rates of Constituents in Monolithic or Compacted Granular Materials Using a Semi-Dynamic Tank Leaching Procedure", January 2013—was used for the leach tests reported in this appendix. This method is included in EPA publication SW-846, entitled Test Methods for Evaluating Solid Waste, Physical/Chemical Methods. EPA Method 1315 can be found online at: https://www.epa.gov/hw-sw846/validated-test-method-1315-mass-transfer-rates-constituents-monolithic-or-compacted

Appendix B tables refer to duplicate monoliths (a,b,c) that mean monoliths from one wet slurry batch. Recall that six 2-in diameter by 4-in tall right cylinders were created from each batch of wet slurry. In PNNL-22747 the term replicate monoliths is used to describe monoliths of the same composition but made using two separate batches of wet slurry and the terms repeat or duplicates are used to describe monoliths made from the same batch of wet slurry. Replicates and duplicates thus differ and can yield different statistical information. We used duplicates (repeats) in our extended leach testing.

B.1 Extended Leach Tests

Eleven monoliths from the original Screening Test were subjected to extend leaching in DIW eluent. The specific monoliths are listed in Table 3.1 in the main text.

Table B.1. Sodium Extended Observed Diffusion Coefficient (cm^2/s)

Sodium Observed Diffusion Coefficient (cm^2/s)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	8.69E-08	1.21E-07	2.52E-07	7.15E-08	3.12E-07	**0.08**	3.97E-08	2.62E-08	7.80E-08	2.20E-08	1.43E-07	1.01E-07
1	3.60E-08	1.17E-08	4.19E-08	4.00E-08	2.92E-08	**1**	3.38E-08	2.37E-08	3.81E-08	1.05E-08	9.89E-08	6.02E-08
2	3.16E-08	6.03E-09	3.64E-08	3.27E-08	2.66E-08	**2**	2.82E-08	1.97E-08	3.78E-08	9.39E-09	8.07E-08	4.90E-08
7	1.17E-08	4.23E-09	2.18E-08	2.03E-08	1.49E-08	**7**	1.40E-08	1.09E-08	1.76E-08	4.38E-09	3.77E-08	2.47E-08
14	1.37E-08	3.91E-09	1.69E-08	1.37E-08	1.00E-08	**14**	1.24E-08	1.18E-08	1.29E-08	4.89E-09	3.21E-08	1.73E-08
28	3.99E-09	2.80E-09	1.07E-08	8.83E-09	6.01E-09	**28**	9.50E-09	8.84E-09	9.47E-09	3.23E-09	1.47E-08	1.42E-08
42	6.16E-09	2.53E-09	7.57E-09	7.02E-09	4.64E-09	**42**	8.22E-09	7.58E-09	6.61E-09	2.90E-09	1.09E-08	9.11E-09
49	5.16E-09	2.35E-09	6.99E-09	6.16E-09	4.32E-09	**49**	7.45E-09	6.66E-09	5.92E-09	2.86E-09	9.26E-09	8.54E-09
63	3.79E-09	2.11E-09	5.43E-09	4.14E-09	3.06E-09	**63**	5.88E-09	5.89E-09	4.60E-09	2.47E-09	6.61E-09	7.82E-09
77	3.40E-09	1.65E-09	3.70E-09	3.58E-09	2.18E-09	**77**	3.07E-09	3.39E-09	3.53E-09	2.12E-09	2.85E-09	3.67E-09
91	1.86E-09	1.06E-09	3.37E-09	2.89E-09	1.56E-09	**91**	3.03E-09	3.44E-09	2.92E-09	2.27E-09	2.65E-09	3.42E-09
119	2.15E-09	1.35E-09	3.43E-09	1.97E-09	1.30E-09	**119**	3.93E-09	4.18E-09	6.37E-09	2.35E-09	9.09E-10	3.73E-09
152	1.93E-09	1.69E-09	2.87E-09	1.76E-09	1.07E-09	**147**	2.77E-09	3.70E-09	1.72E-09	2.07E-09	1.53E-09	2.49E-09
187	1.68E-09	1.55E-09	2.64E-09	1.36E-09	9.49E-10	**182**	2.70E-09	3.68E-09	1.35E-09	2.14E-09	9.70E-10	2.07E-09
222	1.68E-09	1.59E-09	2.70E-09	1.41E-09	9.78E-10	**217**	2.69E-09	3.89E-09	1.54E-09	2.03E-09	9.03E-10	1.89E-09
257	1.23E-09	1.11E-09	2.00E-09	1.08E-09	7.44E-10	**252**	2.04E-09	3.03E-09	1.13E-09	1.72E-09	5.75E-10	1.37E-09
562	7.29E-10	8.08E-10	1.26E-09	4.36E-10	4.23E-10	**557**	1.04E-09	1.78E-09	5.39E-10	1.32E-09	2.44E-10	5.59E-10
609	1.86E-09	2.01E-09	3.59E-09	1.87E-09	1.42E-09	**604**	2.46E-09	3.83E-09	2.06E-09	3.93E-09	9.61E-10	2.08E-09
770	7.11E-10	8.57E-10	1.10E-09	4.82E-10	4.04E-10	**765**	8.46E-10	1.43E-09	5.17E-10	1.68E-09	2.65E-10	5.39E-10
1041		1.20E-09			4.20E-10	**1037**				1.64E-09	2.81E-10	5.64E-10

Table B.2. Nitrate Extended Observed Diffusion Coefficient (cm^2/s)

Nitrate Observed Diffusion Coefficient (cm^2/s)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	3.89E-07	5.02E-07	2.06E-07	3.02E-07	4.18E-07	**0.08**	5.33E-08	5.08E-08	7.19E-08	3.48E-08	2.00E-07	1.15E-07
1	3.96E-08	1.28E-08	4.84E-08	5.72E-08	4.82E-08	**1**	4.11E-08	3.14E-08	5.92E-08	1.51E-08	1.27E-07	6.88E-08
2	3.25E-08	6.23E-09	3.85E-08	4.07E-08	4.17E-08	**2**	3.09E-08	2.75E-08	5.22E-08	1.25E-08	1.05E-07	6.01E-08
7	2.83E-08	5.98E-09	2.97E-08	3.38E-08	3.20E-08	**7**	2.27E-08	1.76E-08	2.95E-08	7.78E-09	6.07E-08	3.37E-08
14	1.67E-08	4.31E-09	1.60E-08	2.08E-08	1.86E-08	**14**	1.61E-08	1.49E-08	1.84E-08	5.41E-09	3.77E-08	1.77E-08
28	1.12E-08	3.35E-09	9.35E-09	1.36E-08	1.01E-08	**28**	1.08E-08	1.19E-08	1.15E-08	4.01E-09	2.30E-08	1.47E-08
42	4.95E-09	2.27E-09	5.82E-09	8.37E-09	4.29E-09	**42**	4.20E-09	6.34E-09	5.56E-09	3.05E-09	1.19E-08	7.22E-09
49	4.61E-09	2.85E-09	4.60E-09	7.77E-09	6.27E-09	**49**	8.27E-09	7.91E-09	6.86E-09	3.45E-09	1.37E-08	8.65E-09
63	3.62E-09	2.53E-09	4.04E-09	5.49E-09	4.08E-09	**63**	5.83E-09	6.68E-09	5.72E-09	3.09E-09	9.92E-09	7.65E-09
77	2.48E-09	1.87E-09	2.35E-09	4.01E-09	2.41E-09	**77**	3.03E-09	4.20E-09	4.82E-09	2.87E-09	4.33E-09	3.97E-09
91	1.40E-09	1.19E-09	2.86E-09	3.71E-09	1.90E-09	**91**	2.53E-09	3.76E-09	3.83E-09	2.81E-09	3.42E-09	3.03E-09
119	1.75E-09	1.37E-09	2.54E-09	2.67E-09	1.61E-09	**119**	3.97E-09	5.28E-09	1.73E-08	3.33E-09	8.45E-10	3.74E-09
152	2.12E-09	2.67E-09	3.25E-09	3.14E-09	1.77E-09	**147**	3.38E-09	5.02E-09	2.88E-09	3.27E-09	2.52E-09	2.94E-09
187	1.78E-09	2.30E-09	2.90E-09	2.42E-09	1.44E-09	**182**	3.24E-09	5.24E-09	2.78E-09	3.64E-09	1.64E-09	2.38E-09
222	1.60E-09	2.29E-09	3.07E-09	2.23E-09	1.46E-09	**217**	3.08E-09	5.17E-09	3.05E-09	3.16E-09	1.23E-09	2.04E-09
257	1.21E-09	1.62E-09	2.38E-09	1.70E-09	1.22E-09	**252**	2.10E-09	4.11E-09	2.51E-09	2.72E-09	8.17E-10	1.42E-09
562	9.85E-10	1.58E-09	2.33E-09	1.15E-09	1.27E-09	**557**	1.54E-09	2.91E-09	2.30E-09	3.29E-09	5.56E-10	8.54E-10
609	8.27E-10	1.90E-09	2.11E-09	1.01E-09	1.07E-09	**604**	1.19E-09	2.39E-09	1.92E-09	4.14E-09	5.54E-10	6.26E-10
770	7.87E-10	1.58E-09	1.49E-09	9.24E-10	8.72E-10	**765**	9.50E-10	1.89E-09	1.38E-09	3.26E-09	4.49E-10	5.10E-10
1041		1.95E-09			8.05E-10	**1037**				3.00E-09	5.12E-10	5.64E-10

Table B.3. Nitrite Extended Observed Diffusion Coefficient (cm^2/s)

Nitrite Observed Diffusion Coefficient (cm^2/s)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	4.06E-07	2.70E-07	1.18E-07	1.52E-07	2.21E-07	**0.08**	6.00E-08	5.67E-08	7.59E-08	3.79E-08	2.15E-07	1.17E-07
1	3.43E-08	1.06E-08	4.17E-08	4.83E-08	4.45E-08	**1**	4.56E-08	3.46E-08	6.27E-08	1.65E-08	1.39E-07	6.65E-08
2	3.52E-08	6.30E-09	4.19E-08	4.35E-08	4.89E-08	**2**	3.57E-08	3.05E-08	5.80E-08	1.38E-08	1.18E-07	6.53E-08
7	2.89E-08	6.37E-09	3.12E-08	3.44E-08	3.55E-08	**7**	2.06E-08	1.57E-08	2.54E-08	6.94E-09	5.08E-08	2.73E-08
14	1.49E-08	3.93E-09	1.39E-08	1.85E-08	1.66E-08	**14**	2.02E-08	1.57E-08	2.19E-08	5.03E-09	3.71E-08	1.57E-08
28	1.13E-08	3.02E-09	8.10E-09	1.12E-08	9.02E-09	**28**	1.22E-08	1.05E-08	1.10E-08	3.24E-09	1.89E-08	1.06E-08
42	5.26E-09	2.28E-09	5.80E-09	7.89E-09	4.44E-09	**42**	4.79E-09	6.56E-09	6.09E-09	2.94E-09	1.24E-08	6.73E-09
49	4.63E-09	2.74E-09	4.44E-09	7.55E-09	6.37E-09	**49**	7.69E-09	6.94E-09	6.46E-09	3.16E-09	1.27E-08	7.73E-09
63	3.57E-09	2.30E-09	3.45E-09	4.71E-09	3.69E-09	**63**	5.96E-09	5.89E-09	5.53E-09	2.81E-09	9.30E-09	6.52E-09
77	2.47E-09	1.73E-09	2.15E-09	3.51E-09	2.24E-09	**77**	3.29E-09	4.04E-09	5.05E-09	2.86E-09	4.39E-09	3.61E-09
91	1.57E-09	1.16E-09	2.71E-09	3.29E-09	1.80E-09	**91**	3.00E-09	3.95E-09	4.45E-09	2.72E-09	3.56E-09	2.95E-09
119	1.84E-09	1.17E-09	1.96E-09	2.04E-09	1.35E-09	**119**	4.62E-09	6.05E-09	3.58E-09	3.43E-09	5.01E-09	3.73E-09
152	2.56E-09	2.49E-09	3.11E-09	2.98E-09	1.59E-09	**147**	3.48E-09	4.60E-09	2.58E-09	2.91E-09	2.43E-09	2.76E-09
187	1.47E-09	1.66E-09	1.99E-09	1.60E-09	9.25E-10	**182**	2.34E-09	3.44E-09	1.77E-09	2.41E-09	1.18E-09	1.68E-09
222	1.24E-09	1.57E-09	2.04E-09	1.37E-09	8.79E-10	**217**	2.21E-09	3.07E-09	1.82E-09	1.94E-09	8.70E-10	1.33E-09
257	9.97E-10	1.22E-09	1.69E-09	1.09E-09	7.77E-10	**252**	1.58E-09	2.65E-09	1.60E-09	1.70E-09	5.98E-10	1.06E-09
562	5.89E-10	9.39E-10	1.17E-09	5.56E-10	5.73E-10	**557**	8.69E-10	1.39E-09	9.45E-10	1.40E-09	3.11E-10	4.77E-10
609	6.28E-10	1.25E-09	1.29E-09	5.41E-10	5.34E-10	**604**	8.50E-10	1.51E-09	1.08E-09	2.31E-09	3.56E-10	4.39E-10
770	6.35E-10	1.17E-09	9.38E-10	5.26E-10	4.51E-10	**765**	7.19E-10	1.21E-09	7.52E-10	1.83E-09	2.98E-10	3.56E-10
1041		1.66E-09			4.46E-10	**1037**				1.91E-09	3.54E-10	4.53E-10

Table B.4. Iodine Extended Observed Diffusion Coefficient (cm^2/s)

Iodine Observed Diffusion Coefficient (cm^2/s)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	3.28E-07	1.07E-07	1.60E-07	1.79E-07	2.81E-07	**0.08**	3.16E-08	4.17E-08	5.89E-08	2.98E-08	1.79E-07	8.41E-08
1	3.77E-08	1.17E-08	3.32E-08	5.80E-08	5.86E-08	**1**	2.83E-08	2.56E-08	5.10E-08	1.42E-08	1.50E-07	5.65E-08
2	3.45E-08	6.18E-09	2.80E-08	4.11E-08	4.51E-08	**2**	2.43E-08	2.25E-08	4.77E-08	1.08E-08	1.19E-07	4.70E-08
7	2.33E-08	4.74E-09	1.94E-08	2.90E-08	2.99E-08	**7**	1.87E-08	1.61E-08	2.74E-08	7.30E-09	7.23E-08	2.93E-08
14	1.45E-08	3.79E-09	1.18E-08	2.00E-08	1.84E-08	**14**	1.35E-08	1.47E-08	1.79E-08	5.38E-09	4.90E-08	1.63E-08
28	9.32E-09	3.19E-09	6.93E-09	1.30E-08	9.07E-09	**28**	8.83E-09	1.08E-08	1.11E-08	3.87E-09	2.78E-08	1.26E-08
42	5.23E-09	2.68E-09	4.49E-09	9.15E-09	5.94E-09	**42**	6.13E-09	8.65E-09	7.72E-09	3.29E-09	1.74E-08	8.33E-09
49	3.56E-09	2.42E-09	3.42E-09	6.92E-09	4.56E-09	**49**	5.22E-09	6.84E-09	5.82E-09	2.88E-09	1.27E-08	6.19E-09
63	2.77E-09	2.22E-09	2.78E-09	5.74E-09	3.41E-09	**63**	4.03E-09	6.10E-09	5.20E-09	2.86E-09	9.07E-09	5.68E-09
77	1.74E-09	1.68E-09	1.61E-09	4.02E-09	2.01E-09	**77**	2.04E-09	3.66E-09	4.43E-09	2.61E-09	3.95E-09	3.25E-09
91	9.51E-10	1.07E-09	1.92E-09	3.85E-09	1.64E-09	**91**	1.85E-09	3.32E-09	3.63E-09	2.65E-09	3.22E-09	2.46E-09
119	1.25E-09	1.37E-09	1.96E-09	2.96E-09	1.49E-09	**119**	2.27E-09	4.21E-09	8.56E-09	2.94E-09	1.13E-09	2.51E-09
152	1.19E-09	2.03E-09	1.95E-09	2.62E-09	1.40E-09	**147**	1.78E-09	3.98E-09	2.30E-09	2.81E-09	1.84E-09	1.76E-09
187	9.92E-10	1.81E-09	1.77E-09	1.98E-09	1.21E-09	**182**	1.77E-09	4.20E-09	2.37E-09	3.25E-09	1.20E-09	1.42E-09
222	7.87E-10	1.70E-09	1.74E-09	1.62E-09	1.14E-09	**217**	1.47E-09	3.66E-09	2.37E-09	2.55E-09	7.82E-10	1.03E-09
257	6.32E-10	1.30E-09	1.48E-09	1.38E-09	1.24E-09	**252**	1.13E-09	3.31E-09	2.27E-09	2.47E-09	6.48E-10	7.89E-10
562	4.24E-10	7.11E-10	1.27E-09	3.74E-10	7.36E-10	**557**	6.80E-10	2.12E-09	8.82E-10	1.20E-09	1.85E-10	3.64E-10
609	5.02E-10	1.10E-09	1.42E-09	6.02E-10	8.26E-10	**604**	7.50E-10	2.08E-09	1.27E-09	2.71E-09	2.92E-10	3.33E-10
770	4.32E-10	1.40E-09	1.13E-09	7.62E-10	9.71E-10	**765**	5.32E-10	1.40E-09	1.49E-09	2.56E-09	3.38E-10	3.29E-10
1041		1.48E-09			8.38E-10	**1037**				2.49E-09	3.81E-10	3.73E-10

Table B.5. Technetium Extended Observed Diffusion Coefficient (cm^2/s)

Technetium Observed Diffusion Coefficient (cm^2/s)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	5.24E-11	2.19E-09	6.91E-09	7.56E-11	1.08E-10	**0.08**	2.30E-09	3.03E-10	3.52E-10	5.21E-11	3.04E-09	1.80E-09
1	1.08E-11	2.28E-10	2.01E-09	7.14E-11	1.65E-11	**1**	9.36E-10	2.95E-10	6.96E-11	1.44E-11	9.74E-10	6.62E-10
2	2.37E-11	2.51E-10	1.03E-09	2.19E-11	1.81E-11	**2**	3.01E-10	2.18E-10	1.32E-10	1.58E-11	1.18E-10	1.83E-10
7	2.76E-11	2.89E-10	4.57E-10	2.26E-11	1.47E-11	**7**	1.78E-10	1.81E-10	1.47E-10	1.36E-11	1.43E-11	1.05E-10
14	5.34E-11	2.75E-10	2.43E-10	5.11E-11	1.24E-11	**14**	1.25E-10	1.74E-10	1.89E-10	6.17E-12	1.40E-11	5.69E-11
28	5.00E-11	2.22E-10	1.49E-10	7.92E-11	1.12E-11	**28**	5.98E-11	1.35E-10	1.53E-10	4.46E-12	1.96E-11	5.77E-11
42	8.41E-11	2.21E-10	1.19E-10	9.68E-11	1.13E-11	**42**	4.27E-11	1.61E-10	1.89E-10	5.35E-12	4.58E-11	6.40E-11
49	8.58E-11	2.20E-10	1.16E-10	9.70E-11	1.17E-11	**49**	4.37E-11	1.83E-10	2.19E-10	6.58E-12	1.60E-10	7.16E-11
63	6.44E-11	2.26E-10	1.11E-10	8.44E-11	1.02E-11	**63**	3.43E-11	1.59E-10	1.61E-10	6.78E-12	2.26E-10	7.25E-11
77	3.73E-11	1.62E-10	7.17E-11	6.08E-11	7.34E-12	**77**	1.73E-11	1.08E-10	1.52E-10	7.17E-12	2.25E-10	4.79E-11
91	2.16E-11	8.26E-11	8.80E-11	5.71E-11	5.79E-12	**91**	1.81E-11	1.02E-10	1.29E-10	8.17E-12	3.50E-10	4.43E-11
119	2.55E-11	1.13E-10	1.09E-10	6.13E-11	8.32E-12	**119**	2.57E-11	1.44E-10	1.37E-09	1.30E-11	5.49E-11	7.26E-11
152	2.17E-11	1.72E-10	9.99E-11	6.90E-11	1.07E-11	**147**	2.31E-11	1.47E-10	1.28E-10	1.66E-11	6.46E-10	7.11E-11
187	2.18E-11	1.50E-10	1.19E-10	7.83E-11	9.43E-12	**182**	2.95E-11	1.69E-10	1.41E-10	2.48E-11	5.51E-10	9.83E-11
222	2.33E-11	1.34E-10	1.29E-10	7.38E-11	8.68E-12	**217**	2.98E-11	1.58E-10	1.47E-10	2.43E-11	3.86E-10	1.07E-10
257	2.64E-11	1.19E-10	1.42E-10	7.74E-11	8.91E-12	**252**	3.30E-11	1.68E-10	1.58E-10	3.05E-11	3.28E-10	1.25E-10
562	1.21E-11	8.25E-11	7.83E-11	1.06E-10	1.05E-11	**557**	1.86E-11	9.25E-11	2.02E-10	5.68E-11	1.39E-10	5.90E-11
609	2.94E-11	9.60E-11	1.46E-10	1.59E-10	1.03E-11	**604**	5.29E-11	1.98E-10	2.84E-10	8.97E-11	1.30E-10	7.03E-11
770	3.85E-11	6.27E-11	1.21E-10	1.10E-10	6.71E-12	**765**	3.47E-11	1.03E-10	1.10E-10	4.65E-11	7.91E-11	3.47E-11
1041		8.53E-11			1.04E-11	**1037**				4.87E-11	1.07E-10	

Table B.6. Chromium Extended Observed Diffusion Coefficient (cm^2/s)

Chromium Observed Diffusion Coefficient (cm^2/s)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	1.79E-12	7.57E-10	4.56E-11	8.10E-13	2.38E-13	**0.08**	4.49E-12	7.71E-11	1.06E-13	<1.18E-13	6.44E-13	3.18E-12
1	1.90E-13	2.83E-11	1.23E-11	5.98E-13	4.76E-14	**1**	3.70E-12	1.48E-11	9.96E-14	<1.94E-14	3.36E-13	2.45E-12
2	2.17E-13	5.22E-12	1.26E-11	5.13E-13	3.30E-14	**2**	5.22E-12	2.89E-12	1.33E-13	<5.70E-14	1.34E-13	1.94E-12
7	5.77E-14	1.17E-12	4.71E-12	2.04E-13	9.81E-15	**7**	5.02E-12	1.11E-12	1.53E-13	2.09E-14	4.57E-14	6.99E-13
14	4.93E-14	7.85E-13	2.19E-12	9.99E-14	9.01E-15	**14**	3.03E-12	5.66E-13	1.87E-13	1.29E-14	5.64E-14	2.65E-13
28	4.37E-14	6.52E-13	1.42E-12	6.75E-14	8.74E-15	**28**	9.10E-13	3.23E-13	1.85E-13	1.87E-14	8.11E-14	1.72E-13
42	5.77E-14	5.55E-13	9.18E-13	4.80E-14	7.56E-15	**42**	2.24E-13	2.10E-13	1.59E-13	8.46E-15	1.08E-13	8.91E-14
49	1.16E-13	7.39E-13	9.57E-13	5.36E-14	1.11E-14	**49**	1.93E-13	2.94E-13	2.07E-13	<3.64E-14	2.13E-13	1.19E-13
63	9.14E-14	6.91E-13	8.31E-13	5.59E-14	1.18E-14	**63**	1.10E-13	1.92E-13	1.52E-13	<1.12E-14	2.22E-13	8.33E-14
77	8.16E-14	5.73E-13	5.09E-13	5.22E-14	1.23E-14	**77**	1.17E-13	2.07E-13	2.00E-13	3.19E-14	1.90E-13	8.97E-14
91	6.16E-14	2.96E-13	5.70E-13	5.14E-14	1.05E-14	**91**	7.13E-14	1.77E-13	1.87E-13	2.59E-14	2.34E-13	7.28E-14
119	6.68E-14	2.84E-13	4.83E-13	4.29E-14	9.62E-15	**119**	8.36E-14	1.89E-13	1.03E-12	3.07E-14	7.12E-14	8.76E-14
152	6.79E-14	3.97E-13	3.77E-13	4.95E-14	7.79E-15	**147**	7.14E-14	1.66E-13	1.54E-13	2.50E-14	4.21E-13	6.67E-14
187	5.73E-14	3.50E-13	2.97E-13	4.71E-14	8.66E-15	**182**	6.79E-14	1.48E-13	1.76E-13	3.57E-14	3.86E-13	6.85E-14
222	6.09E-14	3.26E-13	3.04E-13	5.40E-14	7.95E-15	**217**	7.46E-14	1.42E-13	2.04E-13	3.84E-14	3.38E-13	6.15E-14
257	5.75E-14	3.00E-13	2.49E-13	5.62E-14	7.56E-15	**252**	7.28E-14	1.34E-13	1.91E-13	4.67E-14	2.75E-13	5.25E-14
562	3.39E-14	1.23E-13	1.15E-13	4.38E-14	4.46E-15	**557**	5.16E-14	6.67E-14	6.38E-14	4.23E-14	1.33E-13	3.62E-14
609	5.49E-14	2.42E-13	2.62E-13	1.14E-13	1.22E-14	**604**	1.11E-13	3.98E-13	2.11E-13	9.12E-14	3.02E-13	9.14E-14
770	3.33E-14	1.08E-13	6.75E-14	2.31E-15	4.27E-15	**765**	4.71E-14	6.08E-14	6.43E-14	3.03E-14	7.92E-14	2.71E-14
1041		9.69E-14			3.14E-15	**1037**				1.77E-14	9.55E-14	4.92E-14

Table B.7. Fraction of Sodium Leached versus Total Leach Time

Fraction of Sodium Leached (unitless)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	0.028	0.034	0.049	0.026	0.054	**0.08**	0.019	0.016	0.027	0.014	0.037	0.031
1	0.074	0.059	0.097	0.073	0.095	**1**	0.063	0.053	0.074	0.039	0.112	0.089
2	0.098	0.070	0.124	0.099	0.117	**2**	0.086	0.072	0.101	0.052	0.151	0.120
7	0.143	0.097	0.185	0.157	0.168	**7**	0.135	0.115	0.155	0.080	0.232	0.185
14	0.186	0.119	0.233	0.200	0.204	**14**	0.176	0.155	0.197	0.105	0.298	0.234
28	0.218	0.147	0.287	0.249	0.244	**28**	0.227	0.204	0.248	0.135	0.361	0.296
42	0.250	0.167	0.321	0.282	0.271	**42**	0.263	0.239	0.280	0.156	0.403	0.334
49	0.262	0.175	0.336	0.296	0.283	**49**	0.278	0.253	0.294	0.166	0.420	0.350
63	0.281	0.190	0.359	0.316	0.300	**63**	0.302	0.278	0.315	0.181	0.445	0.378
77	0.298	0.201	0.376	0.333	0.313	**77**	0.318	0.294	0.332	0.194	0.460	0.395
91	0.309	0.209	0.391	0.347	0.323	**91**	0.332	0.309	0.345	0.206	0.474	0.410
119	0.330	0.226	0.418	0.367	0.340	**119**	0.361	0.339	0.382	0.229	0.487	0.438
152	0.351	0.246	0.444	0.387	0.355	**147**	0.382	0.364	0.399	0.247	0.503	0.458
187	0.369	0.263	0.467	0.403	0.369	**182**	0.406	0.392	0.416	0.268	0.518	0.479
222	0.386	0.279	0.488	0.419	0.382	**217**	0.427	0.418	0.432	0.287	0.530	0.497
257	0.399	0.292	0.505	0.431	0.392	**252**	0.445	0.439	0.445	0.303	0.539	0.511
562	0.469	0.365	0.596	0.485	0.445	**557**	0.528	0.548	0.505	0.397	0.580	0.573
609	0.483	0.379	0.616	0.499	0.457	**604**	0.545	0.569	0.520	0.417	0.590	0.588
770	0.510	0.410	0.650	0.522	0.478	**765**	0.575	0.703	0.544	0.460	0.607	0.612
1041		0.462			0.509	**1037**				0.521	0.633	0.648

Table B.8. Fraction of Nitrate Leached versus Total Leach Time

Fraction of Nitrate Leached (unitless)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	0.060	0.068	0.044	0.053	0.062	**0.08**	0.022	0.022	0.026	0.018	0.043	0.033
1	0.108	0.095	0.096	0.110	0.115	**1**	0.071	0.064	0.084	0.047	0.129	0.096
2	0.133	0.106	0.124	0.138	0.143	**2**	0.095	0.087	0.116	0.063	0.174	0.130
7	0.202	0.138	0.195	0.214	0.217	**7**	0.157	0.142	0.187	0.099	0.276	0.206
14	0.249	0.162	0.241	0.267	0.266	**14**	0.204	0.187	0.237	0.126	0.347	0.255
28	0.304	0.192	0.292	0.327	0.318	**28**	0.258	0.244	0.292	0.159	0.426	0.318
42	0.332	0.211	0.322	0.364	0.345	**42**	0.284	0.276	0.322	0.181	0.470	0.352
49	0.344	0.220	0.334	0.379	0.358	**49**	0.300	0.291	0.336	0.192	0.490	0.368
63	0.363	0.236	0.354	0.402	0.378	**63**	0.324	0.317	0.360	0.209	0.522	0.395
77	0.377	0.248	0.368	0.420	0.392	**77**	0.339	0.335	0.380	0.224	0.540	0.413
91	0.386	0.257	0.381	0.436	0.403	**91**	0.352	0.351	0.396	0.238	0.555	0.427
119	0.405	0.273	0.405	0.459	0.421	**119**	0.381	0.385	0.456	0.264	0.569	0.455
152	0.427	0.298	0.432	0.486	0.441	**147**	0.405	0.414	0.478	0.287	0.589	0.477
187	0.446	0.320	0.456	0.508	0.459	**182**	0.431	0.447	0.502	0.315	0.608	0.500
222	0.463	0.339	0.479	0.527	0.474	**217**	0.454	0.477	0.525	0.338	0.622	0.519
257	0.476	0.354	0.497	0.543	0.487	**252**	0.471	0.501	0.544	0.358	0.633	0.533
562	0.556	0.456	0.622	0.630	0.579	**557**	0.573	0.642	0.668	0.507	0.695	0.609
609	0.566	0.470	0.637	0.641	0.590	**604**	0.584	0.658	0.683	0.528	0.702	0.617
770	0.594	0.511	0.676	0.672	0.620	**765**	0.616	0.628	0.721	0.587	0.724	0.641
1041		0.578			0.663	**1037**				0.670	0.759	0.677

Table B.9. Fraction of Nitrite Leached versus Total Leach

Fraction of Nitrite Leached (unitless)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	0.062	0.050	0.033	0.038	0.045	**0.08**	0.024	0.023	0.027	0.019	0.045	0.033
1	0.106	0.074	0.082	0.090	0.096	**1**	0.075	0.068	0.086	0.049	0.134	0.095
2	0.132	0.085	0.110	0.119	0.126	**2**	0.101	0.092	0.120	0.066	0.182	0.130
7	0.202	0.118	0.184	0.196	0.204	**7**	0.160	0.144	0.186	0.100	0.275	0.199
14	0.246	0.141	0.227	0.245	0.251	**14**	0.212	0.190	0.240	0.126	0.346	0.245
28	0.301	0.170	0.274	0.300	0.300	**28**	0.270	0.243	0.294	0.156	0.418	0.299
42	0.330	0.189	0.304	0.336	0.326	**42**	0.297	0.276	0.325	0.177	0.462	0.331
49	0.342	0.198	0.316	0.351	0.340	**49**	0.313	0.290	0.339	0.187	0.482	0.347
63	0.361	0.213	0.334	0.372	0.359	**63**	0.337	0.315	0.363	0.204	0.512	0.372
77	0.375	0.224	0.347	0.389	0.373	**77**	0.353	0.332	0.383	0.219	0.531	0.389
91	0.385	0.233	0.361	0.404	0.383	**91**	0.367	0.349	0.400	0.232	0.547	0.403
119	0.405	0.249	0.381	0.424	0.400	**119**	0.398	0.384	0.427	0.259	0.579	0.431
152	0.429	0.272	0.408	0.450	0.419	**147**	0.422	0.412	0.448	0.281	0.599	0.452
187	0.446	0.291	0.428	0.468	0.433	**182**	0.445	0.439	0.467	0.304	0.615	0.471
222	0.460	0.307	0.446	0.483	0.445	**217**	0.464	0.462	0.485	0.322	0.627	0.486
257	0.472	0.320	0.462	0.496	0.456	**252**	0.479	0.482	0.500	0.338	0.637	0.499
562	0.535	0.398	0.550	0.557	0.517	**557**	0.556	0.579	0.580	0.435	0.682	0.556
609	0.543	0.410	0.562	0.564	0.524	**604**	0.565	0.591	0.591	0.450	0.689	0.563
770	0.569	0.445	0.593	0.588	0.546	**765**	0.593	0.628	0.619	0.494	0.707	0.582
1041		0.507			0.578	**1037**				0.561	0.735	0.615

Table B.10. Fraction of Iodide Leached versus Total Leach Time

Fraction of Iodide Leached (unitless)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	0.055	0.032	0.039	0.041	0.051	**0.08**	0.017	0.020	0.023	0.017	0.041	0.028
1	0.102	0.057	0.082	0.098	0.109	**1**	0.057	0.058	0.077	0.045	0.134	0.085
2	0.127	0.068	0.105	0.126	0.138	**2**	0.079	0.079	0.108	0.060	0.182	0.115
7	0.190	0.097	0.163	0.197	0.209	**7**	0.135	0.132	0.176	0.095	0.293	0.186
14	0.234	0.119	0.203	0.248	0.259	**14**	0.178	0.176	0.225	0.122	0.374	0.233
28	0.284	0.148	0.246	0.307	0.308	**28**	0.227	0.230	0.280	0.154	0.461	0.291
42	0.313	0.169	0.273	0.345	0.339	**42**	0.258	0.267	0.315	0.177	0.514	0.328
49	0.323	0.177	0.283	0.360	0.350	**49**	0.271	0.282	0.328	0.186	0.534	0.341
63	0.340	0.192	0.300	0.384	0.369	**63**	0.291	0.306	0.351	0.203	0.564	0.365
77	0.351	0.203	0.311	0.401	0.381	**77**	0.304	0.323	0.370	0.217	0.581	0.381
91	0.359	0.212	0.322	0.417	0.392	**91**	0.315	0.338	0.385	0.231	0.596	0.394
119	0.376	0.229	0.343	0.442	0.409	**119**	0.336	0.368	0.428	0.256	0.611	0.417
152	0.392	0.250	0.364	0.467	0.427	**147**	0.354	0.394	0.447	0.277	0.629	0.434
187	0.406	0.269	0.383	0.487	0.443	**182**	0.373	0.424	0.469	0.303	0.645	0.451
222	0.418	0.286	0.400	0.503	0.456	**217**	0.389	0.449	0.490	0.324	0.657	0.465
257	0.427	0.300	0.414	0.517	0.470	**252**	0.402	0.471	0.508	0.343	0.666	0.476
562	0.480	0.368	0.506	0.567	0.539	**557**	0.469	0.591	0.585	0.433	0.702	0.525
609	0.487	0.379	0.519	0.575	0.549	**604**	0.478	0.605	0.597	0.450	0.707	0.531
770	0.509	0.417	0.553	0.603	0.581	**765**	0.502	0.644	0.637	0.502	0.726	0.550
1041		0.475			0.624	**1037**				0.579	0.756	0.579

Table B.11. Fraction of Technetium Leached versus Total Leach Time

Fraction of Technetium Leached (unitless)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	0.0007	0.005	0.008	0.001	0.001	**0.08**	0.005	0.002	0.002	0.0007	0.005	0.004
1	0.0015	0.008	0.019	0.003	0.002	**1**	0.012	0.006	0.004	0.0016	0.013	0.010
2	0.0022	0.010	0.023	0.004	0.003	**2**	0.014	0.008	0.005	0.0022	0.014	0.012
7	0.0043	0.017	0.032	0.005	0.004	**7**	0.020	0.013	0.010	0.0037	0.016	0.016
14	0.0070	0.023	0.038	0.008	0.005	**14**	0.024	0.018	0.015	0.0046	0.017	0.019
28	0.0107	0.031	0.044	0.013	0.007	**28**	0.028	0.024	0.022	0.0057	0.020	0.023
42	0.0143	0.037	0.048	0.017	0.008	**42**	0.031	0.029	0.027	0.0066	0.022	0.026
49	0.0159	0.040	0.050	0.018	0.009	**49**	0.032	0.032	0.030	0.0071	0.024	0.028
63	0.0184	0.044	0.054	0.021	0.010	**63**	0.034	0.036	0.034	0.0079	0.029	0.030
77	0.0201	0.048	0.056	0.023	0.011	**77**	0.035	0.039	0.037	0.0086	0.033	0.032
91	0.0213	0.050	0.058	0.025	0.011	**91**	0.036	0.041	0.040	0.0094	0.038	0.034
119	0.0236	0.055	0.063	0.029	0.013	**119**	0.038	0.047	0.057	0.0110	0.042	0.038
152	0.0259	0.061	0.068	0.033	0.014	**147**	0.040	0.052	0.062	0.0127	0.052	0.041
187	0.0280	0.067	0.073	0.037	0.016	**182**	0.043	0.058	0.067	0.0149	0.063	0.046
222	0.0299	0.071	0.078	0.040	0.017	**217**	0.045	0.063	0.072	0.0170	0.071	0.050
257	0.0319	0.075	0.082	0.044	0.018	**252**	0.047	0.068	0.077	0.0191	0.078	0.055
562	0.0408	0.099	0.105	0.070	0.026	**557**	0.058	0.093	0.114	0.0386	0.109	0.075
609	0.0426	0.102	0.109	0.074	0.027	**604**	0.061	0.098	0.120	0.0417	0.112	0.077
770	0.049	0.110	0.120	0.085	0.030	**765**	0.067	0.108	0.130	0.049	0.122	0.087
1041		0.124			0.035	**1037**				0.059	0.137	0.101

Table B.12. Fraction of Chromium Leached versus Total Leach Time

Fraction of Chromium Leached (unitless)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	0.0001	0.0027	0.0007	0.0001	0.0000	**0.08**	0.0002	0.0009	0.0000	0.0000	0.0001	0.0002
1	0.0002	0.0039	0.0015	0.0003	0.0001	**1**	0.0007	0.0018	0.0001	0.0001	0.0002	0.0005
2	0.0003	0.0042	0.0020	0.0004	0.0001	**2**	0.0010	0.0020	0.0002	0.0001	0.0003	0.0007
7	0.0004	0.0047	0.0029	0.0006	0.0002	**7**	0.0019	0.0024	0.0003	0.0002	0.0004	0.0011
14	0.0005	0.0050	0.0034	0.0007	0.0002	**14**	0.0025	0.0027	0.0005	0.0002	0.0004	0.0013
28	0.0006	0.0054	0.0040	0.0008	0.0002	**28**	0.0030	0.0030	0.0007	0.0003	0.0006	0.0015
42	0.0007	0.0057	0.0044	0.0009	0.0003	**42**	0.0032	0.0032	0.0009	0.0003	0.0007	0.0016
49	0.0007	0.0059	0.0046	0.0009	0.0003	**49**	0.0033	0.0033	0.0009	0.0003	0.0008	0.0017
63	0.0008	0.0061	0.0049	0.0010	0.0003	**63**	0.0034	0.0034	0.0011	0.0004	0.0010	0.0018
77	0.0009	0.0063	0.0051	0.0011	0.0004	**77**	0.0035	0.0036	0.0012	0.0004	0.0011	0.0018
91	0.0010	0.0065	0.0053	0.0011	0.0004	**91**	0.0036	0.0037	0.0013	0.0005	0.0012	0.0019
119	0.0011	0.0067	0.0056	0.0012	0.0004	**119**	0.0037	0.0039	0.0018	0.0005	0.0013	0.0021
152	0.0012	0.0070	0.0059	0.0013	0.0005	**147**	0.0038	0.0040	0.0019	0.0006	0.0016	0.0022
187	0.0013	0.0073	0.0061	0.0014	0.0005	**182**	0.0039	0.0042	0.0021	0.0007	0.0019	0.0023
222	0.0014	0.0075	0.0064	0.0015	0.0006	**217**	0.0041	0.0044	0.0023	0.0008	0.0021	0.0024
257	0.0015	0.0077	0.0066	0.0016	0.0006	**252**	0.0042	0.0045	0.0025	0.0009	0.0023	0.0025
562	0.0020	0.0086	0.0074	0.0022	0.0008	**557**	0.0047	0.0052	0.0031	0.0014	0.0033	0.0030
609	0.0021	0.0088	0.0076	0.0023	0.0008	**604**	0.0049	0.0054	0.0033	0.0015	0.0034	0.0031
770	0.0023	0.0091	0.0080	0.0023		**765**	0.0051	0.0056	0.0035	0.0017	0.0037	0.0032
1041		0.0096				**1037**				0.0019	0.0042	0.0036

Table B.13. pH of Eluates from Extended Suite Monoliths

pH of Extended Leach Tests (unitless)												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	11.4	11.0	11.3	11.2	11.4	**0.08**	11.6	11.4	11.6	11.3	11.9	11.7
1	12.0	11.4	11.9	11.9	11.9	**1**	11.9	11.8	11.9	11.6	12.2	12.0
2	12.0	11.2	11.8	11.8	11.8	**2**	11.7	11.6	11.8	11.4	12.0	12.0
7	12.3	11.7	12.2	12.2	12.2	**7**	12.0	11.9	12.0	11.8	12.3	12.2
14	12.1	11.6	12.1	12.0	12.0	**14**	12.0	11.9	12.0	11.7	12.2	12.1
28	12.2	11.6	12.1	12.1	12.1	**28**	12.0	11.8	12.0	11.7	12.1	12.1
42	12.0	11.5	11.9	11.8	11.8	**42**	11.8	11.7	11.8	11.5	11.9	11.9
49	11.6	11.2	11.7	11.6	11.5	**49**	11.6	11.5	11.6	11.3	11.7	11.7
63	11.8	11.3	11.8	11.7	11.6	**63**	11.7	11.6	11.6	11.5	11.7	11.9
77	11.6	11.2	11.6	11.5	11.5	**77**	11.5	11.5	11.5	11.4	11.6	11.7
91	11.7	11.1	11.8	11.6	11.6	**91**	11.5	11.5	11.5	11.3	11.5	11.6
119	11.6	11.1	11.7	11.4	11.4	**119**	11.8	11.6	11.6	11.4	11.6	11.7
152	11.8	11.3	11.7	11.5	11.5	**147**	11.7	11.6	11.5	11.4	11.6	11.7
187	11.7	11.2	11.7	11.4	11.5	**182**	11.7	11.6	11.4	11.4	11.5	11.7
222	11.7	11.1	11.6	11.4	11.4	**217**	11.7	11.6	11.4	11.4	11.5	11.6
257	11.6	11.0	11.6	11.3	11.3	**252**	11.6	11.6	11.3	11.2	11.4	11.5
562	12.2	11.1	12.0	11.6	11.7	**557**	12.1	11.9	11.6	11.4	11.6	12.0
609	11.6	10.8	11.5	11.2	11.3	**604**	11.5	11.5	11.2	11.2	11.3	11.5
770	11.5	10.4	11.6	11.2	11.3	**765**	11.7	11.6	11.1	11.1	11.1	11.5
1041		10.5			11.2	**1037**				11.0	11.0	11.4

Table B.14. Electrical Conductivity of Eluates from Extended Suite Monoliths

Electrical Conductivity (EC) units mS/cm —millisiemens per cm												
Mix Number (see Figure A.1)												
Test Duration (days)	**5a**	**8a**	**10b**	**13a**	**14b**	**Test Duration (days)**	**15a**	**16b**	**17a**	**18b**	**21b**	**24a**
0.08	0.87	0.49	0.63	0.66	0.56	**0.08**	1.77	1.60	1.58	1.26	3.70	2.32
1	4.43	1.52	3.62	3.48	3.81	**1**	4.10	3.33	3.58	2.26	7.97	4.58
2	2.68	0.76	2.22	2.05	2.26	**2**	2.33	1.90	2.13	1.24	4.27	2.70
7	6.41	1.80	4.98	4.55	5.16	**7**	5.63	4.48	4.65	2.96	9.67	5.89
14	4.77	1.56	3.80	3.58	3.71	**14**	4.41	3.77	3.66	2.33	7.08	4.15
28	5.35	1.83	4.32	3.86	4.07	**28**	5.27	4.53	4.04	2.80	7.35	5.08
42	3.63	1.42	3.00	2.72	2.79	**42**	3.61	3.31	2.78	2.10	4.69	3.46
49	1.54	0.67	1.40	1.22	1.24	**49**	1.69	1.55	1.31	1.01	2.08	1.64
63	2.18	0.99	1.94	1.59	1.71	**63**	2.44	2.32	1.70	1.40	2.72	2.34
77	1.79	0.78	1.44	1.25	1.35	**77**	1.70	1.72	1.38	1.23	1.84	1.69
91	1.35	0.58	1.45	1.19	1.08	**91**	1.54	1.59	1.21	1.15	1.57	1.35
119	2.31	0.97	2.17	1.51	1.64	**119**	2.72	2.64	1.62	1.86	2.44	2.25
152	2.41	1.26	2.17	1.45	1.59	**147**	2.32	2.45	1.35	1.63	1.88	1.88
187	2.06	1.06	1.92	1.27	1.42	**182**	2.44	2.66	1.30	1.81	1.69	1.86
222	1.63	0.83	1.48	0.94	1.06	**217**	1.83	2.09	1.05	1.37	1.21	1.39
257	1.39	0.71	1.37	0.87	0.98	**252**	1.61	1.95	0.95	1.26	1.06	1.20
562	5.51	2.79	4.89	2.61	3.57	**557**	6.05	7.30	3.21	5.81	3.15	3.89
609	1.32	0.63	1.30	0.82	0.97	**604**	1.42	1.69	0.90	1.37	0.93	1.08
770	2.12	1.22	1.96	1.14	1.51	**765**	2.36	2.93	1.26	2.63	1.34	1.51
1041		1.96			1.97	**1037**				3.65	1.8	1.86

B.2 Archive Leach Tests

Table B.15. Sodium Archive Test Observed Diffusion Coefficients

Interval (days)	Total Duration (days)	Sodium Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	3Arch-b	3Arch-c	5Arch-a	-	5Arch-c	8Arch-a	8Arch-b	8Arch-c
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	1.88E-08	2.17E-08	2.13E-08	1.30E-08	-	9.69E-09	2.49E-08	2.14E-08	2.86E-08
0.92	**1**	1.19E-08	1.36E-08	1.32E-08	5.75E-09	-	7.12E-09	3.13E-09	3.36E-09	3.63E-09
1	**2**	1.08E-08	1.15E-08	1.28E-08	5.98E-09	-	8.06E-09	3.06E-09	3.20E-09	3.60E-09
5	**7**	8.00E-09	8.05E-09	8.96E-09	5.30E-09	-	5.83E-09	2.78E-09	2.99E-09	3.22E-09
7	**14**	5.98E-09	6.50E-09	7.33E-09	4.43E-09	-	5.23E-09	2.65E-09	2.75E-09	3.16E-09
14	**28**	4.42E-09	4.84E-09	5.74E-09	3.04E-09	-	4.25E-09	2.12E-09	2.41E-09	2.63E-09
14	**42**	4.89E-09	4.91E-09	6.20E-09	3.05E-09	-	4.89E-09	2.27E-09	2.58E-09	2.93E-09
7	**49**	3.55E-09	4.04E-09	4.82E-09	2.57E-09	-	4.84E-09	2.23E-09	2.41E-09	2.38E-09
14	**63**	3.98E-09	4.32E-09	5.32E-09	2.42E-09	-	3.81E-09	1.64E-09	2.00E-09	2.47E-09
30	**93**	2.93E-09	2.78E-09	3.99E-09	1.86E-09	-	2.79E-09	1.25E-09	1.38E-09	1.63E-09
287	**380**	1.98E-09	2.04E-09	2.30E-09	1.48E-09	-	1.79E-09	7.32E-10	9.47E-10	1.02E-09
47	**427**	3.26E-09	2.42E-09	7.88E-09	1.31E-09	-	4.55E-09	1.02E-09	1.23E-09	2.03E-09
163	**590**	1.05E-09	1.05E-09	2.55E-09	8.24E-10		1.59E-09	5.69E-10	6.50E-10	8.03E-10
271	**861**	8.42E-10	7.23E-10	1.71E-09				7.65E-10		1.10E-09

Interval (days)	Total Duration (days)	Sodium Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	10Arch-b	10Arch-c	13Arch-a	13Arch-b	13Arch-c	14Arch-a	14Arch-b	14Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.48E-08	1.45E-08	1.68E-08	6.52E-09	1.26E-08	8.06E-09	5.68E-09	5.76E-09	3.76E-09
0.92	**1**	7.79E-09	6.75E-09	6.31E-09	6.72E-09	8.30E-09	5.71E-09	3.43E-09	3.01E-09	3.48E-09
1	**2**	6.01E-09	5.40E-09	6.28E-09	5.89E-09	5.91E-09	6.02E-09	3.87E-09	3.91E-09	5.09E-09
5	**7**	5.32E-09	4.68E-09	5.60E-09	5.09E-09	4.98E-09	4.59E-09	3.10E-09	3.60E-09	4.14E-09
7	**14**	4.22E-09	3.94E-09	4.96E-09	3.59E-09	3.36E-09	3.75E-09	2.42E-09	2.72E-09	3.30E-09
14	**28**	3.40E-09	3.22E-09	3.85E-09	2.21E-09	2.32E-09	2.85E-09	1.69E-09	2.34E-09	2.44E-09
14	**42**	3.22E-09	3.27E-09	4.55E-09	1.99E-09	2.20E-09	3.25E-09	1.96E-09	2.21E-09	2.47E-09
7	**49**	3.30E-09	3.18E-09	4.28E-09	2.04E-09	1.85E-09	3.30E-09	1.39E-09	1.89E-09	2.70E-09
14	**63**	2.85E-09	2.67E-09	3.68E-09	1.28E-09	1.56E-09	2.73E-09	1.51E-09	1.65E-09	2.25E-09
30	**93**	2.14E-09	2.14E-09	2.88E-09	8.15E-10	1.16E-09	1.75E-09	9.43E-10	1.08E-09	1.63E-09
287	**380**	1.44E-09	1.39E-09	2.01E-09	9.57E-10	9.40E-10	9.56E-10	9.67E-10	1.03E-09	1.31E-09
47	**427**	1.85E-09	1.58E-09	5.17E-09	1.79E-09	1.66E-09	3.36E-09	1.26E-09	1.21E-09	2.89E-09
163	**590**	7.93E-10	9.40E-10	1.83E-09	9.82E-10	1.15E-09	1.06E-09	9.57E-10	1.14E-09	1.15E-09
271	**861**							1.00E-09		1.06E-09

Interval (days)	**Total Duration (days)**	**Sodium Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.16E-08	1.21E-08	1.41E-08	1.88E-08	1.79E-08	2.30E-08	1.39E-08	1.41E-08	1.33E-08
0.92	**1**	7.38E-09	7.73E-09	8.51E-09	6.07E-09	6.15E-09	8.29E-09	8.90E-09	9.23E-09	1.02E-08
1	**2**	5.49E-09	6.05E-09	7.65E-09	5.49E-09	5.93E-09	7.24E-09	8.06E-09	8.33E-09	1.08E-08
5	**7**	5.23E-09	5.69E-09	6.26E-09	4.86E-09	4.78E-09	5.95E-09	6.11E-09	6.78E-09	7.31E-09
7	**14**	3.82E-09	4.18E-09	5.39E-09	4.02E-09	3.97E-09	4.94E-09	4.55E-09	4.30E-09	6.81E-09
14	**28**	3.30E-09	3.65E-09	5.14E-09	3.07E-09	2.96E-09	4.18E-09	2.78E-09	2.22E-09	4.39E-09
14	**42**	3.31E-09	3.32E-09	5.77E-09	2.84E-09	2.95E-09	5.42E-09	2.35E-09	2.28E-09	4.94E-09
7	**49**	3.29E-09	3.27E-09	3.96E-09	2.55E-09	2.24E-09	4.35E-09	2.19E-09	1.99E-09	4.23E-09
14	**63**	2.66E-09	2.72E-09	4.90E-09	2.23E-09	2.32E-09	5.00E-09	1.60E-09	1.78E-09	3.96E-09
30	**93**	2.12E-09	1.97E-09	3.45E-09	1.79E-09	1.68E-09	3.49E-09	1.48E-09	1.02E-09	2.34E-09
287	**380**	1.30E-09	1.37E-09	2.09E-09	1.36E-09	1.35E-09	2.43E-09	1.02E-09	1.47E-09	1.15E-09
47	**427**	1.46E-09	1.57E-09	4.32E-09	1.25E-09	1.50E-09	4.59E-09	1.50E-09	3.35E-09	3.98E-09
163	**590**	7.22E-10	7.11E-10	1.71E-09	6.77E-10	8.66E-10	1.74E-09	1.26E-09	2.02E-09	1.01E-09
271	**861**									

Interval (days)	**Total Duration (days)**	**Sodium Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	7.94E-09	1.05E-08	1.45E-08	2.14E-08	2.03E-08	2.00E-08	2.19E-08	2.73E-08	3.02E-08
0.92	**1**	5.36E-09	7.09E-09	6.30E-09	1.39E-08	1.42E-08	1.62E-08	1.22E-08	1.41E-08	1.22E-08
1	**2**	5.49E-09	5.65E-09	5.80E-09	1.27E-08	1.15E-08	1.69E-08	9.06E-09	1.01E-08	1.05E-08
5	**7**	4.78E-09	4.24E-09	3.63E-09	6.78E-09	9.59E-09	1.43E-08	7.29E-09	6.93E-09	7.07E-09
7	**14**	3.85E-09	3.17E-09	3.31E-09	7.76E-09	8.30E-09	1.27E-08	4.98E-09	5.04E-09	5.67E-09
14	**28**	2.60E-09	2.95E-09	2.46E-09	6.29E-09	6.05E-09	9.26E-09	3.55E-09	3.94E-09	4.19E-09
14	**42**	2.10E-09	2.41E-09	2.84E-09	5.37E-09	5.78E-09	8.86E-09	3.12E-09	3.66E-09	4.06E-09
7	**49**	1.99E-09	1.90E-09	2.34E-09	3.55E-09	4.25E-09	7.45E-09	2.68E-09	2.85E-09	3.12E-09
14	**63**	1.67E-09	2.05E-09	2.42E-09	3.92E-09	4.16E-09	7.02E-09	2.57E-09	2.76E-09	3.42E-09
30	**93**	1.20E-09	1.78E-09	1.93E-09	3.25E-09	3.36E-09	4.82E-09	1.80E-09	1.85E-09	2.88E-09
287	**380**	1.18E-09	1.44E-09	1.32E-09	1.51E-09	1.65E-09	1.60E-09	9.94E-10	1.20E-09	1.33E-09
47	**427**	1.81E-09	1.88E-09	3.00E-09	1.45E-09	1.32E-09	3.67E-09	1.06E-09	9.31E-10	3.92E-09
163	**590**	1.40E-09	1.23E-09	1.44E-09	8.34E-10	4.64E-10	8.82E-10	7.05E-10	5.53E-10	1.35E-09
271	**861**	1.49E-09		2.11E-09	8.35E-10		6.55E-10	7.26E-10		1.38E-09

Table B.16. Nitrate Archive Test Observed Diffusion Coefficients

Interval (days)	Total Duration (days)	Nitrate Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	**-**	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	2.35E-08	2.73E-08	2.72E-08	1.60E-08	-	1.05E-08	3.26E-08	2.97E-08	3.91E-08
0.92	**1**	1.25E-08	1.48E-08	1.40E-08	8.43E-09	-	9.86E-09	2.65E-09	2.81E-09	3.51E-09
1	**2**	1.30E-08	1.44E-08	1.41E-08	7.75E-09	-	9.33E-09	3.11E-09	3.67E-09	4.23E-09
5	**7**	8.98E-09	9.84E-09	9.72E-09	5.96E-09	-	7.29E-09	3.25E-09	3.83E-09	4.48E-09
7	**14**	6.27E-09	6.21E-09	7.54E-09	5.30E-09	-	6.54E-09	3.36E-09	3.62E-09	4.29E-09
14	**28**	5.54E-09	5.55E-09	7.58E-09	4.45E-09	-	6.83E-09	3.32E-09	3.77E-09	4.19E-09
14	**42**	5.19E-09	4.79E-09	7.08E-09	4.01E-09	-	6.01E-09	2.91E-09	3.05E-09	3.83E-09
7	**49**	3.80E-09	3.79E-09	4.69E-09	3.26E-09	-	5.24E-09	2.54E-09	2.56E-09	2.78E-09
14	**63**	4.32E-09	3.98E-09	5.93E-09	2.90E-09	-	4.48E-09	2.02E-09	2.19E-09	2.82E-09
30	**93**	3.73E-09	3.80E-09	6.00E-09	2.31E-09	-	3.98E-09	1.63E-09	1.18E-09	2.41E-09
287	**380**	3.73E-09	3.55E-09	5.18E-09	2.01E-09	-	2.90E-09	1.21E-09	1.31E-09	1.70E-09
47	**427**	2.65E-09	2.48E-09	6.54E-09	1.46E-09	-	2.61E-09	1.35E-09	1.62E-09	1.81E-09
163	**590**	1.19E-09	1.20E-09	4.50E-09	6.41E-10		2.11E-09	8.47E-10	6.16E-10	1.50E-09
271	**861**	1.17E-09	1.02E-09	2.72E-09				1.01E-09		1.68E-09

Interval (days)	Total Duration (days)	Nitrate Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.53E-08	1.57E-08	1.56E-08	9.56E-09	2.20E-08	1.37E-08	6.80E-09	7.87E-09	6.13E-09
0.92	**1**	4.95E-09	5.87E-09	5.00E-09	1.04E-08	1.30E-08	1.04E-08	6.20E-09	5.26E-09	6.58E-09
1	**2**	5.29E-09	5.73E-09	5.36E-09	2.92E-08	1.00E-08	9.36E-09	7.86E-09	7.78E-09	1.01E-08
5	**7**	4.63E-09	4.58E-09	4.76E-09	7.29E-09	7.27E-09	7.06E-09	6.48E-09	6.44E-09	7.79E-09
7	**14**	4.03E-09	3.92E-09	4.22E-09	5.57E-09	5.72E-09	5.52E-09	4.98E-09	5.47E-09	5.86E-09
14	**28**	3.84E-09	3.56E-09	4.38E-09	4.24E-09	4.67E-09	5.38E-09	3.39E-09	5.62E-09	5.31E-09
14	**42**	3.40E-09	3.10E-09	4.02E-09	3.18E-09	3.45E-09	4.83E-09	4.40E-09	4.51E-09	4.49E-09
7	**49**	2.87E-09	2.62E-09	3.25E-09	2.47E-09	3.00E-09	3.84E-09	2.69E-09	3.61E-09	3.81E-09
14	**63**	2.62E-09	2.41E-09	3.53E-09	2.15E-09	2.33E-09	3.84E-09	3.04E-09	3.04E-09	3.47E-09
30	**93**	2.48E-09	2.19E-09	3.41E-09	1.84E-09	1.89E-09	3.28E-09	2.73E-09	2.80E-09	3.59E-09
287	**380**	2.14E-09	1.93E-09	3.45E-09	2.00E-09	1.89E-09	2.41E-09	2.59E-09	2.45E-09	3.95E-09
47	**427**	1.68E-09	1.55E-09	3.40E-09	2.87E-09	3.08E-09	2.65E-09	2.60E-09	2.45E-09	3.78E-09
163	**590**	4.91E-10	5.07E-10	2.55E-09	3.36E-09	1.52E-09	2.45E-09	1.72E-09	1.88E-09	3.13E-09
271	**861**							1.85E-09		2.30E-09

Interval (days)	**Total Duration (days)**	**Nitrate Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.48E-08	1.45E-08	1.59E-08	2.14E-08	1.90E-08	2.67E-08	2.82E-08	2.97E-08	2.89E-08
0.92	**1**	8.10E-09	7.52E-09	9.82E-09	6.43E-09	6.70E-09	9.17E-09	1.20E-08	1.25E-08	1.71E-08
1	**2**	7.37E-09	8.16E-09	9.05E-09	6.81E-09	7.08E-09	8.66E-09	1.31E-08	1.44E-08	1.57E-08
5	**7**	6.19E-09	6.47E-09	7.80E-09	6.10E-09	5.87E-09	7.14E-09	1.09E-08	1.16E-08	1.96E-08
7	**14**	5.27E-09	5.33E-09	6.73E-09	5.37E-09	5.07E-09	6.22E-09	8.81E-09	8.12E-09	1.10E-08
14	**28**	5.03E-09	4.89E-09	7.92E-09	4.81E-09	4.57E-09	6.60E-09	6.63E-09	5.37E-09	9.72E-09
14	**42**	4.50E-09	4.30E-09	7.66E-09	4.30E-09	3.91E-09	6.87E-09	4.12E-09	3.94E-09	7.68E-09
7	**49**	3.59E-09	3.44E-09	4.11E-09	3.25E-09	2.79E-09	4.95E-09	3.20E-09	3.18E-09	4.97E-09
14	**63**	2.91E-09	3.07E-09	5.84E-09	2.94E-09	2.95E-09	5.92E-09	2.93E-09	2.75E-09	5.74E-09
30	**93**	2.49E-09	2.39E-09	5.23E-09	2.72E-09	2.79E-09	5.85E-09	2.37E-09	2.43E-09	5.28E-09
287	**380**	1.93E-09	1.95E-09	3.47E-09	2.11E-09	2.22E-09	4.40E-09	2.67E-09	3.72E-09	3.57E-09
47	**427**	1.22E-09	1.19E-09	2.90E-09	1.71E-09	1.82E-09	3.46E-09	3.72E-09	5.48E-09	3.35E-09
163	**590**	5.25E-10	5.35E-10	2.10E-09	6.88E-10	8.39E-10	2.31E-09	1.68E-09	2.39E-09	2.50E-09
271	**861**									

Interval (days)	**Total Duration (days)**	**Nitrate Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	8.79E-09	1.08E-08	1.51E-08	4.19E-08	4.66E-08	4.00E-08	2.52E-08	3.28E-08	3.89E-08
0.92	**1**	6.81E-09	8.20E-09	8.06E-09	2.61E-08	2.69E-08	3.14E-08	1.15E-08	3.15E-08	1.22E-08
1	**2**	9.11E-09	8.38E-09	8.06E-09	2.84E-08	2.85E-08	3.30E-08	1.05E-08	1.20E-08	1.08E-08
5	**7**	6.83E-09	6.78E-09	5.68E-09	1.43E-08	2.14E-08	2.84E-08	6.47E-09	7.66E-09	7.36E-09
7	**14**	5.58E-09	5.63E-09	4.58E-09	1.70E-08	1.77E-08	2.44E-08	5.32E-09	5.30E-09	5.38E-09
14	**28**	4.46E-09	5.17E-09	4.49E-09	1.57E-08	1.58E-08	2.17E-08	4.24E-09	4.52E-09	5.10E-09
14	**42**	3.07E-09	3.78E-09	4.10E-09	1.19E-08	1.20E-08	1.65E-08	3.08E-09	3.16E-09	4.21E-09
7	**49**	2.31E-09	2.19E-09	2.60E-09	6.83E-09	7.97E-09	9.30E-09	2.39E-09	2.52E-09	2.79E-09
14	**63**	2.23E-09	2.84E-09	3.33E-09	8.34E-09	8.37E-09	1.22E-08	2.19E-09	2.39E-09	3.29E-09
30	**93**	2.03E-09	2.62E-09	3.20E-09	6.60E-09	7.06E-09	9.97E-09	1.81E-09	1.89E-09	3.52E-09
287	**380**	2.25E-09	2.54E-09	2.83E-09	3.49E-09	3.82E-09	4.35E-09	1.49E-09	1.69E-09	2.63E-09
47	**427**	2.80E-09	2.53E-09	3.09E-09	2.48E-09	2.56E-09	3.57E-09	1.20E-09	1.16E-09	2.14E-09
163	**590**	2.08E-09	1.67E-09	3.00E-09	1.16E-09	9.15E-10	1.55E-09	3.24E-10	2.32E-10	1.67E-09
271	**861**	2.53E-09		3.43E-09	1.38E-09		1.04E-09	8.28E-10		1.56E-09

Table B.17. Nitrite Archive Test Observed Diffusion Coefficients

Interval (days)	Total Duration (days)	Nitrite Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	**-**	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	2.13E-08	2.24E-08	2.67E-08	1.61E-08	-	9.17E-09	3.39E-08	2.98E-08	3.61E-08
0.92	**1**	1.07E-08	1.41E-08	1.41E-08	6.58E-09	-	8.25E-09	1.90E-09	2.11E-09	2.45E-09
1	**2**	1.07E-08	1.19E-08	1.28E-08	7.01E-09	-	7.97E-09	2.54E-09	2.90E-09	3.19E-09
5	**7**	7.02E-09	7.72E-09	7.90E-09	3.98E-09	-	5.40E-09	2.22E-09	2.26E-09	3.00E-09
7	**14**	5.64E-09	5.67E-09	6.24E-09	3.79E-09	-	5.09E-09	2.60E-09	2.89E-09	3.16E-09
14	**28**	4.32E-09	4.06E-09	5.57E-09	3.51E-09	-	5.65E-09	2.07E-09	2.52E-09	2.84E-09
14	**42**	4.43E-09	4.18E-09	5.75E-09	2.82E-09	-	4.62E-09	2.38E-09	2.23E-09	2.57E-09
7	**49**	4.15E-09	4.06E-09	4.91E-09	3.40E-09	-	4.94E-09	2.29E-09	2.27E-09	2.37E-09
14	**63**	3.70E-09	3.37E-09	5.33E-09	2.16E-09	-	3.89E-09	1.61E-09	1.76E-09	2.09E-09
30	**93**	3.02E-09	3.12E-09	4.57E-09	1.76E-09	-	2.93E-09	1.26E-09	1.20E-09	1.76E-09
287	**380**	2.08E-09	1.96E-09	2.67E-09	1.41E-09	-	1.84E-09	7.93E-10	8.29E-10	1.03E-09
47	**427**	1.03E-09	9.68E-10	4.84E-09	5.94E-10	-	1.97E-09	2.32E-10	2.34E-10	1.22E-09
163	**590**	2.74E-09	2.76E-09	3.11E-09	2.29E-09		1.58E-09	1.32E-09	4.41E-09	1.10E-09
271	**861**	1.32E-09	1.16E-09	1.95E-09				8.47E-10		1.42E-09

Interval (days)	Total Duration (days)	Nitrite Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.53E-08	1.57E-08	1.38E-08	1.06E-08	2.04E-08	1.17E-08	6.36E-09	8.17E-09	5.62E-09
0.92	**1**	4.32E-09	4.99E-09	4.13E-09	7.80E-09	9.44E-09	8.87E-09	4.69E-09	4.12E-09	5.67E-09
1	**2**	4.27E-09	4.40E-09	4.20E-09	7.68E-09	7.23E-09	7.17E-09	6.32E-09	6.68E-09	7.82E-09
5	**7**	2.91E-09	2.79E-09	3.26E-09	4.16E-09	4.19E-09	4.70E-09	3.74E-09	4.45E-09	5.46E-09
7	**14**	2.58E-09	2.57E-09	3.19E-09	3.41E-09	3.53E-09	3.98E-09	3.05E-09	3.82E-09	4.34E-09
14	**28**	2.40E-09	2.24E-09	3.07E-09	2.72E-09	2.90E-09	3.73E-09	2.08E-09	3.62E-09	3.59E-09
14	**42**	2.43E-09	2.03E-09	2.85E-09	2.09E-09	2.22E-09	3.24E-09	2.64E-09	2.81E-09	3.16E-09
7	**49**	2.91E-09	2.65E-09	2.82E-09	2.08E-09	2.59E-09	3.21E-09	2.63E-09	3.55E-09	3.24E-09
14	**63**	2.01E-09	1.85E-09	2.67E-09	1.70E-09	1.74E-09	2.87E-09	1.97E-09	2.18E-09	2.59E-09
30	**93**	1.79E-09	1.68E-09	2.38E-09	1.46E-09	1.45E-09	2.16E-09	1.90E-09	2.06E-09	2.42E-09
287	**380**	1.15E-09	1.13E-09	1.89E-09	1.10E-09	8.13E-10	1.23E-09	1.23E-09	1.27E-09	1.76E-09
47	**427**	2.48E-10	3.44E-10	2.31E-09	8.03E-10	8.07E-10	1.63E-09	8.19E-10	7.44E-10	2.30E-09
163	**590**	3.18E-09	3.06E-09	1.75E-09	1.84E-08	4.18E-09	1.51E-09	3.51E-09	3.89E-09	1.90E-09
271	**861**							1.45E-09		1.56E-09

Interval (days)	**Total Duration (days)**	**Nitrite Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.47E-08	1.44E-08	1.38E-08	1.83E-08	1.81E-08	2.31E-08	1.83E-08	2.05E-08	2.20E-08
0.92	**1**	6.53E-09	6.30E-09	8.19E-09	4.50E-09	4.68E-09	7.36E-09	9.15E-09	8.92E-09	1.19E-08
1	**2**	6.14E-09	6.70E-09	6.87E-09	5.27E-09	5.41E-09	6.77E-09	8.02E-09	8.47E-09	1.08E-08
5	**7**	3.75E-09	4.22E-09	4.92E-09	3.61E-09	3.46E-09	4.80E-09	5.90E-09	6.35E-09	7.59E-09
7	**14**	3.48E-09	3.48E-09	4.81E-09	3.28E-09	3.03E-09	4.52E-09	5.36E-09	5.10E-09	7.02E-09
14	**28**	3.18E-09	3.15E-09	5.58E-09	2.74E-09	2.54E-09	4.20E-09	3.80E-09	2.85E-09	5.48E-09
14	**42**	3.01E-09	2.88E-09	5.33E-09	2.52E-09	2.27E-09	4.74E-09	2.47E-09	2.41E-09	4.54E-09
7	**49**	3.46E-09	3.16E-09	3.66E-09	2.71E-09	2.37E-09	4.08E-09	2.51E-09	2.51E-09	3.93E-09
14	**63**	2.18E-09	2.20E-09	4.70E-09	2.32E-09	2.32E-09	4.30E-09	1.87E-09	1.77E-09	4.15E-09
30	**93**	2.00E-09	1.91E-09	3.54E-09	1.86E-09	1.96E-09	3.72E-09	1.59E-09	1.61E-09	2.98E-09
287	**380**	1.12E-09	1.03E-09	1.69E-09	1.08E-09	1.17E-09	2.24E-09	1.16E-09	1.51E-09	1.48E-09
47	**427**	3.54E-10	3.58E-10	2.07E-09	3.79E-10	3.77E-10	2.33E-09	1.27E-09	1.83E-09	1.98E-09
163	**590**	2.29E-09	2.28E-09	1.52E-09	2.44E-09	2.60E-09	1.63E-09	2.83E-09	3.37E-09	1.44E-09
271	**861**									

Interval (days)	**Total Duration (days)**	**Nitrite Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	7.63E-09	9.33E-09	1.26E-08	3.64E-08	3.91E-08	3.73E-08	1.74E-08	2.39E-08	3.06E-08
0.92	**1**	5.18E-09	5.81E-09	6.15E-09	2.42E-08	2.51E-08	2.94E-08	8.30E-09	1.12E-08	9.87E-09
1	**2**	6.88E-09	6.36E-09	5.66E-09	2.41E-08	2.38E-08	2.95E-08	6.89E-09	8.55E-09	8.27E-09
5	**7**	4.09E-09	3.69E-09	3.57E-09	1.08E-08	1.63E-08	1.99E-08	4.35E-09	4.75E-09	5.06E-09
7	**14**	3.24E-09	3.48E-09	3.00E-09	1.39E-08	1.45E-08	1.84E-08	3.72E-09	4.05E-09	3.95E-09
14	**28**	2.46E-09	2.83E-09	2.70E-09	1.19E-08	1.11E-08	1.48E-08	2.67E-09	2.92E-09	3.17E-09
14	**42**	1.81E-09	2.21E-09	2.57E-09	9.25E-09	9.32E-09	1.25E-08	2.17E-09	2.27E-09	2.88E-09
7	**49**	1.80E-09	1.64E-09	2.05E-09	6.40E-09	7.60E-09	9.14E-09	2.39E-09	2.45E-09	2.52E-09
14	**63**	1.59E-09	1.98E-09	2.29E-09	7.23E-09	7.48E-09	1.00E-08	1.62E-09	1.73E-09	2.48E-09
30	**93**	1.31E-09	1.77E-09	1.93E-09	5.29E-09	5.20E-09	7.05E-09	1.37E-09	1.41E-09	2.33E-09
287	**380**	1.01E-09	1.28E-09	1.26E-09	1.98E-09	2.10E-09	2.10E-09	7.89E-10	8.61E-10	1.19E-09
47	**427**	7.25E-10	6.40E-10	1.82E-09	8.35E-10	6.16E-10	2.06E-09	3.85E-10	3.21E-10	1.56E-09
163	**590**	4.19E-09	3.31E-09	1.75E-09	2.63E-09	2.44E-09	1.13E-09	1.56E-09	1.37E-09	1.14E-09
271	**861**	1.88E-09		2.17E-09	1.36E-09		8.23E-10	8.80E-10		1.32E-09

Table B.18. Iodine Archive Test Observed Diffusion Coefficients

Interval (days)	**Total Duration (days)**	**Iodine Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	**-**	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	2.82E-08	3.19E-08	3.31E-08	9.21E-09	-	6.65E-09	1.61E-08	1.55E-08	2.14E-08
0.92	**1**	1.45E-08	1.70E-08	1.65E-08	6.12E-09	-	7.02E-09	2.35E-09	2.79E-09	3.14E-09
1	**2**	1.43E-08	1.57E-08	1.58E-08	5.94E-09	-	7.06E-09	2.84E-09	3.27E-09	3.55E-09
5	**7**	1.00E-08	1.11E-08	1.09E-08	5.06E-09	-	5.84E-09	2.82E-09	3.15E-09	3.68E-09
7	**14**	6.57E-09	7.88E-09	7.93E-09	4.13E-09	-	5.25E-09	2.70E-09	2.96E-09	3.75E-09
14	**28**	4.84E-09	5.55E-09	7.54E-09	3.50E-09	-	5.03E-09	2.50E-09	2.51E-09	3.13E-09
14	**42**	5.35E-09	5.31E-09	7.08E-09	2.78E-09	-	4.58E-09	2.18E-09	2.44E-09	2.84E-09
7	**49**	4.29E-09	4.41E-09	5.17E-09	2.47E-09	-	4.26E-09	2.14E-09	2.23E-09	2.09E-09
14	**63**	4.86E-09	4.57E-09	6.72E-09	2.15E-09	-	3.53E-09	1.74E-09	1.87E-09	2.41E-09
30	**93**	4.01E-09	4.18E-09	6.32E-09	1.57E-09	-	2.78E-09	1.38E-09	1.54E-09	1.93E-09
287	**380**	3.91E-09	3.83E-09	4.37E-09	1.13E-09	-	1.96E-09	8.94E-10	9.53E-10	8.55E-10
47	**427**	3.04E-09	2.86E-09	6.58E-09	1.09E-09	-	1.76E-09	1.19E-09	1.59E-09	1.23E-09
163	**590**	2.09E-09	2.13E-09	5.21E-09	6.98E-10		1.40E-09	9.98E-10	1.09E-09	8.69E-10
271	**861**	1.28E-09	1.11E-09	2.84E-09				7.82E-10		1.26E-09

Interval (days)	**Total Duration (days)**	**Iodine Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	7.58E-09	7.77E-09	7.76E-09	6.12E-09	1.43E-08	8.91E-09	5.45E-09	6.71E-09	5.08E-09
0.92	**1**	2.70E-09	2.92E-09	2.67E-09	7.22E-09	8.55E-09	6.81E-09	5.33E-09	4.67E-09	5.47E-09
1	**2**	2.77E-09	2.80E-09	2.80E-09	7.56E-09	7.32E-09	6.98E-09	6.87E-09	6.79E-09	8.97E-09
5	**7**	2.58E-09	2.48E-09	2.57E-09	6.11E-09	6.09E-09	6.02E-09	5.90E-09	6.47E-09	7.83E-09
7	**14**	2.29E-09	2.20E-09	2.48E-09	4.92E-09	4.94E-09	4.79E-09	4.70E-09	5.41E-09	6.11E-09
14	**28**	2.12E-09	1.97E-09	2.51E-09	3.59E-09	4.09E-09	3.97E-09	3.19E-09	5.16E-09	4.92E-09
14	**42**	2.14E-09	1.68E-09	2.52E-09	2.69E-09	2.59E-09	3.93E-09	4.06E-09	4.35E-09	4.18E-09
7	**49**	1.89E-09	1.78E-09	2.35E-09	2.11E-09	2.43E-09	3.46E-09	2.68E-09	3.46E-09	4.12E-09
14	**63**	1.73E-09	1.76E-09	2.55E-09	1.83E-09	2.11E-09	3.69E-09	3.15E-09	3.37E-09	3.78E-09
30	**93**	1.60E-09	1.46E-09	2.37E-09	1.57E-09	1.74E-09	3.03E-09	2.80E-09	2.87E-09	3.63E-09
287	**380**	1.31E-09	1.26E-09	1.67E-09	1.51E-09	1.44E-09	1.29E-09	3.05E-09	2.44E-09	2.37E-09
47	**427**	1.09E-09	1.02E-09	2.19E-09	2.56E-09	2.76E-09	1.96E-09	3.41E-09	2.76E-09	3.36E-09
163	**590**	6.46E-10	6.79E-10	1.88E-09	2.06E-09	2.29E-09	2.35E-09	2.45E-09	2.69E-09	3.23E-09
271	**861**							1.94E-09		2.30E-09

Interval (days)	**Total Duration (days)**	**Iodine Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	8.81E-09	8.66E-09	8.68E-09	1.53E-08	1.40E-08	1.82E-08	1.91E-08	2.24E-08	2.02E-08
0.92	**1**	4.86E-09	5.10E-09	5.69E-09	4.99E-09	5.09E-09	6.98E-09	9.78E-09	1.03E-08	1.31E-08
1	**2**	4.37E-09	4.62E-09	5.54E-09	5.09E-09	5.34E-09	6.75E-09	1.03E-08	1.07E-08	1.21E-08
5	**7**	3.57E-09	4.07E-09	4.97E-09	4.91E-09	4.98E-09	5.58E-09	8.97E-09	9.64E-09	1.11E-08
7	**14**	3.05E-09	3.24E-09	4.35E-09	4.39E-09	4.12E-09	4.90E-09	7.41E-09	6.85E-09	9.60E-09
14	**28**	2.95E-09	2.74E-09	4.82E-09	3.61E-09	3.28E-09	4.77E-09	4.88E-09	3.92E-09	7.82E-09
14	**42**	2.72E-09	2.39E-09	4.67E-09	3.22E-09	2.85E-09	5.66E-09	2.90E-09	3.07E-09	7.05E-09
7	**49**	2.09E-09	2.15E-09	2.77E-09	2.83E-09	2.40E-09	3.74E-09	2.54E-09	2.69E-09	4.86E-09
14	**63**	1.81E-09	1.88E-09	4.34E-09	2.61E-09	2.50E-09	5.19E-09	2.44E-09	2.38E-09	5.48E-09
30	**93**	1.42E-09	1.40E-09	3.28E-09	2.20E-09	2.28E-09	4.95E-09	2.10E-09	2.20E-09	4.81E-09
287	**380**	9.73E-10	9.52E-10	1.81E-09	1.45E-09	1.56E-09	3.20E-09	2.13E-09	3.41E-09	1.51E-09
47	**427**	6.73E-10	6.63E-10	1.71E-09	1.59E-09	1.41E-09	2.60E-09	3.07E-09	4.85E-09	1.90E-09
163	**590**	5.05E-10	5.11E-10	1.50E-09	7.15E-10	8.30E-10	1.94E-09	2.56E-09	3.56E-09	2.36E-09
271	**861**									

Interval (days)	**Total Duration (days)**	**Iodine Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	7.02E-09	8.90E-09	1.26E-08	4.46E-08	4.60E-08	3.94E-08	1.53E-08	1.88E-08	2.29E-08
0.92	**1**	5.18E-09	6.43E-09	6.14E-09	3.04E-08	3.06E-08	3.50E-08	7.03E-09	8.63E-09	7.25E-09
1	**2**	7.08E-09	6.80E-09	6.49E-09	3.20E-08	3.08E-08	3.76E-08	6.12E-09	7.36E-09	6.62E-09
5	**7**	6.19E-09	6.36E-09	4.81E-09	1.60E-08	2.52E-08	3.07E-08	4.28E-09	5.03E-09	4.66E-09
7	**14**	5.24E-09	5.04E-09	3.92E-09	1.82E-08	1.98E-08	2.76E-08	3.47E-09	3.55E-09	3.39E-09
14	**28**	3.81E-09	4.19E-09	3.53E-09	1.53E-08	1.52E-08	2.21E-08	2.57E-09	2.63E-09	3.14E-09
14	**42**	2.54E-09	3.19E-09	3.21E-09	1.17E-08	1.17E-08	1.72E-08	2.02E-09	2.15E-09	2.52E-09
7	**49**	2.14E-09	2.22E-09	2.40E-09	7.67E-09	9.52E-09	9.91E-09	1.87E-09	1.98E-09	1.69E-09
14	**63**	2.25E-09	2.71E-09	3.01E-09	8.66E-09	8.96E-09	1.41E-08	1.62E-09	1.67E-09	2.55E-09
30	**93**	1.76E-09	2.39E-09	2.86E-09	6.76E-09	7.29E-09	1.06E-08	1.27E-09	1.28E-09	2.41E-09
287	**380**	1.58E-09	2.13E-09	1.56E-09	3.51E-09	3.89E-09	2.90E-09	7.67E-10	9.89E-10	1.37E-09
47	**427**	2.73E-09	2.85E-09	2.37E-09	2.51E-09	2.10E-09	2.38E-09	6.35E-10	7.43E-10	1.50E-09
163	**590**	2.66E-09	2.03E-09	2.99E-09	1.76E-09	1.44E-09	1.71E-09	6.28E-10	5.01E-10	1.35E-09
271	**861**	2.28E-09		3.01E-09	1.51E-09		1.06E-09	5.58E-10		1.14E-09

Table B.19. Technetium Archive Test Observed Diffusion Coefficients

Interval (days)	**Total Duration (days)**	**Technetium Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	**-**	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	5.78E-14	3.80E-14	7.57E-14	5.46E-14	-	7.03E-14	1.20E-10	1.17E-10	2.39E-10
0.92	**1**	9.45E-13	5.28E-13	2.24E-12	8.02E-13	-	2.10E-12	1.75E-11	1.76E-11	4.45E-11
1	**2**	2.67E-12	2.55E-12	9.38E-12	2.24E-12	-	6.68E-12	1.86E-11	1.71E-11	5.60E-11
5	**7**	2.24E-12	3.09E-12	1.72E-11	1.70E-12	-	1.20E-11	1.68E-11	1.48E-11	8.57E-11
7	**14**	2.01E-12	2.42E-12	3.35E-11	2.35E-12	-	2.50E-11	1.19E-11	1.04E-11	1.26E-10
14	**28**	1.81E-12	2.85E-12	4.33E-11	1.56E-12	-	2.84E-11	5.64E-12	4.35E-12	1.46E-10
14	**42**	2.35E-12	2.77E-12	5.08E-11	1.64E-12	-	3.38E-11	5.90E-12	4.53E-12	1.38E-10
7	**49**	2.09E-12	3.26E-12	3.86E-11	1.92E-12	-	4.49E-11	6.48E-12	5.41E-12	1.28E-10
14	**63**	2.82E-12	3.61E-12	7.11E-11	1.28E-12	-	5.28E-11	5.19E-12	3.87E-12	1.50E-10
30	**93**	2.31E-12	2.88E-12	8.43E-11	1.06E-12	-	4.54E-11	3.22E-12	2.87E-12	1.38E-10
287	**380**	3.12E-12	3.49E-12	1.20E-10	6.67E-13	-	2.57E-11	1.90E-12	1.88E-12	8.75E-11
47	**427**	1.77E-12	1.45E-12	2.55E-10	4.64E-13	-	6.01E-11	4.53E-12	4.86E-12	9.30E-11
163	**590**	4.88E-13	4.70E-13	1.73E-10	2.36E-13		4.34E-11	1.01E-11	5.80E-12	6.15E-11
271	**861**	2.10E-13	2.05E-13	1.68E-10				8.82E-12		6.27E-11

Interval (days)	**Total Duration (days)**	**Technetium Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	2.04E-11	6.81E-12	6.01E-12	3.87E-12	1.12E-11	1.06E-12	7.48E-14	<3.23E-14	1.19E-12
0.92	**1**	7.73E-12	5.00E-12	1.09E-11	4.52E-12	4.65E-12	2.57E-12	6.03E-13	2.34E-13	2.99E-12
1	**2**	1.06E-11	8.31E-12	3.46E-11	5.00E-12	4.11E-12	4.09E-12	1.96E-12	8.75E-13	6.07E-12
5	**7**	9.71E-12	8.54E-12	5.69E-11	4.92E-12	4.08E-12	6.77E-12	2.11E-12	1.12E-12	7.10E-12
7	**14**	1.14E-11	9.94E-12	7.26E-11	4.78E-12	3.88E-12	1.32E-11	2.49E-12	1.57E-12	8.01E-12
14	**28**	8.54E-12	7.14E-12	8.81E-11	5.02E-12	5.25E-12	2.13E-11	2.15E-12	1.52E-12	7.54E-12
14	**42**	6.86E-12	6.25E-12	8.22E-11	5.28E-12	6.97E-12	3.23E-11	7.39E-12	1.55E-12	6.25E-12
7	**49**	6.90E-12	8.23E-12	8.42E-11	1.04E-11	1.24E-11	3.79E-11	3.34E-12	1.53E-12	7.32E-12
14	**63**	5.91E-12	5.39E-12	9.48E-11	8.15E-12	9.38E-12	4.83E-11	4.94E-12	1.34E-12	7.41E-12
30	**93**	3.68E-12	3.52E-12	9.00E-11	7.85E-12	9.47E-12	5.58E-11	3.74E-12	1.52E-12	9.07E-12
287	**380**	3.58E-12	2.32E-12	6.85E-11	2.23E-11	1.73E-11	1.49E-10	1.62E-12	7.20E-13	1.09E-11
47	**427**	2.31E-12	3.30E-12	7.66E-11	4.70E-11	3.48E-11	3.07E-10	8.11E-13	5.22E-13	7.14E-12
163	**590**	1.51E-12	1.48E-12	6.61E-11	2.70E-11	3.86E-11	1.87E-10	4.43E-13	3.53E-13	3.89E-12
271	**861**							2.45E-13		4.44E-12

Interval (days)	**Total Duration (days)**	**Technetium Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	4.31E-14	5.85E-14	2.13E-13	2.62E-11	9.11E-12	2.67E-11	2.86E-12	3.05E-12	1.75E-12
0.92	**1**	5.13E-13	7.25E-13	1.73E-12	3.37E-11	3.04E-11	1.30E-10	9.56E-12	8.01E-12	5.83E-11
1	**2**	1.23E-12	1.96E-12	5.46E-12	3.96E-11	3.45E-11	1.39E-10	2.34E-11	1.82E-11	1.16E-10
5	**7**	1.26E-12	1.84E-12	1.24E-11	2.68E-11	2.04E-11	1.05E-10	1.46E-11	1.33E-11	1.13E-10
7	**14**	1.55E-12	2.26E-12	2.74E-11	3.13E-11	2.62E-11	1.12E-10	1.44E-11	1.43E-11	1.72E-10
14	**28**	1.17E-12	1.74E-12	3.14E-11	2.79E-11	2.19E-11	1.39E-10	1.18E-11	1.74E-11	1.68E-10
14	**42**	1.40E-12	1.32E-12	3.70E-11	3.30E-11	2.71E-11	1.60E-10	1.31E-11	1.75E-11	1.95E-10
7	**49**	1.79E-12	1.93E-12	2.55E-11	4.00E-11	2.51E-11	1.56E-10	2.17E-11	2.64E-11	2.03E-10
14	**63**	1.21E-12	1.74E-12	4.15E-11	3.54E-11	3.14E-11	1.77E-10	1.93E-11	1.99E-11	1.90E-10
30	**93**	1.09E-12	1.09E-12	3.65E-11	2.23E-11	2.16E-11	1.69E-10	1.58E-11	1.92E-11	2.16E-10
287	**380**	7.39E-13	7.00E-13	2.32E-11	1.23E-11	9.49E-12	1.24E-10	1.26E-11	1.83E-11	2.94E-10
47	**427**	5.90E-13	7.19E-13	4.58E-11	9.19E-12	1.02E-11	1.40E-10	2.08E-11	7.23E-11	4.62E-10
163	**590**	1.24E-13	1.22E-13	2.75E-11	2.77E-12	2.50E-12	7.15E-11	2.02E-11	3.65E-11	1.61E-10
271	**861**									

Interval (days)	**Total Duration (days)**	**Technetium Observed Diffusion Coefficient (cm^2/s)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	6.87E-13	3.08E-14	2.72E-12	5.65E-14	5.53E-14	4.24E-14	<5.75E-14	6.03E-13	1.11E-13
0.92	**1**	2.91E-12	6.68E-12	1.24E-11	2.27E-13	1.60E-13	7.89E-13	5.15E-14	3.81E-13	1.94E-13
1	**2**	7.08E-12	1.14E-11	2.06E-11	1.22E-12	8.87E-13	3.54E-12	3.12E-13	9.57E-13	1.27E-12
5	**7**	6.65E-12	8.23E-12	1.69E-11	7.29E-13	1.25E-12	3.27E-11	4.33E-13	8.01E-13	2.22E-12
7	**14**	7.00E-12	8.33E-12	2.01E-11	2.72E-12	3.26E-12	1.55E-10	8.15E-13	8.81E-13	3.20E-12
14	**28**	5.93E-12	6.92E-12	2.02E-11	3.20E-12	4.79E-12	2.23E-10	5.26E-13	5.43E-13	5.66E-12
14	**42**	6.16E-12	5.81E-12	2.49E-11	4.68E-12	5.98E-12	2.89E-10	4.72E-13	6.05E-13	5.96E-12
7	**49**	5.89E-12	4.50E-12	2.20E-11	4.72E-12	5.65E-12	3.05E-10	9.77E-13	9.74E-13	5.59E-12
14	**63**	7.01E-12	6.00E-12	2.72E-11	8.19E-12	8.64E-12	4.04E-10	1.02E-12	9.24E-13	6.62E-12
30	**93**	5.53E-12	6.10E-12	3.10E-11	7.50E-12	7.81E-12	3.88E-10	6.48E-13	5.19E-13	1.05E-11
287	**380**	4.19E-12	4.03E-12	4.39E-11	9.22E-12	1.01E-11	2.19E-10	5.69E-13	5.65E-13	1.38E-11
47	**427**	5.13E-12	4.64E-12	3.04E-11	5.26E-12	4.29E-12	2.41E-10	8.36E-13	2.93E-13	2.14E-11
163	**590**	4.08E-12	1.90E-12	2.24E-11	2.35E-12	2.09E-12	1.57E-10	3.60E-13	1.28E-13	2.23E-11
271	**861**	7.67E-13		5.95E-11	2.03E-12		1.30E-10	5.02E-13		3.62E-11

Table B.20. Chromium Archive Test Observed Diffusion Coefficients

Interval (days)	Total Duration (days)	Chromium Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	3Arch-b	3Arch-c	5Arch-a	-	5Arch-c	8Arch-a	8Arch-b	8Arch-c
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	1.14E-15	1.03E-15	2.82E-13	4.18E-16	-	7.98E-14	7.71E-13	9.99E-13	2.26E-12
0.92	**1**	7.49E-17	6.17E-17	1.13E-13	1.10E-15	-	2.13E-14	1.26E-12	1.75E-12	2.32E-12
1	**2**	3.65E-16	1.41E-15	5.49E-14	2.29E-16	-	<1.58E-14	3.08E-13	4.80E-13	5.38E-13
5	**7**	1.31E-16	1.05E-15	8.47E-14	9.13E-18	-	1.80E-14	6.39E-14	9.29E-14	3.32E-13
7	**14**	<1.77E-15	<1.76E-15	9.05E-14	7.26E-17	-	2.63E-14	<6.11E-15	<9.72E-15	2.08E-13
14	**28**	1.16E-17	4.21E-17	6.08E-14	<1.14E-15	-	1.08E-14	1.30E-14	1.31E-14	9.94E-14
14	**42**	<1.50E-15	<1.50E-15	9.71E-14	<1.93E-15	-	3.63E-14	<2.21E-14	<2.22E-14	2.58E-13
7	**49**	<7.86E-15	<7.84E-15	1.07E-13	<1.01E-14	-	5.01E-14	<1.16E-13	<1.16E-13	2.03E-13
14	**63**	<2.41E-15	<2.41E-15	5.42E-14	<3.11E-15	-	3.73E-14	<3.55E-14	<3.57E-14	3.60E-13
30	**93**	<7.24E-16	<7.22E-16	7.04E-14	<9.34E-16	-	2.20E-14	<1.06E-14	<1.07E-14	2.07E-13
287	**380**	7.31E-17	3.85E-17	3.25E-14	8.59E-18	-	1.61E-14	3.91E-17	6.49E-17	1.13E-13
47	**427**	1.37E-17	5.99E-17	2.33E-12	1.07E-17	-	1.76E-13	3.92E-17	5.22E-16	4.23E-13
163	**590**	<1.61E-16	<1.61E-16	4.85E-14	5.20E-17		2.78E-14	<1.91E-16	5.22E-21	9.69E-15
271	**861**	4.77E-18	3.80E-19	3.89E-14				2.71E-16		7.86E-15

Interval (days)	Total Duration (days)	Chromium Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	10Arch-b	10Arch-c	13Arch-a	13Arch-b	13Arch-c	14Arch-a	14Arch-b	14Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.18E-12	6.39E-13	3.69E-12	2.41E-12	1.40E-12	<6.50E-14	5.42E-15	1.66E-15	<3.45E-14
0.92	**1**	5.94E-13	5.87E-13	2.55E-12	9.35E-14	1.05E-13	1.74E-14	5.59E-16	1.07E-16	2.15E-14
1	**2**	3.97E-13	3.87E-13	1.63E-12	6.63E-14	9.01E-14	<3.16E-14	<1.65E-14	<1.63E-14	<1.67E-14
5	**7**	2.02E-13	2.39E-13	8.94E-13	5.82E-14	4.30E-14	2.67E-14	1.10E-16	2.36E-16	1.93E-14
7	**14**	8.12E-14	1.00E-13	4.28E-13	2.04E-14	1.32E-14	1.63E-14	<4.81E-18	<2.33E-15	7.37E-15
14	**28**	2.84E-14	3.46E-14	1.37E-13	3.55E-15	1.11E-15	4.66E-15	<1.18E-15	<1.17E-15	1.26E-15
14	**42**	1.13E-14	1.09E-14	1.40E-13	4.22E-17	<4.02E-15	1.60E-14	<2.00E-15	<1.98E-15	4.08E-15
7	**49**	1.86E-17	1.61E-16	1.17E-13	<2.04E-14	<2.10E-14	2.36E-14	<1.05E-14	<1.04E-14	<1.07E-14
14	**63**	1.40E-15	7.26E-16	1.33E-13	<6.27E-15	<6.41E-15	1.32E-11	<3.23E-15	<3.19E-15	6.76E-15
30	**93**	<1.75E-15	4.12E-19	7.22E-14	<1.88E-15	<1.91E-15	1.18E-14	<9.68E-16	<9.58E-16	1.77E-15
287	**380**	3.81E-17	4.68E-17	5.30E-14	<5.68E-17	8.06E-18	1.68E-14	<2.92E-17	2.89E-17	9.55E-16
47	**427**	3.31E-17	2.84E-17	1.60E-13	5.30E-17	3.16E-17	8.77E-14	6.13E-17	4.68E-17	3.50E-15
163	**590**	<3.90E-16	<3.75E-16	4.51E-14	<4.19E-16	<4.21E-16	1.69E-14	<2.16E-16	<2.13E-16	1.04E-15
271	**861**							6.78E-19		4.16E-16

Interval (days)	Total Duration (days)	Chromium Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	15Arch-b	15Arch-c	16Arch-a	16Arch-b	16Arch-c	17Arch-a	17Arch-b	17Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	5.22E-15	1.33E-14	1.33E-13	3.19E-13	8.06E-13	1.71E-12	1.14E-14	6.46E-15	<6.20E-14
0.92	**1**	1.26E-14	8.29E-15	7.25E-14	1.16E-13	3.75E-13	1.43E-12	3.57E-15	6.02E-15	2.27E-14
1	**2**	2.30E-15	4.45E-15	4.98E-14	1.65E-14	2.32E-14	2.15E-13	6.58E-15	3.93E-15	<3.01E-14
5	**7**	2.58E-15	1.26E-14	4.01E-13	4.40E-15	2.82E-15	1.92E-13	2.31E-15	2.41E-15	7.05E-14
7	**14**	6.05E-17	1.58E-15	2.49E-13	1.58E-17	<7.69E-15	1.25E-13	<4.37E-15	<4.35E-15	5.66E-14
14	**28**	3.08E-20	1.80E-17	1.73E-13	<3.87E-15	<3.85E-15	6.14E-14	2.87E-17	9.96E-18	2.67E-14
14	**42**	1.93E-15	<1.95E-15	2.22E-13	<6.57E-15	<6.53E-15	1.15E-13	<3.72E-15	<3.69E-15	7.20E-14
7	**49**	<1.01E-14	<1.02E-14	1.08E-13	<3.44E-14	<3.43E-14	1.22E-13	<1.95E-14	<1.94E-14	6.59E-14
14	**63**	<3.11E-15	3.07E-16	1.46E-13	<1.06E-14	<1.05E-14	1.28E-13	7.95E-18	<5.94E-15	8.35E-14
30	**93**	<9.32E-16	<9.43E-16	6.08E-14	<3.17E-15	<3.16E-15	7.74E-14	<1.79E-15	<1.78E-15	5.80E-14
287	**380**	4.47E-17	3.81E-17	2.46E-14	2.60E-21	<9.53E-17	3.35E-14	<5.42E-17	1.40E-18	6.20E-14
47	**427**	5.37E-18	3.69E-17	1.01E-13	2.40E-16	1.44E-16	2.75E-13	5.05E-18	2.69E-16	1.43E-13
163	**590**	<2.08E-16	<2.10E-16	2.32E-14	<7.06E-16	<7.03E-16	3.48E-14	<4.00E-16	1.21E-16	4.10E-14
271	**861**									

Interval (days)	Total Duration (days)	Chromium Observed Diffusion Coefficient (cm^2/s)								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	18Arch-b	18Arch-c	21Arch-a	21Arch-b	21Arch-c	24Arch-a	24Arch-b	24Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.28E-14	2.99E-15	<1.18E-13	7.99E-15	9.09E-15	4.32E-14	2.85E-14	4.81E-14	1.45E-13
0.92	**1**	6.09E-17	4.89E-17	<1.95E-14	3.79E-15	3.60E-15	1.77E-14	1.70E-14	2.98E-14	7.27E-14
1	**2**	1.01E-16	7.56E-17	<5.74E-14	1.64E-15	1.46E-15	<1.24E-14	2.36E-14	3.39E-14	4.33E-14
5	**7**	2.96E-16	5.81E-16	2.22E-14	2.49E-17	3.55E-16	1.51E-14	1.57E-14	1.82E-14	9.12E-14
7	**14**	<8.32E-15	<8.13E-15	9.99E-15	<1.76E-15	<1.77E-15	1.50E-14	1.04E-15	3.29E-15	6.83E-14
14	**28**	<4.16E-15	<4.06E-15	<4.09E-15	<8.81E-16	5.40E-18	1.28E-14	2.72E-15	1.26E-15	4.01E-14
14	**42**	<7.07E-15	<6.91E-15	1.24E-14	<1.50E-15	<1.50E-15	3.07E-14	<3.52E-15	<3.47E-15	5.51E-14
7	**49**	<3.71E-14	<3.62E-14	<3.65E-14	<7.85E-15	<7.88E-15	3.10E-14	<1.85E-14	<1.82E-14	6.63E-14
14	**63**	<1.14E-14	<1.11E-14	2.96E-14	2.41E-15	<2.42E-15	4.79E-14	4.98E-17	<5.58E-15	6.18E-14
30	**93**	<3.41E-15	<3.33E-15	1.49E-14	<7.23E-16	<7.26E-16	4.54E-14	<1.70E-15	<1.67E-15	4.63E-14
287	**380**	<1.03E-16	<1.01E-16	9.32E-15	2.27E-17	2.38E-17	2.98E-14	4.67E-17	1.85E-16	2.49E-14
47	**427**	1.67E-16	<1.78E-15	3.04E-14	3.69E-19	9.43E-18	6.15E-14	8.05E-18	3.16E-17	7.68E-14
163	**590**	<7.60E-16	<7.43E-16	1.12E-14	<1.61E-16	<1.62E-16	1.95E-14	<3.79E-16	<3.73E-16	2.48E-14
271	**861**	5.09E-17		9.23E-15	2.07E-17		1.78E-14	5.09E-18		2.30E-14

Table B.21. Fraction of Sodium Leached versus Total Leach Time Archive Monoliths

Interval (days)	Total Duration (days)	Fraction of Sodium Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	3Arch-b	3Arch-c	5Arch-a	-	5Arch-c	8Arch-a	8Arch-b	8Arch-c
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	0.013	0.014	0.014	0.011	-	0.010	0.015	0.014	0.016
0.92	**1**	0.039	0.042	0.041	0.029	-	0.030	0.029	0.028	0.031
1	**2**	0.054	0.057	0.057	0.040	-	0.042	0.036	0.036	0.039
5	**7**	0.091	0.094	0.096	0.070	-	0.074	0.058	0.058	0.063
7	**14**	0.119	0.123	0.128	0.094	-	0.100	0.077	0.078	0.083
14	**28**	0.153	0.159	0.167	0.123	-	0.134	0.101	0.103	0.110
14	**42**	0.181	0.187	0.198	0.145	-	0.162	0.120	0.123	0.132
7	**49**	0.192	0.198	0.210	0.154	-	0.174	0.128	0.132	0.140
14	**63**	0.211	0.218	0.233	0.169	-	0.194	0.141	0.146	0.156
30	**93**	0.242	0.248	0.270	0.194	-	0.224	0.161	0.167	0.179
287	**380**	0.389	0.397	0.428	0.321	-	0.364	0.250	0.269	0.284
47	**427**	0.412	0.416	0.463	0.335	-	0.390	0.263	0.282	0.302
163	**590**	0.451	0.455	0.524	0.370		0.439	0.292	0.313	0.337
271	**861**	0.500	0.501	0.594				0.339		0.393

Interval (days)	Total Duration (days)	Fraction of Sodium Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	10Arch-b	10Arch-c	13Arch-a	13Arch-b	13Arch-c	14Arch-a	14Arch-b	14Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.012	0.012	0.013	0.008	0.011	0.009	0.007	0.007	0.006
0.92	**1**	0.033	0.031	0.032	0.027	0.033	0.027	0.021	0.021	0.020
1	**2**	0.044	0.042	0.043	0.038	0.043	0.038	0.030	0.029	0.030
5	**7**	0.074	0.070	0.074	0.067	0.072	0.066	0.053	0.054	0.056
7	**14**	0.098	0.094	0.100	0.089	0.094	0.088	0.071	0.073	0.077
14	**28**	0.128	0.123	0.133	0.114	0.119	0.116	0.093	0.099	0.103
14	**42**	0.151	0.146	0.160	0.132	0.137	0.139	0.110	0.117	0.122
7	**49**	0.161	0.156	0.172	0.140	0.145	0.149	0.117	0.125	0.131
14	**63**	0.177	0.172	0.191	0.151	0.157	0.165	0.129	0.138	0.146
30	**93**	0.204	0.199	0.222	0.167	0.177	0.189	0.147	0.157	0.169
287	**380**	0.330	0.323	0.372	0.269	0.278	0.292	0.250	0.263	0.288
47	**427**	0.347	0.339	0.400	0.286	0.294	0.315	0.264	0.277	0.309
163	**590**	0.381	0.376	0.453	0.324	0.335	0.354	0.301	0.318	0.350
271	**861**							0.355		0.405

Interval (days)	**Total Duration (days)**	**Fraction of Sodium Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.010	0.011	0.011	0.013	0.013	0.015	0.011	0.012	0.011
0.92	**1**	0.031	0.032	0.033	0.032	0.032	0.037	0.034	0.035	0.035
1	**2**	0.041	0.042	0.045	0.042	0.043	0.049	0.046	0.047	0.050
5	**7**	0.071	0.073	0.078	0.071	0.071	0.081	0.079	0.081	0.085
7	**14**	0.094	0.097	0.104	0.094	0.094	0.107	0.104	0.106	0.116
14	**28**	0.124	0.128	0.141	0.123	0.123	0.141	0.131	0.130	0.150
14	**42**	0.146	0.151	0.172	0.144	0.144	0.171	0.150	0.149	0.178
7	**49**	0.156	0.161	0.182	0.153	0.153	0.183	0.158	0.157	0.190
14	**63**	0.173	0.178	0.204	0.168	0.168	0.205	0.171	0.170	0.210
30	**93**	0.199	0.203	0.238	0.192	0.192	0.239	0.193	0.189	0.237
287	**380**	0.318	0.325	0.387	0.314	0.313	0.404	0.299	0.315	0.350
47	**427**	0.333	0.340	0.413	0.328	0.328	0.431	0.314	0.338	0.375
163	**590**	0.365	0.373	0.463	0.360	0.364	0.482	0.357	0.393	0.413
271	**861**									

Interval (days)	**Total Duration (days)**	**Fraction of Sodium Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.009	0.010	0.012	0.014	0.014	0.014	0.014	0.016	0.017
0.92	**1**	0.026	0.030	0.031	0.042	0.042	0.044	0.041	0.044	0.043
1	**2**	0.036	0.041	0.041	0.058	0.057	0.062	0.054	0.058	0.057
5	**7**	0.065	0.068	0.066	0.092	0.097	0.111	0.089	0.093	0.092
7	**14**	0.088	0.088	0.087	0.124	0.131	0.152	0.115	0.119	0.120
14	**28**	0.114	0.117	0.113	0.165	0.171	0.202	0.146	0.152	0.154
14	**42**	0.132	0.136	0.135	0.195	0.201	0.240	0.168	0.176	0.179
7	**49**	0.140	0.144	0.143	0.205	0.213	0.255	0.177	0.185	0.189
14	**63**	0.153	0.158	0.159	0.225	0.233	0.281	0.193	0.202	0.207
30	**93**	0.173	0.182	0.184	0.257	0.266	0.321	0.218	0.226	0.238
287	**380**	0.286	0.308	0.304	0.386	0.400	0.452	0.322	0.341	0.359
47	**427**	0.303	0.325	0.326	0.401	0.415	0.476	0.335	0.353	0.383
163	**590**	0.348	0.368	0.372	0.436	0.441	0.512	0.367	0.382	0.428
271	**861**	0.414		0.450	0.485		0.555	0.413		0.491

Table B.22. Fraction of Nitrate Leached versus Total Leach Time Archive Monoliths

Interval (days)	**Total Duration (days)**	**Fraction of Nitrate Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	**-**	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	0.015	0.016	0.016	0.012	-	0.010	0.017	0.017	0.019
0.92	**1**	0.041	0.045	0.044	0.034	-	0.034	0.030	0.029	0.033
1	**2**	0.057	0.061	0.061	0.046	-	0.047	0.037	0.038	0.042
5	**7**	0.096	0.102	0.101	0.078	-	0.083	0.061	0.063	0.070
7	**14**	0.125	0.131	0.133	0.105	-	0.113	0.082	0.085	0.094
14	**28**	0.164	0.170	0.178	0.140	-	0.156	0.112	0.117	0.128
14	**42**	0.193	0.197	0.212	0.165	-	0.187	0.134	0.139	0.153
7	**49**	0.203	0.208	0.224	0.175	-	0.200	0.143	0.148	0.162
14	**63**	0.224	0.227	0.248	0.192	-	0.221	0.157	0.163	0.178
30	**93**	0.259	0.263	0.292	0.219	-	0.257	0.180	0.182	0.207
287	**380**	0.460	0.458	0.530	0.367	-	0.436	0.295	0.302	0.343
47	**427**	0.481	0.478	0.562	0.382	-	0.456	0.309	0.318	0.360
163	**590**	0.522	0.520	0.643	0.413		0.512	0.345	0.348	0.407
271	**861**	0.580	0.573	0.731				0.399		0.476

Interval (days)	**Total Duration (days)**	**Fraction of Nitrate Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.012	0.012	0.012	0.009	0.014	0.011	0.008	0.009	0.008
0.92	**1**	0.029	0.031	0.029	0.034	0.042	0.036	0.027	0.026	0.027
1	**2**	0.039	0.041	0.040	0.057	0.055	0.049	0.039	0.038	0.041
5	**7**	0.067	0.069	0.068	0.093	0.091	0.084	0.073	0.072	0.077
7	**14**	0.090	0.093	0.092	0.120	0.118	0.111	0.099	0.099	0.105
14	**28**	0.123	0.124	0.127	0.154	0.154	0.150	0.129	0.138	0.143
14	**42**	0.146	0.146	0.153	0.177	0.177	0.177	0.155	0.165	0.169
7	**49**	0.155	0.155	0.163	0.185	0.187	0.188	0.164	0.175	0.180
14	**63**	0.171	0.171	0.182	0.200	0.202	0.208	0.182	0.193	0.198
30	**93**	0.200	0.198	0.215	0.224	0.227	0.241	0.212	0.223	0.232
287	**380**	0.353	0.344	0.411	0.372	0.371	0.403	0.380	0.387	0.439
47	**427**	0.369	0.359	0.434	0.393	0.392	0.424	0.400	0.407	0.463
163	**590**	0.396	0.387	0.496	0.463	0.440	0.484	0.451	0.460	0.530
271	**861**							0.524		0.611

Interval (days)	**Total Duration (days)**	**Fraction of Nitrate Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.012	0.012	0.012	0.014	0.013	0.016	0.016	0.017	0.016
0.92	**1**	0.033	0.032	0.036	0.033	0.033	0.039	0.042	0.043	0.048
1	**2**	0.045	0.045	0.049	0.045	0.045	0.052	0.058	0.060	0.065
5	**7**	0.078	0.078	0.085	0.077	0.076	0.088	0.102	0.105	0.123
7	**14**	0.104	0.105	0.115	0.104	0.103	0.117	0.136	0.138	0.162
14	**28**	0.141	0.141	0.161	0.140	0.138	0.160	0.178	0.176	0.213
14	**42**	0.168	0.167	0.195	0.166	0.163	0.193	0.204	0.201	0.248
7	**49**	0.178	0.177	0.206	0.176	0.172	0.206	0.214	0.211	0.261
14	**63**	0.195	0.195	0.230	0.193	0.189	0.230	0.231	0.228	0.284
30	**93**	0.224	0.222	0.271	0.223	0.220	0.274	0.259	0.256	0.326
287	**380**	0.369	0.368	0.464	0.375	0.376	0.496	0.430	0.458	0.524
47	**427**	0.382	0.381	0.485	0.391	0.393	0.520	0.454	0.487	0.547
163	**590**	0.410	0.410	0.541	0.423	0.428	0.579	0.503	0.547	0.608
271	**861**									

Interval (days)	**Total Duration (days)**	**Fraction of Nitrate Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.009	0.010	0.012	0.020	0.021	0.019	0.015	0.018	0.019
0.92	**1**	0.029	0.032	0.033	0.058	0.060	0.061	0.041	0.060	0.045
1	**2**	0.042	0.045	0.046	0.082	0.083	0.087	0.055	0.075	0.060
5	**7**	0.076	0.079	0.077	0.131	0.144	0.156	0.088	0.111	0.095
7	**14**	0.104	0.106	0.102	0.179	0.192	0.213	0.115	0.138	0.122
14	**28**	0.138	0.144	0.137	0.244	0.258	0.289	0.149	0.173	0.160
14	**42**	0.160	0.168	0.163	0.288	0.301	0.340	0.171	0.196	0.185
7	**49**	0.169	0.177	0.171	0.302	0.317	0.357	0.180	0.204	0.195
14	**63**	0.184	0.193	0.190	0.331	0.346	0.392	0.194	0.220	0.213
30	**93**	0.209	0.223	0.222	0.377	0.394	0.449	0.219	0.245	0.247
287	**380**	0.366	0.390	0.398	0.572	0.598	0.666	0.346	0.381	0.416
47	**427**	0.386	0.409	0.420	0.592	0.615	0.687	0.360	0.394	0.435
163	**590**	0.442	0.459	0.487	0.633	0.652	0.734	0.382	0.413	0.484
271	**861**	0.527		0.586	0.696		0.789	0.431		0.552

Table B.23. Fraction of Nitrite Leached versus Total Leach Time Archive Monoliths

Interval (days)	Total Duration (days)	Fraction of Nitrite Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	3Arch-b	3Arch-c	5Arch-a	-	5Arch-c	8Arch-a	8Arch-b	8Arch-c
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	0.014	0.014	0.016	0.012	-	0.009	0.018	0.017	0.018
0.92	**1**	0.039	0.043	0.044	0.032	-	0.031	0.028	0.028	0.030
1	**2**	0.053	0.058	0.060	0.043	-	0.043	0.035	0.035	0.038
5	**7**	0.088	0.094	0.096	0.069	-	0.074	0.055	0.055	0.061
7	**14**	0.115	0.121	0.125	0.092	-	0.100	0.073	0.074	0.081
14	**28**	0.149	0.154	0.164	0.123	-	0.139	0.097	0.100	0.109
14	**42**	0.176	0.180	0.194	0.144	-	0.166	0.117	0.119	0.129
7	**49**	0.187	0.191	0.206	0.154	-	0.178	0.125	0.127	0.137
14	**63**	0.206	0.209	0.229	0.169	-	0.198	0.138	0.141	0.152
30	**93**	0.238	0.241	0.267	0.193	-	0.229	0.158	0.161	0.176
287	**380**	0.388	0.386	0.437	0.317	-	0.371	0.251	0.255	0.281
47	**427**	0.401	0.399	0.464	0.326	-	0.388	0.257	0.261	0.295
163	**590**	0.464	0.462	0.532	0.384		0.437	0.301	0.342	0.335
271	**861**	0.526	0.519	0.606				0.350		0.399

Interval (days)	Total Duration (days)	Fraction of Nitrite Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	10Arch-b	10Arch-c	13Arch-a	13Arch-b	13Arch-c	14Arch-a	14Arch-b	14Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.012	0.012	0.011	0.010	0.014	0.011	0.008	0.009	0.007
0.92	**1**	0.028	0.029	0.027	0.031	0.037	0.033	0.024	0.024	0.025
1	**2**	0.037	0.038	0.036	0.043	0.049	0.044	0.035	0.036	0.038
5	**7**	0.059	0.060	0.060	0.070	0.076	0.073	0.061	0.063	0.068
7	**14**	0.078	0.079	0.081	0.091	0.097	0.096	0.081	0.086	0.093
14	**28**	0.103	0.104	0.109	0.118	0.125	0.127	0.105	0.117	0.124
14	**42**	0.123	0.122	0.131	0.137	0.144	0.150	0.125	0.139	0.147
7	**49**	0.132	0.131	0.140	0.145	0.153	0.159	0.134	0.149	0.157
14	**63**	0.146	0.145	0.156	0.157	0.166	0.176	0.148	0.164	0.173
30	**93**	0.171	0.168	0.184	0.179	0.188	0.202	0.173	0.190	0.201
287	**380**	0.283	0.280	0.329	0.289	0.282	0.317	0.290	0.308	0.341
47	**427**	0.289	0.287	0.348	0.300	0.293	0.333	0.301	0.319	0.360
163	**590**	0.357	0.355	0.399	0.465	0.372	0.380	0.373	0.395	0.413
271	**861**							0.438		0.481

Interval (days)	Total Duration (days)	Fraction of Nitrite Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	15Arch-b	15Arch-c	16Arch-a	16Arch-b	16Arch-c	17Arch-a	17Arch-b	17Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.012	0.012	0.011	0.01	0.01	0.01	0.013	0.014	0.014
0.92	**1**	0.031	0.030	0.033	0.03	0.03	0.03	0.036	0.036	0.040
1	**2**	0.042	0.042	0.045	0.04	0.04	0.05	0.048	0.049	0.055
5	**7**	0.067	0.069	0.074	0.06	0.06	0.07	0.080	0.082	0.091
7	**14**	0.089	0.090	0.099	0.09	0.08	0.10	0.107	0.109	0.122
14	**28**	0.118	0.119	0.138	0.11	0.11	0.13	0.139	0.136	0.160
14	**42**	0.140	0.141	0.168	0.13	0.13	0.16	0.159	0.156	0.187
7	**49**	0.150	0.150	0.178	0.14	0.14	0.17	0.168	0.165	0.198
14	**63**	0.165	0.165	0.200	0.16	0.15	0.19	0.181	0.178	0.218
30	**93**	0.190	0.190	0.234	0.18	0.18	0.22	0.204	0.201	0.250
287	**380**	0.301	0.296	0.371	0.29	0.29	0.38	0.317	0.330	0.376
47	**427**	0.308	0.303	0.389	0.30	0.30	0.40	0.331	0.347	0.394
163	**590**	0.367	0.361	0.436	0.358	0.362	0.443	0.396	0.417	0.440
271	**861**									

Interval (days)	Total Duration (days)	Fraction of Nitrite Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	18Arch-b	18Arch-c	21Arch-a	21Arch-b	21Arch-c	24Arch-a	24Arch-b	24Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.008	0.009	0.011	0.018	0.019	0.019	0.013	0.015	0.017
0.92	**1**	0.026	0.028	0.030	0.056	0.057	0.059	0.034	0.040	0.041
1	**2**	0.037	0.039	0.040	0.077	0.078	0.083	0.046	0.053	0.054
5	**7**	0.064	0.064	0.065	0.120	0.131	0.141	0.073	0.082	0.083
7	**14**	0.084	0.086	0.085	0.163	0.175	0.191	0.096	0.105	0.107
14	**28**	0.110	0.113	0.112	0.220	0.230	0.254	0.123	0.133	0.136
14	**42**	0.127	0.132	0.133	0.258	0.268	0.299	0.141	0.152	0.158
7	**49**	0.135	0.139	0.141	0.272	0.283	0.315	0.150	0.161	0.166
14	**63**	0.147	0.153	0.156	0.299	0.311	0.347	0.162	0.174	0.182
30	**93**	0.168	0.177	0.181	0.341	0.352	0.395	0.184	0.196	0.210
287	**380**	0.273	0.296	0.299	0.488	0.503	0.546	0.276	0.293	0.325
47	**427**	0.283	0.306	0.316	0.499	0.513	0.564	0.284	0.300	0.340
163	**590**	0.362	0.376	0.367	0.561	0.573	0.605	0.332	0.345	0.382
271	**861**	0.436		0.447	0.624		0.653	0.382		0.444

Table B.24. Fraction of Iodide Leached versus Total Leach Time Archive Monoliths

Interval (days)	Total Duration (days)	Fraction of Iodide Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	**-**	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	0.016	0.017	0.018	0.009	-	0.008	0.012	0.012	0.014
0.92	**1**	0.045	0.048	0.048	0.028	-	0.028	0.024	0.025	0.028
1	**2**	0.061	0.065	0.066	0.039	-	0.040	0.031	0.033	0.036
5	**7**	0.103	0.109	0.109	0.068	-	0.071	0.053	0.056	0.061
7	**14**	0.133	0.141	0.141	0.092	-	0.098	0.072	0.076	0.084
14	**28**	0.169	0.180	0.187	0.122	-	0.135	0.098	0.102	0.113
14	**42**	0.198	0.209	0.220	0.143	-	0.162	0.117	0.121	0.134
7	**49**	0.209	0.220	0.233	0.152	-	0.173	0.125	0.130	0.142
14	**63**	0.231	0.241	0.258	0.167	-	0.192	0.138	0.143	0.157
30	**93**	0.267	0.278	0.304	0.189	-	0.222	0.159	0.166	0.183
287	**380**	0.474	0.481	0.522	0.301	-	0.368	0.258	0.267	0.279
47	**427**	0.495	0.502	0.554	0.313	-	0.384	0.272	0.283	0.293
163	**590**	0.551	0.558	0.641	0.346		0.430	0.310	0.323	0.329
271	**861**	0.611	0.614	0.732				0.357		0.389

Interval (days)	Total Duration (days)	Fraction of Iodide Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.008	0.009	0.009	0.008	0.012	0.009	0.007	0.008	0.007
0.92	**1**	0.021	0.022	0.021	0.028	0.034	0.029	0.025	0.024	0.024
1	**2**	0.028	0.029	0.028	0.040	0.045	0.041	0.036	0.036	0.037
5	**7**	0.049	0.050	0.050	0.072	0.078	0.073	0.068	0.069	0.074
7	**14**	0.067	0.067	0.068	0.098	0.104	0.098	0.093	0.096	0.102
14	**28**	0.091	0.090	0.094	0.129	0.137	0.131	0.123	0.134	0.139
14	**42**	0.109	0.107	0.115	0.150	0.157	0.156	0.148	0.160	0.164
7	**49**	0.117	0.114	0.123	0.158	0.166	0.166	0.157	0.171	0.175
14	**63**	0.130	0.128	0.139	0.171	0.180	0.185	0.175	0.189	0.195
30	**93**	0.153	0.150	0.167	0.194	0.204	0.217	0.205	0.220	0.229
287	**380**	0.272	0.268	0.304	0.322	0.329	0.336	0.388	0.383	0.389
47	**427**	0.285	0.281	0.322	0.342	0.350	0.354	0.411	0.404	0.412
163	**590**	0.312	0.312	0.375	0.397	0.408	0.413	0.472	0.467	0.480
271	**861**							0.547		0.561

Interval (days)	Total Duration (days)	Fraction of Iodide Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.009	0.009	0.009	0.012	0.011	0.013	0.013	0.015	0.014
0.92	**1**	0.026	0.026	0.027	0.029	0.029	0.033	0.037	0.039	0.041
1	**2**	0.035	0.035	0.037	0.039	0.039	0.045	0.051	0.053	0.056
5	**7**	0.060	0.062	0.066	0.068	0.068	0.076	0.090	0.094	0.100
7	**14**	0.080	0.083	0.090	0.092	0.092	0.102	0.122	0.124	0.136
14	**28**	0.108	0.110	0.126	0.123	0.121	0.139	0.158	0.157	0.182
14	**42**	0.129	0.129	0.153	0.146	0.143	0.169	0.180	0.179	0.216
7	**49**	0.137	0.137	0.162	0.155	0.151	0.180	0.189	0.188	0.228
14	**63**	0.150	0.151	0.182	0.171	0.167	0.203	0.204	0.204	0.251
30	**93**	0.172	0.172	0.215	0.198	0.195	0.243	0.230	0.231	0.291
287	**380**	0.275	0.274	0.354	0.324	0.325	0.433	0.383	0.424	0.420
47	**427**	0.285	0.284	0.371	0.340	0.340	0.453	0.405	0.451	0.437
163	**590**	0.312	0.311	0.417	0.372	0.375	0.507	0.467	0.524	0.497
271	**861**									

Interval (days)	Total Duration (days)	Fraction of Iodide Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.008	0.009	0.011	0.020	0.021	0.019	0.012	0.013	0.015
0.92	**1**	0.025	0.028	0.030	0.062	0.062	0.064	0.032	0.036	0.035
1	**2**	0.037	0.040	0.041	0.087	0.087	0.091	0.043	0.047	0.046
5	**7**	0.069	0.073	0.070	0.139	0.152	0.163	0.070	0.077	0.075
7	**14**	0.096	0.099	0.093	0.189	0.204	0.224	0.092	0.099	0.096
14	**28**	0.128	0.133	0.124	0.253	0.268	0.301	0.118	0.125	0.125
14	**42**	0.148	0.155	0.146	0.296	0.311	0.353	0.136	0.144	0.145
7	**49**	0.156	0.164	0.155	0.311	0.328	0.370	0.143	0.152	0.152
14	**63**	0.171	0.180	0.172	0.341	0.357	0.407	0.156	0.165	0.168
30	**93**	0.195	0.208	0.203	0.388	0.406	0.466	0.176	0.185	0.197
287	**380**	0.326	0.361	0.334	0.583	0.612	0.643	0.268	0.289	0.319
47	**427**	0.347	0.382	0.353	0.603	0.630	0.662	0.278	0.300	0.334
163	**590**	0.410	0.437	0.420	0.654	0.676	0.713	0.308	0.327	0.379
271	**861**	0.490		0.513	0.720		0.767	0.348		0.436

Table B.25. Fraction of Technetium Leached versus Total Leach Time Archive Monoliths

Interval (days)	Total Duration (days)	Fraction of Technetium Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	**-**	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	0.0000	0.0000	0.0000	0.0000	-	0.0000	0.0011	0.0010	0.0015
0.92	**1**	0.0003	0.0002	0.0004	0.0002	-	0.0004	0.0021	0.0020	0.0031
1	**2**	0.0005	0.0004	0.0008	0.0004	-	0.0007	0.0027	0.0026	0.0041
5	**7**	0.0011	0.0011	0.0025	0.0010	-	0.0022	0.0043	0.0042	0.0080
7	**14**	0.0016	0.0017	0.0046	0.0015	-	0.0040	0.0056	0.0054	0.0121
14	**28**	0.0023	0.0026	0.0081	0.0022	-	0.0068	0.0069	0.0065	0.0184
14	**42**	0.0029	0.0032	0.0109	0.0027	-	0.0091	0.0078	0.0073	0.0231
7	**49**	0.0032	0.0035	0.0120	0.0029	-	0.0103	0.0083	0.0077	0.0251
14	**63**	0.0037	0.0041	0.0146	0.0033	-	0.0125	0.0090	0.0083	0.0289
30	**93**	0.0046	0.0051	0.0199	0.0039	-	0.0164	0.0100	0.0093	0.0357
287	**380**	0.0104	0.0112	0.0560	0.0066	-	0.0332	0.0146	0.0138	0.0666
47	**427**	0.0109	0.0117	0.0623	0.0069	-	0.0362	0.0154	0.0147	0.0704
163	**590**	0.0118	0.0126	0.0782	0.0074		0.0442	0.0193	0.0176	0.0799
271	**861**	0.0125	0.0133	0.1000				0.0243		0.0934

Interval (days)	Total Duration (days)	Fraction of Technetium Leached (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.0004	0.0003	0.0002	0.0002	0.0003	0.0001	0.0000	0.0000	0.0001
0.92	**1**	0.0011	0.0008	0.0010	0.0007	0.0008	0.0005	0.0002	0.0001	0.0005
1	**2**	0.0016	0.0012	0.0019	0.0010	0.0011	0.0008	0.0004	0.0003	0.0009
5	**7**	0.0028	0.0024	0.0050	0.0019	0.0020	0.0018	0.0010	0.0007	0.0019
7	**14**	0.0041	0.0036	0.0082	0.0027	0.0027	0.0032	0.0016	0.0012	0.0030
14	**28**	0.0056	0.0050	0.0131	0.0039	0.0039	0.0056	0.0024	0.0018	0.0044
14	**42**	0.0066	0.0060	0.0167	0.0048	0.0049	0.0079	0.0034	0.0023	0.0054
7	**49**	0.0071	0.0065	0.0184	0.0054	0.0055	0.0089	0.0038	0.0025	0.0059
14	**63**	0.0079	0.0072	0.0215	0.0063	0.0065	0.0111	0.0045	0.0029	0.0067
30	**93**	0.0090	0.0083	0.0270	0.0079	0.0083	0.0154	0.0056	0.0036	0.0084
287	**380**	0.0152	0.0134	0.0546	0.0235	0.0220	0.0559	0.0098	0.0064	0.0193
47	**427**	0.0158	0.0141	0.0580	0.0262	0.0243	0.0628	0.0101	0.0067	0.0203
163	**590**	0.0173	0.0156	0.0680	0.0325	0.0318	0.0795	0.0110	0.0074	0.0227
271	**861**							0.0118		0.0263

Interval (days)	**Total Duration (days)**	**Fraction of Technetium Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.0000	0.0000	0.0000	0.0005	0.0003	0.0005	0.0002	0.0002	0.0001
0.92	**1**	0.0002	0.0002	0.0004	0.0019	0.0016	0.0033	0.0009	0.0008	0.0020
1	**2**	0.0003	0.0004	0.0007	0.0028	0.0024	0.0049	0.0016	0.0014	0.0035
5	**7**	0.0008	0.0010	0.0021	0.0049	0.0043	0.0092	0.0032	0.0030	0.0079
7	**14**	0.0013	0.0015	0.0040	0.0070	0.0062	0.0131	0.0046	0.0043	0.0127
14	**28**	0.0018	0.0022	0.0069	0.0097	0.0086	0.0193	0.0063	0.0065	0.0194
14	**42**	0.0023	0.0027	0.0093	0.0120	0.0107	0.0244	0.0078	0.0082	0.0250
7	**49**	0.0025	0.0029	0.0102	0.0131	0.0116	0.0266	0.0086	0.0091	0.0275
14	**63**	0.0029	0.0033	0.0122	0.0150	0.0133	0.0309	0.0100	0.0105	0.0319
30	**93**	0.0035	0.0039	0.0156	0.0177	0.0160	0.0384	0.0123	0.0130	0.0403
287	**380**	0.0063	0.0067	0.0314	0.0293	0.0262	0.0756	0.0240	0.0272	0.0971
47	**427**	0.0066	0.0070	0.0340	0.0305	0.0275	0.0803	0.0258	0.0305	0.1055
163	**590**	0.0070	0.0074	0.0404	0.0325	0.0294	0.0907	0.0313	0.0379	0.1210
271	**861**									

Interval (days)	**Total Duration (days)**	**Fraction of Technetium Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	0.0001	0.0000	0.0002	0.0000	0.0000	0.0000	0.0000	0.0001	0.0000
0.92	**1**	0.0005	0.0006	0.0010	0.0001	0.0001	0.0002	0.0001	0.0002	0.0001
1	**2**	0.0009	0.0011	0.0016	0.0003	0.0002	0.0005	0.0002	0.0004	0.0003
5	**7**	0.0019	0.0023	0.0033	0.0006	0.0007	0.0028	0.0004	0.0007	0.0009
7	**14**	0.0029	0.0034	0.0050	0.0012	0.0014	0.0074	0.0008	0.0011	0.0016
14	**28**	0.0042	0.0047	0.0073	0.0022	0.0025	0.0151	0.0011	0.0015	0.0028
14	**42**	0.0051	0.0057	0.0093	0.0030	0.0035	0.0219	0.0014	0.0018	0.0038
7	**49**	0.0056	0.0061	0.0101	0.0034	0.0039	0.0249	0.0016	0.0019	0.0042
14	**63**	0.0064	0.0068	0.0118	0.0043	0.0048	0.0312	0.0019	0.0022	0.0050
30	**93**	0.0077	0.0083	0.0150	0.0059	0.0064	0.0425	0.0024	0.0027	0.0069
287	**380**	0.0145	0.0149	0.0369	0.0159	0.0169	0.0912	0.0049	0.0051	0.0191
47	**427**	0.0154	0.0158	0.0391	0.0168	0.0177	0.0973	0.0052	0.0054	0.0209
163	**590**	0.0178	0.0174	0.0449	0.0187	0.0195	0.1120	0.0059	0.0058	0.0267
271	**861**	0.0193		0.0580	0.0211		0.1320	0.0071		0.0369

Table B.26. Fraction of Chromium Leached versus Total Leach Time Archive Monoliths

Interval (days)	**Total Duration (days)**	**Fraction of Chromium Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	**-**	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	-	DIW	VZP	VZP	DIW
0.08	**0.08**	3.27E-06	3.09E-06	5.14E-05	1.98E-06	-	2.74E-05	8.50E-05	9.66E-05	1.46E-04
0.92	**1**	5.33E-06	1.23E-06	1.31E-04	9.89E-06	-	6.22E-05	3.53E-04	4.12E-04	5.09E-04
1	**2**	7.98E-06	6.42E-06	1.64E-04	1.20E-05	-	7.96E-05	4.30E-04	5.08E-04	6.11E-04
5	**7**	1.27E-05	1.97E-05	2.84E-04	1.32E-05	-	1.35E-04	5.35E-04	6.33E-04	8.49E-04
7	**14**	2.81E-05	3.51E-05	3.95E-04	1.01E-05	-	1.95E-04	5.63E-04	6.70E-04	1.02E-03
14	**28**	2.99E-05	3.84E-05	5.23E-04	2.76E-05	-	2.49E-04	6.23E-04	7.29E-04	1.18E-03
14	**42**	4.53E-05	5.38E-05	6.47E-04	4.52E-05	-	3.25E-04	6.82E-04	7.88E-04	1.38E-03
7	**49**	6.07E-05	6.91E-05	7.04E-04	6.27E-05	-	3.64E-04	7.41E-04	8.47E-04	1.46E-03
14	**63**	7.61E-05	8.45E-05	7.77E-04	8.02E-05	-	4.24E-04	8.00E-04	9.07E-04	1.65E-03
30	**93**	9.16E-05	9.98E-05	9.29E-04	9.78E-05	-	5.10E-04	8.59E-04	9.66E-04	1.91E-03
287	**380**	1.20E-04	1.20E-04	1.52E-03	1.07E-04	-	9.28E-04	8.80E-04	9.93E-04	3.03E-03
47	**427**	1.18E-04	1.17E-04	2.12E-03	1.06E-04	-	1.09E-03	8.82E-04	1.00E-03	3.28E-03
163	**590**	1.34E-04	1.33E-04	2.39E-03	9.74E-05		1.30E-03	5.92E-05	3.09E-07	4.22E-04
271	**861**	1.30E-04	1.31E-04	2.72E-03				9.10E-05		4.90E-04

Interval (days)	**Total Duration (days)**	**Fraction of Chromium Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.05E-04	7.79E-05	1.88E-04	1.50E-04	1.14E-04	2.47E-05	7.15E-06	3.96E-06	1.79E-05
0.92	**1**	2.89E-04	2.62E-04	5.73E-04	2.23E-04	1.92E-04	5.63E-05	1.28E-05	6.44E-06	5.27E-05
1	**2**	3.77E-04	3.49E-04	7.52E-04	2.59E-04	2.33E-04	8.11E-05	3.07E-05	2.43E-05	7.06E-05
5	**7**	5.62E-04	5.52E-04	1.15E-03	3.59E-04	3.32E-04	1.49E-04	3.51E-05	3.07E-05	1.28E-04
7	**14**	6.68E-04	6.70E-04	1.39E-03	4.11E-04	3.72E-04	1.96E-04	5.30E-05	4.85E-05	1.59E-04
14	**28**	7.55E-04	7.67E-04	1.58E-03	4.42E-04	3.89E-04	2.31E-04	7.09E-05	6.63E-05	1.77E-04
14	**42**	7.97E-04	8.09E-04	1.73E-03	4.45E-04	4.14E-04	2.82E-04	8.88E-05	8.42E-05	2.03E-04
7	**49**	7.98E-04	8.11E-04	1.80E-03	4.70E-04	4.39E-04	3.09E-04	1.07E-04	1.02E-04	2.21E-04
14	**63**	8.10E-04	8.20E-04	1.91E-03	4.95E-04	4.63E-04	1.45E-03	1.25E-04	1.20E-04	2.46E-04
30	**93**	8.34E-04	8.20E-04	2.07E-03	5.19E-04	4.88E-04	1.52E-03	1.42E-04	1.38E-04	2.70E-04
287	**380**	8.54E-04	8.43E-04	2.83E-03	5.44E-04	4.98E-04	1.95E-03	1.60E-04	1.55E-04	3.72E-04
47	**427**	8.52E-04	8.41E-04	2.99E-03	5.41E-04	5.00E-04	2.06E-03	1.57E-04	1.53E-04	3.95E-04
163	**590**	8.76E-04	8.64E-04	3.25E-03	5.66E-04	5.25E-04	2.22E-03	1.75E-04	1.71E-04	4.34E-04
271	**861**							1.74E-04		4.68E-04

Interval (days)	**Total Duration (days)**	**Fraction of Chromium Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	6.99E-06	1.11E-05	3.51E-05	5.47E-05	8.72E-05	1.28E-04	1.03E-05	7.81E-06	2.42E-05
0.92	**1**	3.38E-05	3.28E-05	9.88E-05	1.36E-04	2.34E-04	4.17E-04	2.46E-05	2.64E-05	6.02E-05
1	**2**	4.04E-05	4.20E-05	1.30E-04	1.54E-04	2.55E-04	4.83E-04	3.59E-05	3.51E-05	8.44E-05
5	**7**	6.14E-05	8.83E-05	3.89E-04	1.81E-04	2.77E-04	6.66E-04	5.58E-05	5.54E-05	1.94E-04
7	**14**	6.42E-05	1.03E-04	5.71E-04	1.80E-04	3.09E-04	7.98E-04	8.01E-05	7.97E-05	2.82E-04
14	**28**	6.43E-05	1.05E-04	7.85E-04	2.12E-04	3.42E-04	9.28E-04	8.29E-05	8.14E-05	3.67E-04
14	**42**	8.18E-05	1.23E-04	9.72E-04	2.45E-04	3.74E-04	1.07E-03	1.07E-04	1.06E-04	4.75E-04
7	**49**	9.93E-05	1.40E-04	1.03E-03	2.77E-04	4.06E-04	1.13E-03	1.32E-04	1.30E-04	5.19E-04
14	**63**	1.17E-04	1.46E-04	1.15E-03	3.09E-04	4.39E-04	1.24E-03	1.32E-04	1.54E-04	6.10E-04
30	**93**	1.34E-04	1.63E-04	1.29E-03	3.42E-04	4.71E-04	1.40E-03	1.57E-04	1.79E-04	7.49E-04
287	**380**	1.56E-04	1.84E-04	1.80E-03	3.42E-04	5.03E-04	2.01E-03	1.81E-04	1.83E-04	1.57E-03
47	**427**	1.57E-04	1.86E-04	1.93E-03	3.36E-04	4.98E-04	2.22E-03	1.82E-04	1.89E-04	1.72E-03
163	**590**	1.75E-04	2.04E-04	2.11E-03	3.68E-04	5.31E-04	2.45E-03	2.06E-04	2.02E-04	1.97E-03
271	**861**									

Interval (days)	**Total Duration (days)**	**Fraction of Chromium Leached (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	1.09E-05	5.31E-06	3.34E-05	8.65E-06	9.22E-06	2.00E-05	1.64E-05	2.13E-05	3.70E-05
0.92	**1**	1.28E-05	6.98E-06	6.67E-05	2.33E-05	2.35E-05	5.16E-05	4.75E-05	6.25E-05	1.01E-04
1	**2**	1.42E-05	8.19E-06	1.00E-04	2.89E-05	2.88E-05	6.70E-05	6.88E-05	8.82E-05	1.30E-04
5	**7**	2.13E-05	1.82E-05	1.62E-04	3.10E-05	3.66E-05	1.18E-04	1.21E-04	1.44E-04	2.55E-04
7	**14**	5.48E-05	5.14E-05	1.99E-04	4.64E-05	5.20E-05	1.62E-04	1.32E-04	1.65E-04	3.51E-04
14	**28**	8.83E-05	8.46E-05	2.32E-04	6.18E-05	5.32E-05	2.21E-04	1.60E-04	1.84E-04	4.56E-04
14	**42**	1.22E-04	1.18E-04	2.76E-04	7.73E-05	6.86E-05	2.91E-04	1.83E-04	2.07E-04	5.49E-04
7	**49**	1.55E-04	1.51E-04	3.10E-04	9.27E-05	8.41E-05	3.21E-04	2.07E-04	2.31E-04	5.94E-04
14	**63**	1.89E-04	1.84E-04	3.64E-04	1.08E-04	9.95E-05	3.90E-04	2.09E-04	2.54E-04	6.73E-04
30	**93**	2.22E-04	2.18E-04	4.34E-04	1.24E-04	1.15E-04	5.12E-04	2.33E-04	2.78E-04	7.96E-04
287	**380**	2.56E-04	2.51E-04	7.54E-04	1.39E-04	1.31E-04	1.08E-03	2.55E-04	3.23E-04	1.32E-03
47	**427**	2.51E-04	2.67E-04	8.23E-04	1.40E-04	1.32E-04	1.18E-03	2.54E-04	3.21E-04	1.43E-03
163	**590**	3.06E-04	3.01E-04	9.52E-04	1.55E-04	1.48E-04	1.35E-03	2.78E-04	3.44E-04	1.62E-03
271	**861**	3.18E-04		1.11E-03	1.63E-04		1.57E-03	2.82E-04		1.88E-03

Table B.27. pH of Archive Eluates

Interval (days)	Total Duration (days)	pH of Archive Eluates (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	—	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	—	DIW	VZP	VZP	DIW
0.08	**0.08**	10.3	10.3	11.6	9.5	—	10.8	9.2	9.2	10.5
0.92	**1**	10.8	10.7	12.0	10.3	—	11.7	9.8	9.7	11.2
1	**2**	10.6	10.6	11.9	10.3	—	11.6	9.5	9.5	11.0
5	**7**	11.2	11.2	12.1	10.5	—	11.8	10.2	10.2	11.3
7	**14**	10.8	10.7	11.9	10.3	—	11.7	10.4	10.4	11.2
14	**28**	11.0	11.1	12.0	10.5	—	11.9	10.5	10.5	11.4
14	**42**	10.6	10.6	11.9	10.3	—	11.8	10.3	10.4	11.3
7	**49**	10.3	10.3	11.6	10.2	—	11.5	10.2	10.3	10.9
14	**63**	10.5	10.4	11.8	10.4	—	11.7	10.4	10.4	11.2
30	**93**	10.7	10.7	12.0	10.4	—	11.8	10.3	10.2	11.3
287	**380**	12.3	12.3	12.5	12.0	—	12.4	10.0	10.1	11.4
47	**427**	10.5	10.4	12.0	10.3	—	11.8	9.4	9.7	10.9
163	**590**	10.5	10.7	12	10.3		11.9	8.44	8.98	10.7
271	**861**	10.4	10.4	12				8.4		10.7

Interval (days)	Total Duration (days)	pH of Archive Eluates (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	**10Arch-b**	**10Arch-c**	**13Arch-a**	**13Arch-b**	**13Arch-c**	**14Arch-a**	**14Arch-b**	**14Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	9.8	9.8	11.0	9.6	9.7	10.9	9.5	9.5	10.9
0.92	**1**	10.5	10.4	11.8	10.5	10.6	11.6	10.4	10.4	11.4
1	**2**	10.4	10.4	11.5	10.4	10.4	11.4	10.2	10.2	11.2
5	**7**	10.5	10.5	11.8	10.6	10.6	11.7	10.5	10.5	11.7
7	**14**	10.4	10.4	11.7	10.4	10.4	11.6	10.4	10.3	11.5
14	**28**	10.5	10.5	11.8	10.4	10.5	11.7	10.4	10.5	11.6
14	**42**	10.4	10.4	11.7	10.4	10.3	11.5	10.3	10.3	11.5
7	**49**	10.3	10.3	11.4	10.3	10.3	11.3	10.2	10.1	11.2
14	**63**	10.3	10.4	11.6	10.3	10.3	11.4	10.3	10.3	11.4
30	**93**	10.3	10.3	11.7	10.3	10.3	11.5	10.3	10.2	11.5
287	**380**	11.7	11.7	12.3	10.6	10.1	11.8	10.4	10.7	12.0
47	**427**	10.2	10.3	11.7	10.2	10.2	11.4	10.2	10.2	11.6
163	**590**	10.3	10.1	11.8	10.2	9.88	11.3	9.99	9.85	11.5
271	**861**							9.94		11.5

Interval (days)	**Total Duration (days)**	**pH of Archive Eluates (unitless)**								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	**15Arch-b**	**15Arch-c**	**16Arch-a**	**16Arch-b**	**16Arch-c**	**17Arch-a**	**17Arch-b**	**17Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	9.86	9.94	11.1	9.8	9.83	11	9.75	9.8	11.0
0.92	**1**	10.5	10.5	11.8	10.5	10.5	11.6	10.6	10.6	11.7
1	**2**	10.4	10.4	11.5	10.4	10.4	11.4	10.5	10.5	11.5
5	**7**	10.6	10.5	11.9	10.5	10.5	11.7	10.6	10.5	11.7
7	**14**	10.4	10.4	11.7	10.4	10.4	11.6	10.5	10.5	11.7
14	**28**	10.5	10.5	11.8	10.4	10.4	11.7	10.4	10.4	11.7
14	**42**	10.4	10.4	11.8	10.3	10.3	11.6	10.3	10.4	11.6
7	**49**	10.3	10.4	11.4	10.3	10.1	11.3	10.2	10.3	11.3
14	**63**	10.4	10.4	11.7	10.3	10.3	11.5	10.3	10.3	11.5
30	**93**	10.4	10.4	11.8	10.3	10.2	11.7	10.3	10.3	11.6
287	**380**	11.9	12	12.4	11	11.1	12.2	10.6	10.7	11.8
47	**427**	10.3	10.4	11.8	10.2	10.1	11.6	10.2	10.3	11.5
163	**590**	10.3	10.4	11.9	10.2	10.0	11.8	10.3	9.9	11.3
271	**861**									

Interval (days)	Total Duration (days)	pH of Archive Eluates (unitless)								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	9.6	9.6	10.9	10.0	10.0	11.2	10.0	10.1	11.4
0.92	**1**	10.3	10.4	11.4	10.6	10.5	11.7	10.5	10.6	11.8
1	**2**	10.2	10.3	11.3	10.5	10.5	11.6	10.5	10.5	11.7
5	**7**	10.5	10.4	11.6	10.5	10.5	11.8	10.6	10.5	11.9
7	**14**	10.4	10.4	11.4	10.4	10.5	11.7	10.4	10.4	11.8
14	**28**	10.4	10.4	11.5	10.5	10.5	11.8	10.5	10.6	11.8
14	**42**	10.3	10.3	11.4	10.4	10.3	11.7	10.4	10.4	11.8
7	**49**	10.2	10.1	11.1	10.1	10.2	11.5	10.3	10.3	11.4
14	**63**	10.3	10.3	11.4	10.4	10.3	11.6	10.4	10.4	11.6
30	**93**	10.3	10.2	11.5	10.3	10.3	11.7	10.4	10.3	11.8
287	**380**	10.5	10.4	11.8	11.3	11.6	12.1	11.6	11.8	12.2
47	**427**	10.1	9.8	11.4	10.2	10.3	11.7	10.3	10.3	11.7
163	**590**	10.3	9.93	11.2	9.88	10.4	11.7	10.3	10.2	11.8
271	**861**	10.1		11.1	10.1		11.6	10.1		11.8

Table B.28. Electrical Conductivity of Archive Eluates

Interval (days)	Total Duration (days)	EC of Archive Eluates (mS/cm)								
		Mix Number and VZP Duplicates (a,b)								
		3Arch-a	**3Arch-b**	**3Arch-c**	**5Arch-a**	—	**5Arch-c**	**8Arch-a**	**8Arch-b**	**8Arch-c**
Eluent		VZP	VZP	DIW	VZP	—	—	VZP	VZP	DIW
0.08	**0.08**	4.37	4.42	1.69	4.24	—	0.89	4.23	4.15	0.794
0.92	**1**	4.88	4.99	3.18	4.57	—	2.09	4.07	4.13	0.795
1	**2**	4.44	4.44	1.92	4.26	—	1.32	3.95	3.95	0.478
5	**7**	5.71	5.80	4.56	4.96	—	3.13	4.37	4.51	1.35
7	**14**	5.12	5.17	3.81	4.84	—	2.82	4.47	4.5	1.29
14	**28**	5.65	5.69	4.77	5.12	—	3.68	4.65	4.67	1.69
14	**42**	4.21	4.20	3.19	4.01	—	2.36	3.68	3.69	1.08
7	**49**	3.66	3.66	1.44	3.57	—	1.18	3.46	3.47	0.497
14	**63**	4.10	4.07	2.39	3.83	—	1.81	3.64	3.65	0.837
30	**93**	4.49	4.51	3.76	4.10	—	2.80	3.76	3.79	1.22
287	**380**	12.90	12.10	14.10	8.97	—	10.90	5.69	5.76	3.93
47	**427**	3.86	3.81	3.45	3.65	—	2.37	3.51	3.54	0.757
163	**590**	4.92	4.95	5.65	4.56		4.21	4.16	4.20	1.45
271	**861**	5.11	5.00	5.88				4.56		2.11

Interval (days)	Total Duration (days)	EC of Archive Eluates (mS/cm)								
		Mix Number and VZP Duplicates (a,b)								
		10Arch-a	10Arch-b	10Arch-c	13Arch-a	13Arch-b	13Arch-c	14Arch-a	14Arch-b	14Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	4.12	4.13	0.897	4.01	4.1	0.574	4.02	4.06	0.516
0.92	**1**	4.24	4.19	1.54	4.39	4.45	1.29	4.39	4.34	1.21
1	**2**	4.03	4.08	0.912	4.14	4.13	0.766	4.16	4.16	0.863
5	**7**	4.64	4.66	2.35	4.66	4.62	1.95	4.96	4.99	2.16
7	**14**	4.54	4.56	2.1	4.56	4.56	1.69	4.72	4.78	1.89
14	**28**	4.75	4.72	2.74	4.57	4.6	2.08	4.79	5.05	2.38
14	**42**	3.73	3.72	1.77	3.63	3.65	1.39	3.92	3.93	1.53
7	**49**	3.48	3.48	0.869	3.44	3.46	0.707	3.51	3.52	0.737
14	**63**	3.69	3.69	1.39	3.61	3.62	1.05	3.8	3.82	1.17
30	**93**	3.89	3.89	2.15	3.73	3.75	1.53	4.06	4.09	1.82
287	**380**	7.28	7.15	8.45	6.01	5.95	4.76	7.62	7.55	7.39
47	**427**	3.56	3.55	1.93	3.63	3.64	1.27	3.77	3.76	1.69
163	**590**	4.17	4.21	3.12	4.46	4.49	1.96	4.95	5.04	2.91
271	**861**							5.37		3.58

Interval (days)	Total Duration (days)	EC of Archive Eluates (mS/cm)								
		Mix Number and VZP Duplicates (a,b)								
		15Arch-a	15Arch-b	15Arch-c	16Arch-a	16Arch-b	16Arch-c	17Arch-a	17Arch-b	17Arch-c
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	4.22	4.23	1.00	4.42	4.35	1.22	4.18	4.20	0.70
0.92	**1**	4.59	4.57	1.97	4.65	4.64	1.93	4.50	4.51	1.62
1	**2**	4.18	4.21	1.20	4.30	4.32	1.17	4.22	4.23	1.02
5	**7**	5.06	5.10	3.12	5.25	5.20	2.90	4.96	4.93	2.50
7	**14**	4.88	4.89	2.69	5.04	5.01	2.51	4.77	4.70	2.23
14	**28**	5.08	5.11	3.64	5.46	5.28	3.28	4.81	4.75	2.65
14	**42**	4.02	4.01	2.46	4.11	4.08	2.25	3.73	3.73	1.72
7	**49**	3.60	3.60	1.02	3.65	3.60	1.00	3.49	3.49	0.80
14	**63**	3.89	3.90	1.89	3.99	3.99	1.85	3.72	3.71	1.26
30	**93**	4.16	4.15	2.87	4.33	4.35	2.88	3.85	3.86	1.80
287	**380**	8.68	8.88	10.70	8.43	8.67	11.20	6.56	7.17	5.43
47	**427**	3.64	3.63	2.30	3.77	3.79	2.23	3.70	3.84	1.50
163	**590**	4.47	4.47	4.08	4.71	4.83	4.03	4.69	4.93	2.05
271	**861**									

Interval (days)	**Total Duration (days)**	**EC of Archive Eluates (mS/cm)**								
		Mix Number and VZP Duplicates (a,b)								
		18Arch-a	**18Arch-b**	**18Arch-c**	**21Arch-a**	**21Arch-b**	**21Arch-c**	**24Arch-a**	**24Arch-b**	**24Arch-c**
Eluent		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
0.08	**0.08**	4.12	4.18	0.85	4.55	4.59	1.32	4.23	4.29	1.21
0.92	**1**	4.58	4.68	1.50	5.39	5.26	2.92	4.55	4.64	2.00
1	**2**	4.28	4.31	0.92	4.69	4.61	1.85	4.19	4.18	1.23
5	**7**	5.24	5.21	2.17	5.87	6.19	4.59	4.84	4.87	2.86
7	**14**	4.99	5.01	1.88	5.81	5.89	4.07	4.61	4.68	2.40
14	**28**	5.20	5.29	2.42	6.36	6.34	5.04	4.81	4.85	2.97
14	**42**	3.96	4.03	1.58	4.59	4.60	3.17	3.75	3.77	1.89
7	**49**	3.56	3.54	0.71	3.77	3.81	1.37	3.50	3.50	0.92
14	**63**	3.89	3.96	1.24	4.33	4.34	2.31	3.73	3.74	1.44
30	**93**	4.11	4.24	1.92	4.79	4.84	3.38	3.85	3.86	2.21
287	**380**	8.11	8.54	7.35	9.64	10.20	9.70	6.69	7.15	7.21
47	**427**	3.90	3.87	1.55	3.84	3.81	2.26	3.49	3.46	1.76
163	**590**	5.46	5.22	3.05	4.96	4.82	3.08	4.16	4.01	2.89
271	**861**	6.17		4.31	5.45		3.41	4.4		3.54

B.3 I-Spike Leach Tests

Table B.29. Na Observed Diffusion Coefficient (cm^2/s) I-spike Leach Tests

		Na in I-spiked Monoliths Observed Diffusion Coefficient (cm^2/s)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	4.28E-08	6.34E-08	5.32E-08	4.92E-08	5.65E-08	6.03E-08	6.84E-08	7.18E-08	7.32E-08	7.08E-08	5.98E-08	5.24E-08
1	1.60E-08	1.48E-08	9.44E-09	1.35E-08	1.00E-08	1.42E-08	1.39E-08	1.52E-08	2.04E-08	1.54E-08	2.82E-08	1.23E-08
2	4.61E-09	7.99E-09	6.87E-09	1.22E-08	8.03E-09	6.02E-09	9.09E-09	1.11E-08	1.20E-08	6.65E-09	6.87E-09	7.24E-09
7	9.38E-09	9.51E-09	9.86E-09	1.03E-08	1.11E-08	1.07E-08	9.58E-09	9.65E-09	1.07E-08	1.18E-08	1.14E-08	9.71E-09
14	5.08E-09	4.82E-09	6.20E-09	6.04E-09	5.82E-09	5.65E-09	5.63E-09	6.00E-09	6.11E-09	6.71E-09	3.81E-09	3.09E-09
28	4.25E-09	3.75E-09	4.05E-09	4.02E-09	3.59E-09	4.10E-09	4.65E-09	4.72E-09	4.38E-09	4.39E-09	4.78E-09	4.30E-09
42	3.25E-09	3.24E-09	3.29E-09	3.59E-09	3.75E-09	3.51E-09	4.25E-09	4.25E-09	4.31E-09	3.96E-09	4.46E-09	4.29E-09
49	1.81E-09	1.93E-09	2.63E-09	2.18E-09	1.51E-09	2.15E-09	4.00E-09	4.28E-09	4.24E-09	3.53E-09	4.97E-09	4.35E-09
63	1.57E-09	2.60E-09	1.92E-09	1.84E-09	2.18E-09	1.82E-09	3.60E-09	3.22E-09	2.81E-09	3.05E-09	2.44E-09	2.93E-09
100	1.77E-09	1.70E-09	1.85E-09	1.69E-09	2.05E-09	2.22E-09	2.55E-09	2.69E-09	2.88E-09	2.45E-09	2.27E-09	2.73E-09
369	1.02E-09	1.01E-09	1.18E-09	1.16E-09	1.17E-09	1.07E-09	1.65E-09	1.62E-09	1.58E-09	1.56E-09	1.59E-09	1.69E-09
414	1.40E-09	1.24E-09	1.41E-09	1.35E-09	1.23E-09	1.38E-09	3.82E-09	4.19E-09	3.94E-09	4.09E-09	3.57E-09	3.67E-09
569	5.34E-10	4.34E-10	5.35E-10	5.30E-10	4.61E-10	4.97E-10	1.18E-09	1.31E-09	1.26E-09	1.27E-09	1.19E-09	1.25E-09

Table B.30. Nitrate Observed Diffusion Coefficient (cm^2/s) I-spike Leach Tests

		Nitrate in I-spiked Monoliths Observed Diffusion Coefficient (cm^2/s)											
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	3.70E-08	4.40E-08	3.17E-08	4.08E-08	3.74E-08	3.57E-08	5.46E-08	5.58E-08	6.87E-08	6.34E-08	6.29E-08	4.73E-08
1	1.57E-08	1.61E-08	1.08E-08	1.53E-08	1.08E-08	1.28E-08	1.53E-08	1.47E-08	2.35E-08	1.74E-08	2.55E-08	1.17E-08
2	6.42E-09	1.19E-08	6.92E-09	9.91E-09	2.05E-08	7.63E-09	9.10E-09	8.84E-09	8.40E-09	7.19E-09	9.62E-09	7.11E-09
7	8.96E-09	9.13E-09	8.11E-09	8.61E-09	8.65E-09	7.87E-09	9.38E-09	9.11E-09	9.20E-09	9.86E-09	9.81E-09	9.29E-09
14	4.36E-09	4.19E-09	3.76E-09	4.11E-09	4.30E-09	4.32E-09	5.15E-09	5.39E-09	5.28E-09	5.31E-09	3.21E-09	2.24E-09
28	3.48E-09	3.02E-09	2.72E-09	2.51E-09	2.63E-09	2.46E-09	4.33E-09	4.46E-09	3.30E-09	3.58E-09	3.38E-09	3.74E-09
42	2.66E-09	2.75E-09	2.65E-09	2.42E-09	2.44E-09	2.33E-09	3.70E-09	3.78E-09	3.56E-09	3.57E-09	3.79E-09	4.30E-09
49	2.67E-09	2.37E-09	2.39E-09	2.47E-09	2.21E-09	2.29E-09	3.47E-09	3.63E-09	3.58E-09	3.45E-09	3.61E-09	3.97E-09
63	2.18E-09	1.94E-09	2.13E-09	1.94E-09	1.92E-09	1.91E-09	3.46E-09	3.35E-09	3.54E-09	3.30E-09	2.55E-09	3.02E-09
100	1.94E-09	1.81E-09	2.08E-09	3.53E-09	2.02E-09	1.89E-09	3.63E-09	3.45E-09	3.26E-09	4.61E-09	3.18E-09	3.63E-09
369	1.98E-09	1.98E-09	2.14E-09	2.25E-09	2.17E-09	1.99E-09	3.55E-09	3.57E-09	3.66E-09	3.51E-09	3.40E-09	3.56E-09
414	1.49E-09	1.39E-09	1.36E-09	1.40E-09	1.42E-09	1.39E-09	3.16E-09	3.53E-09	3.48E-09	3.49E-09	3.17E-09	3.26E-09
569	4.10E-10	4.00E-10	6.90E-10	6.55E-10	6.13E-10	6.37E-10	1.83E-09	2.17E-09	2.59E-09	2.68E-09	2.31E-09	1.84E-09

Table B.31. Nitrite Observed Diffusion Coefficient (cm^2/s) I-spike Leach Tests

		Nitrite in I-spiked Monoliths Observed Diffusion Coefficient (cm^2/s)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	3.22E-08	3.75E-08	2.83E-08	3.51E-08	3.29E-08	3.09E-08	3.75E-08	3.84E-08	4.56E-08	4.30E-08	4.22E-08	3.28E-08
1	1.02E-08	1.02E-08	7.15E-09	1.01E-08	7.57E-09	8.75E-09	9.48E-09	9.34E-09	1.43E-08	1.13E-08	1.60E-08	7.59E-09
2	4.57E-09	9.05E-09	5.40E-09	6.92E-09	6.39E-09	6.14E-09	6.50E-09	6.30E-09	6.13E-09	5.28E-09	6.91E-09	5.11E-09
7	5.33E-09	5.71E-09	5.25E-09	5.28E-09	5.57E-09	4.89E-09	6.00E-09	5.77E-09	5.76E-09	6.24E-09	6.21E-09	5.86E-09
14	3.07E-09	2.76E-09	2.87E-09	3.03E-09	3.06E-09	3.06E-09	3.52E-09	3.56E-09	3.59E-09	3.49E-09	2.15E-09	1.54E-09
28	2.19E-09	1.94E-09	1.87E-09	1.79E-09	1.80E-09	1.56E-09	2.82E-09	2.80E-09	2.16E-09	2.24E-09	2.15E-09	2.33E-09
42	1.96E-09	1.93E-09	1.85E-09	1.75E-09	1.80E-09	1.66E-09	2.42E-09	2.53E-09	2.32E-09	2.38E-09	2.53E-09	2.81E-09
49	1.70E-09	1.85E-09	1.84E-09	1.80E-09	1.65E-09	1.85E-09	2.42E-09	2.51E-09	2.47E-09	2.42E-09	2.57E-09	2.74E-09
63	1.53E-09	1.48E-09	1.70E-09	1.48E-09	1.49E-09	1.47E-09	2.21E-09	2.12E-09	2.35E-09	2.21E-09	1.73E-09	2.00E-09
100	1.18E-09	1.14E-09	1.31E-09	1.28E-09	1.24E-09	1.17E-09	2.12E-09	2.01E-09	1.88E-09	1.82E-09	1.84E-09	2.11E-09
369	1.38E-09	1.31E-09	1.37E-09	1.44E-09	1.49E-09	1.30E-09	2.36E-09	2.19E-09	2.21E-09	2.17E-09	2.09E-09	2.17E-09
414	4.64E-10	4.56E-10	3.95E-10	4.25E-10	4.05E-10	3.87E-10	1.71E-09	1.92E-09	1.88E-09	1.91E-09	1.74E-09	1.77E-09
569	6.80E-10	6.60E-10	8.92E-10	8.61E-10	8.31E-10	8.39E-10	9.73E-10	1.14E-09	1.33E-09	1.39E-09	1.21E-09	9.86E-10

Table B.32. Iodide in Iodide-spiked Monoliths Observed Diffusion Coefficient (cm^2/s)

		Iodide in I-spiked Monoliths Observed Diffusion Coefficient (cm^2/s)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	7.48E-08	9.76E-08	6.45E-08	8.03E-08	8.65E-08	7.85E-08	8.74E-08	7.35E-08	1.06E-07	9.55E-08	1.08E-07	7.95E-08
1	2.20E-08	2.10E-08	1.61E-08	2.29E-08	1.75E-08	2.32E-08	2.05E-08	1.93E-08	3.12E-08	2.41E-08	3.71E-08	1.83E-08
2	1.02E-08	1.69E-08	1.05E-08	1.55E-08	1.57E-08	1.14E-08	1.26E-08	1.16E-08	1.23E-08	1.06E-08	1.52E-08	1.14E-08
7	1.28E-08	1.21E-08	1.14E-08	1.20E-08	1.19E-08	1.19E-08	1.28E-08	1.18E-08	1.36E-08	1.38E-08	1.49E-08	1.42E-08
14	6.23E-09	5.48E-09	5.01E-09	5.38E-09	6.96E-09	6.24E-09	6.51E-09	6.54E-09	6.16E-09	7.08E-09	4.42E-09	3.26E-09
28	4.31E-09	3.75E-09	3.36E-09	3.23E-09	3.42E-09	3.41E-09	5.00E-09	5.57E-09	4.34E-09	4.58E-09	4.89E-09	5.22E-09
42	4.32E-09	4.12E-09	3.38E-09	3.22E-09	3.69E-09	3.66E-09	4.91E-09	4.66E-09	4.78E-09	4.75E-09	5.39E-09	6.08E-09
49	6.28E-09	6.17E-09	3.53E-09	3.37E-09	3.49E-09	3.64E-09	4.55E-09	4.36E-09	5.00E-09	4.68E-09	5.25E-09	5.83E-09
63	4.04E-09	3.69E-09	3.03E-09	2.96E-09	3.14E-09	2.82E-09	4.32E-09	4.16E-09	4.88E-09	4.25E-09	3.65E-09	4.41E-09
100	4.41E-09	4.10E-09	3.33E-09	3.03E-09	3.23E-09	2.97E-09	5.89E-09	5.55E-09	4.60E-09	4.25E-09	4.68E-09	5.29E-09
369	3.31E-09	3.49E-09	3.20E-09	3.78E-09	3.34E-09	3.10E-09	4.94E-09	4.76E-09	5.21E-09	4.72E-09	4.74E-09	4.73E-09
414	4.94E-09	4.98E-09	2.31E-09	2.24E-09	2.79E-09	2.91E-09	5.88E-09	6.61E-09	6.11E-09	5.37E-09	5.16E-09	5.20E-09
569	3.36E-08	3.71E-08	2.77E-09	3.02E-09	1.73E-09	1.95E-09	5.20E-09	6.25E-09	4.01E-09	4.82E-09	4.13E-09	4.45E-09

Table B.33. Na Fraction Leached in I-spike Leach Tests

		Na in I-spiked Monoliths Fraction Leached (unitless))									
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	0.020	0.024	0.022	0.021	0.023	0.024	0.025	0.026	0.026	0.026	0.024	0.022
1	0.050	0.054	0.046	0.049	0.047	0.052	0.053	0.055	0.060	0.055	0.064	0.048
2	0.060	0.066	0.057	0.064	0.059	0.063	0.066	0.070	0.075	0.067	0.075	0.060
7	0.100	0.106	0.098	0.106	0.103	0.106	0.107	0.110	0.118	0.111	0.119	0.100
14	0.126	0.132	0.127	0.134	0.131	0.134	0.134	0.139	0.146	0.141	0.142	0.121
28	0.160	0.164	0.160	0.166	0.162	0.168	0.169	0.174	0.180	0.176	0.178	0.154
42	0.182	0.187	0.183	0.190	0.186	0.191	0.195	0.200	0.206	0.201	0.205	0.180
49	0.190	0.194	0.192	0.198	0.193	0.200	0.206	0.211	0.217	0.211	0.217	0.192
63	0.202	0.210	0.206	0.211	0.207	0.213	0.225	0.229	0.234	0.229	0.232	0.208
100	0.231	0.239	0.236	0.240	0.239	0.246	0.259	0.265	0.271	0.263	0.265	0.244
369	0.329	0.338	0.342	0.344	0.344	0.347	0.384	0.389	0.392	0.385	0.388	0.370
414	0.344	0.351	0.357	0.358	0.357	0.362	0.408	0.413	0.416	0.409	0.411	0.393
569	0.371	0.376	0.384	0.385	0.383	0.388	0.448	0.456	0.458	0.451	0.452	0.434

Table B.34. Nitrate Fraction Leached in I-spike Leach Tests

		Nitrate in I-spiked Monoliths Fraction Leached (unitless)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	0.019	0.020	0.017	0.019	0.019	0.018	0.022	0.023	0.025	0.024	0.024	0.021
1	0.048	0.051	0.042	0.049	0.043	0.046	0.052	0.052	0.061	0.056	0.062	0.046
2	0.060	0.066	0.054	0.062	0.063	0.058	0.065	0.065	0.074	0.067	0.076	0.058
7	0.099	0.106	0.091	0.100	0.102	0.095	0.105	0.104	0.114	0.109	0.117	0.098
14	0.123	0.129	0.114	0.124	0.126	0.119	0.131	0.131	0.140	0.135	0.138	0.115
28	0.153	0.158	0.141	0.149	0.152	0.145	0.165	0.165	0.170	0.166	0.168	0.146
42	0.174	0.179	0.161	0.169	0.172	0.164	0.189	0.190	0.193	0.190	0.192	0.172
49	0.183	0.188	0.170	0.177	0.180	0.173	0.199	0.200	0.204	0.200	0.203	0.183
63	0.198	0.201	0.184	0.191	0.194	0.186	0.218	0.218	0.222	0.218	0.219	0.200
100	0.228	0.231	0.216	0.232	0.225	0.217	0.259	0.259	0.261	0.265	0.258	0.241
369	0.366	0.369	0.359	0.377	0.368	0.355	0.442	0.443	0.447	0.448	0.438	0.424
414	0.380	0.383	0.373	0.391	0.383	0.370	0.463	0.465	0.469	0.470	0.459	0.446
569	0.404	0.407	0.404	0.421	0.412	0.400	0.513	0.520	0.529	0.531	0.516	0.496

Table B.35. Nitrite Fraction Leached in I-spike Leach Tests

		Nitrite in I-spiked Monoliths Fraction Leached (unitless)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	0.017	0.019	0.016	0.018	0.018	0.017	0.019	0.019	0.021	0.020	0.020	0.017
1	0.041	0.043	0.037	0.042	0.038	0.040	0.042	0.042	0.049	0.045	0.050	0.038
2	0.051	0.056	0.047	0.053	0.049	0.051	0.053	0.053	0.060	0.055	0.062	0.048
7	0.081	0.088	0.077	0.083	0.080	0.080	0.085	0.084	0.091	0.088	0.094	0.079
14	0.101	0.107	0.097	0.103	0.100	0.100	0.106	0.106	0.113	0.110	0.111	0.094
28	0.126	0.130	0.119	0.125	0.122	0.121	0.134	0.133	0.137	0.134	0.135	0.118
42	0.143	0.147	0.136	0.141	0.139	0.137	0.153	0.153	0.156	0.154	0.155	0.139
49	0.150	0.155	0.144	0.149	0.146	0.145	0.162	0.162	0.164	0.162	0.164	0.148
63	0.163	0.167	0.157	0.161	0.158	0.157	0.177	0.176	0.179	0.177	0.177	0.162
100	0.186	0.191	0.182	0.185	0.183	0.181	0.208	0.207	0.209	0.206	0.207	0.194
369	0.301	0.303	0.296	0.301	0.301	0.293	0.357	0.351	0.353	0.350	0.348	0.336
414	0.309	0.311	0.304	0.309	0.309	0.300	0.373	0.368	0.370	0.367	0.364	0.352
569	0.340	0.341	0.339	0.343	0.343	0.335	0.410	0.408	0.413	0.410	0.405	0.389

Table B.36. Iodide in Iodide-spiked Monoliths Fraction Leached

		Iodide in I-spiked Monoliths Fraction Leached (unitless)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	0.026	0.030	0.025	0.027	0.028	0.027	0.028	0.026	0.031	0.030	0.032	0.027
1	0.062	0.065	0.055	0.063	0.060	0.064	0.062	0.059	0.073	0.067	0.078	0.059
2	0.076	0.083	0.069	0.080	0.077	0.079	0.078	0.074	0.088	0.081	0.095	0.074
7	0.123	0.129	0.114	0.125	0.122	0.124	0.125	0.119	0.136	0.130	0.145	0.123
14	0.151	0.156	0.140	0.152	0.153	0.153	0.154	0.148	0.165	0.160	0.170	0.143
28	0.185	0.188	0.170	0.181	0.183	0.184	0.190	0.187	0.199	0.195	0.206	0.181
42	0.212	0.214	0.193	0.203	0.207	0.208	0.218	0.214	0.226	0.223	0.235	0.211
49	0.225	0.227	0.203	0.213	0.218	0.219	0.230	0.226	0.238	0.235	0.248	0.225
63	0.245	0.246	0.221	0.230	0.235	0.236	0.250	0.246	0.260	0.255	0.267	0.245
100	0.291	0.291	0.261	0.268	0.274	0.274	0.303	0.297	0.307	0.300	0.314	0.295
369	0.469	0.474	0.436	0.456	0.452	0.447	0.519	0.509	0.528	0.512	0.527	0.506
414	0.495	0.501	0.454	0.474	0.472	0.467	0.548	0.540	0.557	0.540	0.554	0.533
569	0.711	0.728	0.516	0.538	0.521	0.520	0.633	0.633	0.631	0.622	0.630	0.611

Table B.37. pH of Eluates from I-spike Leach Tests

		pH in Eluates from I-spiked Monoliths (unitless)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	10.40	10.50	10.60	10.60	10.50	10.50	11.80	11.90	12.00	12.00	11.80	11.80
1	11.20	11.30	11.10	11.40	11.40	11.40	11.80	11.90	12.00	12.20	11.90	12.00
2	10.50	10.60	10.60	10.60	10.60	10.60	11.80	11.80	11.90	11.90	11.90	11.90
7	11.70	11.70	11.80	11.80	11.80	11.80	12.10	12.20	12.20	12.20	12.20	12.20
14	11.50	11.50	11.60	11.50	11.50	11.60	12.20	12.20	12.20	12.20	12.10	12.20
28	11.60	11.60	11.60	11.50	11.60	11.60	12.10	12.10	12.10	12.10	12.10	12.10
42	10.90	10.90	10.90	10.80	10.80	10.90	12.00	12.00	12.00	12.00	12.00	12.00
49	10.40	10.40	10.40	10.40	10.40	10.40	11.70	11.70	11.70	11.70	11.70	11.60
63	10.40	10.40	10.30	10.40	10.40	10.40	11.80	11.80	11.80	11.80	11.70	11.70
100	10.60	10.90	10.80	10.80	10.80	10.80	12.00	12.00	12.00	12.00	12.00	12.00
369	12.10	12.10	12.20	12.10	12.10	12.10	12.50	12.40	12.40	12.50	12.40	12.40
414	10.30	10.30	10.30	10.20	10.30	10.30	11.70	11.70	11.70	11.70	11.70	11.70
569	10.40	10.40	10.40	10.30	10.40	10.40	11.90	11.90	11.90	11.80	12.00	11.90

Table B.38. Electrical Conductivity in Eluates from Iodide in Iodide-spiked Monoliths

		EC in I-spiked Monoliths Eluates (mS/cm)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Eluent	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**VZP**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**	**DIW**
Sample ID	**CS-T1-VZ-1**	**CS-T1-VZ-2**	**CS-T2-VZ-1**	**CS-T2-VZ-2**	**CS-T3-VZ-1**	**CS-T3-VZ-2**	**CS-T1-DI-3**	**CS-T1-DI-4**	**CS-T2-DI-3**	**CS-T2-DI-4**	**CS-T3-DI-3**	**CS-T3-DI-4**
Test Duration (days)												
0.08	4.64	4.78	4.58	4.70	4.68	4.72	2.80	2.66	2.92	2.94	2.91	2.76
1	4.85	5.05	4.88	5.01	4.70	4.88	2.97	3.38	3.35	3.02	3.59	2.47
2	4.29	4.21	4.22	4.29	4.23	4.25	1.70	1.90	1.91	1.78	1.77	1.70
7	6.93	7.32	7.04	7.12	7.16	7.10	6.02	6.33	6.26	6.58	6.48	6.35
14	4.32	4.80	4.75	4.72	4.79	4.83	3.90	3.97	3.95	3.94	4.14	3.99
28	5.12	5.09	5.03	4.94	5.09	5.13	4.39	4.47	4.39	4.45	4.40	4.43
42	4.06	4.08	4.07	3.98	4.02	4.02	3.08	3.14	3.09	3.10	3.14	3.15
49	3.60	3.60	3.59	3.58	3.58	3.59	1.43	1.45	1.44	1.44	1.46	1.45
63	3.77	3.76	3.76	3.74	3.74	3.75	2.08	2.08	2.08	2.07	1.85	1.91
100	4.33	4.93	4.42	4.35	4.42	4.37	3.81	3.83	3.82	3.77	3.80	3.83
369	9.77	9.76	10.30	9.77	9.90	10.00	11.70	11.50	11.60	11.60	11.70	11.80
414	3.69	3.68	3.67	3.65	3.66	3.68	2.45	2.52	2.42	2.45	2.44	2.41
569	4.44	4.36	4.41	4.34	4.34	4.38	4.09	4.25	4.14	3.97	4.12	4.05

B.4 Tc-Gluconate Leach Tests

Table B.39. Sodium Observed Diffusion Coefficient (cm^2/s) in Tc-Gluconate Leach Tests

Sodium Observed Diffusion Coefficient (cm^2/s)										
Test Duration		**Tc Form**	**Tc-gluconate**				**Pertechnetate**			
Interval (days)	**Total (days)**	**Eluent**	**Vadose Zone Pore Water**		**Deionized Water**		**Vadose Zone Pore Water**		**Deionized Water**	
		Sample ID	**CS-T4-VZ-1**	**CS-T4-VZ-2**	**CS-T4-DI-3**	**CS-T4-DI-4**	**CS-T5-VZ-1**	**CS-T5-VZ-2**	**CS-T5-DI-3**	**CS-T5-DI-4**
0.08	0.08		3.58E-09	5.46E-09	1.85E-09	6.74E-09	3.51E-08	3.39E-08	3.50E-09	4.46E-09
.092	1		1.87E-08	1.78E-08	1.63E-08	1.64E-08	1.97E-08	1.69E-08	1.62E-08	1.59E-08
1	2		1.24E-08	1.16E-08	1.16E-08	1.20E-08	9.13E-09	8.96E-09	1.51E-08	1.51E-08
5	7		8.59E-09	1.02E-08	9.64E-09	8.74E-09	8.17E-09	8.83E-09	1.00E-08	8.64E-09
7	14		5.06E-09	6.13E-09	6.65E-09	6.40E-09	5.18E-09	5.19E-09	4.96E-09	5.67E-09
14	28		3.58E-09	3.90E-09	4.36E-09	4.09E-09	2.90E-09	3.16E-09	3.15E-09	3.80E-09
14	42		3.22E-09	3.43E-09	3.76E-09	3.49E-09	2.21E-09	2.29E-09	3.07E-09	3.09E-09
7	49		2.57E-09	2.71E-09	3.66E-09	3.88E-09	1.94E-09	1.95E-09	3.12E-09	3.08E-09
14	63		2.74E-09	2.51E-09	3.17E-09	3.35E-09	1.84E-09	1.72E-09	2.50E-09	2.66E-09
31	94		2.58E-09	2.58E-09	3.36E-09	3.03E-09	1.79E-09	1.52E-09	2.22E-09	2.66E-09
267	361		1.39E-09	1.24E-09	1.77E-09	1.62E-09	9.61E-10	9.23E-10	1.20E-09	1.23E-09
47	408		1.50E-09	2.11E-09	3.99E-09	3.77E-09	1.06E-09	1.02E-09	3.53E-09	3.92E-09
161	569		7.79E-10	7.79E-10	1.57E-09	1.20E-09	5.21E-10	5.50E-10	1.47E-09	1.26E-09
272	841		5.58E-10		1.17E-09		4.85E-10		1.07E-09	

Table B.40. Nitrate Observed Diffusion Coefficient (cm^2/s) in Tc-Gluconate Leach Tests

Nitrate Observed Diffusion Coefficient (cm^2/s)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Eluent	Vadose Zone Pore Water		Deionized Water		Vadose Zone Pore Water		Deionized Water	
		Sample ID	CS-T4-VZ-1	CS-T4-VZ-2	CS-T4-DI-3	CS-T4-DI-4	CS-T5-VZ-1	CS-T5-VZ-2	CS-T5-DI-3	CS-T5-DI-4
0.08	0.08		3.96E-09	5.67E-09	9.63E-10	3.26E-09	3.41E-08	3.36E-08	1.79E-09	2.39E-09
.092	1		1.54E-08	1.57E-08	1.44E-08	1.42E-08	1.55E-08	1.44E-08	1.30E-08	1.26E-08
1	2		9.07E-09	9.65E-09	9.87E-09	1.14E-08	5.81E-09	7.82E-09	1.14E-08	1.21E-08
5	7		8.12E-09	8.50E-09	8.78E-09	8.72E-09	6.98E-09	6.58E-09	6.97E-09	7.33E-09
7	14		3.91E-09	5.08E-09	5.60E-09	5.75E-09	3.72E-09	3.79E-09	3.96E-09	5.20E-09
14	28		2.79E-09	2.86E-09	3.80E-09	3.77E-09	2.17E-09	2.39E-09	3.07E-09	3.84E-09
14	42		2.58E-09	2.68E-09	3.67E-09	3.73E-09	1.82E-09	2.02E-09	2.94E-09	3.38E-09
7	49		2.25E-09	2.65E-09	3.30E-09	3.42E-09	1.78E-09	1.65E-09	2.66E-09	3.15E-09
14	63		2.34E-09	2.31E-09	3.26E-09	3.62E-09	1.55E-09	1.46E-09	2.50E-09	3.02E-09
31	94		2.57E-09	2.61E-09	3.83E-09	3.51E-09	1.49E-09	1.30E-09	2.40E-09	3.13E-09
267	361		2.47E-09	2.30E-09	3.42E-09	3.05E-09	1.75E-09	1.74E-09	3.25E-09	3.34E-09
47	408		1.52E-09	1.53E-09	3.21E-09	2.43E-09	1.29E-09	1.32E-09	3.60E-09	3.23E-09
161	569		7.63E-10	7.65E-10	2.51E-09	1.89E-09	6.18E-10	6.56E-10	2.75E-09	1.95E-09
272	841		6.64E-10		1.79E-09		6.98E-10		1.85E-09	

Table B.41. Nitrite Technetium Observed Diffusion Coefficient (cm^2/s) in Tc-Gluconate Leach Tests

Nitrite Observed Diffusion Coefficient (cm^2/s)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Eluent	Vadose Zone Pore Water		Deionized Water		Vadose Zone Pore Water		Deionized Water	
		Sample ID	CS-T4-VZ-1	CS-T4-VZ-2	CS-T4-DI-3	CS-T4-DI-4	CS-T5-VZ-1	CS-T5-VZ-2	CS-T5-DI-3	CS-T5-DI-4
0.08	0.08		3.32E-09	4.21E-09	9.11E-10	2.81E-09	2.05E-08	2.00E-08	1.65E-09	2.20E-09
.092	1		1.04E-08	9.64E-09	8.90E-09	8.77E-09	9.98E-09	8.28E-09	8.66E-09	8.34E-09
1	2		4.74E-09	5.45E-09	5.85E-09	6.46E-09	3.68E-09	4.96E-09	6.99E-09	7.49E-09
5	7		4.32E-09	4.56E-09	4.46E-09	4.31E-09	3.95E-09	3.78E-09	3.86E-09	4.09E-09
7	14		2.10E-09	2.50E-09	3.04E-09	3.13E-09	2.09E-09	2.02E-09	2.38E-09	2.95E-09
14	28		1.70E-09	1.73E-09	2.38E-09	2.27E-09	1.40E-09	1.47E-09	2.01E-09	2.43E-09
14	42		1.53E-09	1.56E-09	2.27E-09	2.26E-09	1.15E-09	1.26E-09	1.92E-09	2.16E-09
7	49		1.61E-09	1.82E-09	2.17E-09	2.19E-09	1.32E-09	1.21E-09	1.73E-09	2.02E-09
14	63		1.39E-09	1.44E-09	2.10E-09	2.38E-09	1.11E-09	1.01E-09	1.73E-09	1.99E-09
31	94		1.46E-09	1.46E-09	2.03E-09	1.90E-09	9.33E-10	8.47E-10	1.34E-09	1.69E-09
267	361		1.06E-09	9.71E-10	1.46E-09	1.29E-09	7.88E-10	7.96E-10	1.37E-09	1.41E-09
47	408		4.98E-10	4.70E-10	1.71E-09	1.28E-09	3.71E-10	4.15E-10	1.98E-09	1.75E-09
161	569		1.19E-09	1.21E-09	1.28E-09	9.30E-10	1.09E-09	1.12E-09	1.43E-09	1.00E-09
272	841		4.56E-10		9.41E-10		4.97E-10		1.05E-09	

Table B.42. Technetium Observed Diffusion Coefficient (cm^2/s) Tc-Gluconate Leach Tests

Technetium Observed Diffusion Coefficient (cm^2/s)										
Test Duration		**Tc Form**	**Tc-gluconate**				**Pertechnetate**			
Interval (days)	**Total (days)**	**Eluent**	**Vadose Zone Pore Water**		**Deionized Water**		**Vadose Zone Pore Water**		**Deionized Water**	
		Sample ID	**CS-T4-VZ-1**	**CS-T4-VZ-2**	**CS-T4-DI-3**	**CS-T4-DI-4**	**CS-T5-VZ-1**	**CS-T5-VZ-2**	**CS-T5-DI-3**	**CS-T5-DI-4**
0.08	0.08		7.17E-11	1.68E-10	2.40E-11	5.52E-11	4.76E-11	4.73E-13	1.34E-13	1.19E-13
.092	1		1.84E-10	2.13E-10	2.39E-10	1.96E-10	1.18E-11	1.31E-12	1.08E-11	3.44E-12
1	2		3.46E-11	3.59E-11	1.13E-10	1.17E-10	2.80E-12	9.76E-13	2.99E-11	9.53E-12
5	7		1.85E-11	1.95E-11	1.44E-10	1.45E-10	2.05E-12	1.67E-12	3.14E-11	1.20E-11
7	14		9.58E-12	1.34E-11	1.72E-10	1.97E-10	1.13E-12	1.03E-12	3.08E-11	1.76E-11
14	28		1.40E-11	1.87E-11	2.36E-10	2.81E-10	1.25E-12	1.18E-12	3.22E-11	2.23E-11
14	42		2.45E-11	3.24E-11	2.83E-10	2.91E-10	1.44E-12	1.35E-12	3.40E-11	2.55E-11
7	49		3.24E-11	4.54E-11	2.60E-10	2.35E-10	1.46E-12	1.63E-12	3.09E-11	2.45E-11
14	63		2.62E-11	2.81E-11	2.43E-10	2.16E-10	9.64E-13	1.10E-12	2.39E-11	1.93E-11
31	94		3.04E-11	3.58E-11	2.40E-10	1.85E-10	9.84E-13	8.72E-13	1.65E-11	1.54E-11
267	361		2.52E-11	2.63E-11	1.50E-10	1.15E-10	1.88E-12	1.99E-12	1.18E-11	1.20E-11
47	408		1.70E-11	1.36E-11	1.36E-10	9.55E-11	9.19E-13	1.13E-12	1.89E-11	3.10E-11
161	569		1.02E-11	7.81E-12	6.09E-11	5.24E-11	5.91E-13	9.06E-13	1.48E-11	2.04E-11
272	841		7.30E-12		5.72E-11		6.63E-13		1.22E-11	

Table B.43. Chromium Observed Diffusion Coefficient (cm^2/s) in Tc-Gluconate Leach Tests

Chromium Observed Diffusion Coefficient (cm^2/s)										
Test Duration		**Tc Form**	**Tc-gluconate**				**Pertechnetate**			
Interval (days)	**Total (days)**	**Eluent**	**Vadose Zone Pore Water**		**Deionized Water**		**Vadose Zone Pore Water**		**Deionized Water**	
		Sample ID	**CS-T4-VZ-1**	**CS-T4-VZ-2**	**CS-T4-DI-3**	**CS-T4-DI-4**	**CS-T5-VZ-1**	**CS-T5-VZ-2**	**CS-T5-DI-3**	**CS-T5-DI-4**
0.08	0.08		6.44E-15	7.21E-14	1.34E-14	4.93E-14	7.28E-12	<2.63E-14	1.63E-14	2.42E-14
.092	1		1.41E-13	3.01E-13	7.62E-13	7.80E-13	1.07E-12	4.55E-15	2.53E-13	2.38E-13
1	2		6.10E-15	8.42E-15	2.09E-13	2.59E-13	7.85E-14	1.28E-17	1.80E-13	1.59E-13
5	7		1.96E-15	1.81E-15	2.02E-13	2.24E-13	7.68E-15	3.01E-14	1.64E-13	1.48E-13
7	14		7.08E-17	3.11E-16	1.93E-13	2.09E-13	3.33E-16	4.46E-17	1.48E-13	1.48E-13
14	28		1.23E-17	5.24E-17	1.18E-13	1.27E-13	8.12E-17	2.69E-17	7.48E-14	8.10E-14
14	42		<1.46E-15	<1.48E-15	1.17E-13	1.04E-13	<1.51E-15	<1.55E-15	8.19E-14	8.49E-14
7	49		<7.63E-15	<7.77E-15	9.70E-14	8.14E-14	<7.91E-15	<8.11E-15	6.67E-14	7.93E-14
14	63		<2.34E-15	<2.38E-15	6.36E-14	5.76E-14	<2.42E-15	<2.49E-15	3.71E-14	3.30E-14
31	94		<6.66E-16	<6.79E-16	5.25E-14	4.43E-14	<6.91E-16	<7.08E-16	2.34E-14	8.63E-14
267	361		4.06E-17	2.79E-17	1.93E-14	1.55E-14	1.12E-16	5.19E-17	8.35E-15	1.38E-14
47	408		1.91E-18	3.97E-20	1.84E-13	1.18E-13	4.95E-17	<3.80E-16	7.27E-14	1.62E-13
161	569		<1.54E-16	<1.57E-16	1.42E-14	1.29E-14	<1.60E-16	<1.64E-16	9.29E-15	1.04E-14
272	841		3.90E-18		9.20E-15		1.48E-18		7.35E-15	

Table B.44. Sodium Fraction Leached in Tc-Gluconate Leach Tests

Sodium Fraction Leached from Tc-Gluconate Monoliths (unitless)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Eluent	Vadose Zone Pore Water		Deionized Water		Vadose Zone Pore Water		Deionized Water	
		Sample ID	CS-T4-VZ-1	CS-T4-VZ-2	CS-T4-DI-3	CS-T4-DI-4	CS-T5-VZ-1	CS-T5-VZ-2	CS-T5-DI-3	CS-T5-DI-4
0.08	0.08		0.006	0.007	0.004	0.009	0.018	0.018	0.006	0.007
.092	1		0.039	0.039	0.035	0.042	0.052	0.049	0.036	0.039
1	2		0.054	0.054	0.050	0.058	0.065	0.062	0.053	0.056
5	7		0.092	0.095	0.090	0.100	0.103	0.100	0.095	0.097
7	14		0.118	0.124	0.120	0.132	0.129	0.127	0.121	0.125
14	28		0.149	0.156	0.154	0.167	0.157	0.156	0.150	0.159
14	42		0.172	0.180	0.179	0.193	0.176	0.175	0.172	0.182
7	49		0.181	0.189	0.189	0.205	0.184	0.182	0.182	0.192
14	63		0.197	0.204	0.207	0.224	0.197	0.195	0.198	0.209
31	94		0.227	0.234	0.241	0.259	0.222	0.218	0.225	0.241
267	361		0.344	0.344	0.372	0.395	0.319	0.312	0.334	0.356
47	408		0.359	0.362	0.398	0.422	0.332	0.325	0.358	0.382
161	569		0.393	0.396	0.446	0.467	0.360	0.354	0.405	0.428
272	841		0.434		0.505		0.399		0.461	

Table B.45. Nitrate Fraction Leached in Tc-Gluconate Leach Tests

Nitrate Fraction Leached from Tc-Gluconate Monoliths (unitless)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Eluent	Vadose Zone Pore Water		Deionized Water		Vadose Zone Pore Water		Deionized Water	
		Sample ID	CS-T4-VZ-1	CS-T4-VZ-2	CS-T4-DI-3	CS-T4-DI-4	CS-T5-VZ-1	CS-T5-VZ-2	CS-T5-DI-3	CS-T5-DI-4
0.08	0.08		0.006	0.007	0.003	0.006	0.018	0.018	0.004	0.005
.092	1		0.036	0.037	0.032	0.037	0.048	0.046	0.032	0.033
1	2		0.049	0.051	0.045	0.053	0.058	0.058	0.046	0.049
5	7		0.086	0.089	0.084	0.094	0.093	0.092	0.081	0.086
7	14		0.109	0.115	0.112	0.125	0.116	0.114	0.104	0.114
14	28		0.137	0.142	0.144	0.159	0.140	0.139	0.133	0.148
14	42		0.157	0.163	0.168	0.185	0.157	0.157	0.155	0.172
7	49		0.165	0.172	0.178	0.196	0.164	0.164	0.164	0.182
14	63		0.180	0.187	0.196	0.217	0.177	0.176	0.179	0.200
31	94		0.210	0.217	0.232	0.255	0.199	0.197	0.208	0.235
267	361		0.365	0.366	0.414	0.441	0.330	0.327	0.387	0.424
47	408		0.381	0.382	0.437	0.462	0.345	0.341	0.411	0.447
161	569		0.415	0.416	0.498	0.520	0.375	0.373	0.475	0.504
272	841		0.459		0.571		0.421		0.549	

Table B.46. Nitrite Fraction Leached in Tc-Gluconate Leach Tests

Nitrite Fraction Leached from Tc-Gluconate Monoliths (unitless)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Eluent	Vadose Zone Pore Water		Deionized Water		Vadose Zone Pore Water		Deionized Water	
		Sample ID	CS-T4-VZ-1	CS-T4-VZ-2	CS-T4-DI-3	CS-T4-DI-4	CS-T5-VZ-1	CS-T5-VZ-2	CS-T5-DI-3	CS-T5-DI-4
0.08	0.08		0.006	0.006	0.003	0.006	0.014	0.014	0.004	0.005
.092	1		0.030	0.030	0.026	0.030	0.038	0.035	0.026	0.028
1	2		0.039	0.040	0.036	0.042	0.046	0.045	0.038	0.040
5	7		0.067	0.068	0.064	0.071	0.072	0.070	0.064	0.068
7	14		0.083	0.086	0.084	0.093	0.089	0.087	0.082	0.089
14	28		0.105	0.108	0.109	0.120	0.109	0.106	0.105	0.116
14	42		0.121	0.123	0.128	0.141	0.122	0.120	0.122	0.135
7	49		0.127	0.131	0.136	0.149	0.129	0.126	0.130	0.143
14	63		0.139	0.143	0.151	0.166	0.139	0.136	0.143	0.158
31	94		0.162	0.165	0.177	0.194	0.157	0.153	0.164	0.183
267	361		0.263	0.262	0.296	0.315	0.245	0.241	0.280	0.306
47	408		0.272	0.271	0.313	0.330	0.253	0.249	0.298	0.324
161	569		0.315	0.313	0.357	0.371	0.293	0.290	0.344	0.364
272	841		0.351		0.409		0.332		0.400	

Table B.47. Technetium Fraction Leached in Tc-Gluconate Leach Tests

Technetium Fraction Leached from Tc-Gluconate Monoliths (unitless)										
Test Duration		**Tc Form**	**Tc-gluconate**				**Pertechnetate**			
Interval (days)	**Total (days)**	**Eluent→**	**Vadose Zone Pore Water**		**Deionized Water**		**Vadose Zone Pore Water**		**Deionized Water**	
		ID→	**CS-T4-VZ-1**	**CS-T4-VZ-2**	**CS-T4-DI-3**	**CS-T4-DI-4**	**CS-T5-VZ-1**	**CS-T5-VZ-2**	**CS-T5-DI-3**	**CS-T5-DI-4**
0.08	0.08		0.0008	0.0012	0.0005	0.0008	0.0007	0.0001	0.0000	0.0000
.092	1		0.0041	0.0047	0.0042	0.0044	0.0015	0.0003	0.0008	0.0005
1	2		0.0049	0.0056	0.0056	0.0060	0.0017	0.0005	0.0016	0.0009
5	7		0.0067	0.0074	0.0106	0.0114	0.0023	0.0010	0.0039	0.0024
7	14		0.0078	0.0087	0.0154	0.0170	0.0027	0.0014	0.0059	0.0041
14	28		0.0097	0.0110	0.0234	0.0264	0.0033	0.0019	0.0089	0.0066
14	42		0.0117	0.0132	0.0301	0.0337	0.0038	0.0024	0.0112	0.0087
7	49		0.0127	0.0144	0.0329	0.0366	0.0040	0.0026	0.0122	0.0096
14	63		0.0143	0.0161	0.0378	0.0416	0.0043	0.0029	0.0137	0.0111
31	94		0.0176	0.0196	0.0469	0.0502	0.0049	0.0035	0.0161	0.0135
267	361		0.0332	0.0355	0.0850	0.0864	0.0092	0.0079	0.0269	0.0248
47	408		0.0349	0.0370	0.0897	0.0907	0.0096	0.0083	0.0286	0.0272
161	569		0.0388	0.0404	0.0993	0.1002	0.0105	0.0095	0.0334	0.0330
272	841		0.0434		0.1123		0.0119		0.0394	

Table B.48. Chromium Fraction Leached in Tc-Gluconate Leach Tests

Chromium Fraction Leached from Tc-Gluconate Monoliths (unitless)										
Test Duration		**Tc Form**	**Tc-gluconate**				**Pertechnetate**			
Interval (days)	**Total (days)**	**Eluent→**	**Vadose Zone Pore Water**		**Deionized Water**		**Vadose Zone Pore Water**		**Deionized Water**	
		ID→	**CS-T4-VZ-1**	**CS-T4-VZ-2**	**CS-T4-DI-3**	**CS-T4-DI-4**	**CS-T5-VZ-1**	**CS-T5-VZ-2**	**CS-T5-DI-3**	**CS-T5-DI-4**
0.08	0.08		7.76E-06	2.59E-05	1.12E-05	2.32E-05	2.62E-04	1.56E-05	1.23E-05	1.58E-05
.092	1		9.79E-05	1.57E-04	2.21E-04	2.53E-04	5.12E-04	3.18E-05	1.34E-04	1.39E-04
1	2		1.09E-04	1.70E-04	2.84E-04	3.28E-04	5.50E-04	3.22E-05	1.92E-04	1.96E-04
5	7		1.27E-04	1.87E-04	4.69E-04	5.40E-04	5.86E-04	1.04E-04	3.60E-04	3.63E-04
7	14		1.30E-04	1.94E-04	6.30E-04	7.21E-04	5.93E-04	1.06E-04	5.02E-04	5.11E-04
14	28		1.32E-04	1.98E-04	8.09E-04	9.21E-04	5.98E-04	1.09E-04	6.44E-04	6.66E-04
14	42		1.47E-04	2.13E-04	9.45E-04	1.06E-03	6.13E-04	1.24E-04	7.58E-04	7.88E-04
7	49		1.62E-04	2.28E-04	1.00E-03	1.11E-03	6.29E-04	1.40E-04	8.03E-04	8.39E-04
14	63		1.77E-04	2.43E-04	1.08E-03	1.20E-03	6.44E-04	1.55E-04	8.64E-04	8.99E-04
31	94		1.93E-04	2.59E-04	1.21E-03	1.33E-03	6.60E-04	1.71E-04	9.54E-04	1.08E-03
267	361		2.13E-04	2.75E-04	1.65E-03	1.75E-03	6.93E-04	1.93E-04	1.24E-03	1.46E-03
47	408		2.13E-04	2.75E-04	1.82E-03	1.90E-03	6.96E-04	2.01E-04	1.35E-03	1.63E-03
161	569		2.28E-04	2.91E-04	1.97E-03	2.05E-03	7.12E-04	2.17E-04	1.47E-03	1.76E-03
272	841		2.32E-04		2.13E-03		7.14E-04		1.62E-03	

Table B.49. pH in Eluates from Tc-Gluconate Leach Tests

pH in Eluates (unitless)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Eluent→	Vadose Zone Pore Water		Deionized Water		Vadose Zone Pore Water		Deionized Water	
		ID→	CS-T4-VZ-1	CS-T4-VZ-2	CS-T4-DI-3	CS-T4-DI-4	CS-T5-VZ-1	CS-T5-VZ-2	CS-T5-DI-3	CS-T5-DI-4
0.08	0.08		10.4	10.4	11.4	11.4	10.6	10.7	11.4	11.4
.092	1		11.2	11.3	12.0	12.1	11.2	11.3	12.1	12.1
1	2		10.6	10.5	11.8	11.8	10.6	10.6	11.8	11.7
5	7		11.6	11.6	12.1	12.1	11.6	11.6	12.1	12.1
7	14		11.4	11.5	12.1	12.1	11.3	11.3	12.1	12.1
14	28		11.5	11.5	12.1	12.1	11.4	11.4	12.1	12.1
14	42		10.8	10.8	12.0	12.0	10.7	10.7	12.0	11.9
7	49		10.4	10.4	11.7	11.6	10.4	10.4	11.6	11.6
14	63		10.5	10.5	11.8	11.8	10.5	10.4	11.8	11.8
31	94		10.9	10.9	12.0	12.0	10.6	10.6	12.0	12.0
267	361		12.3	12.3	12.5	12.5	12.1	12.0	12.5	12.4
47	408		10.4	10.4	11.8	11.8	10.3	10.3	11.8	11.7
161	569		10.7	10.8	12.0	11.9	10.4	10.4	12.0	11.9
272	841		10.3		11.9		10.3		11.9	

Table B.50. EC in Eluates from Tc-Gluconate Leach Tests

Electrical Conductivity in Eluates (mS/cm)										
Test Duration		**Tc Form**	**Tc-gluconate**				**Pertechnetate**			
Interval (days)	**Total (days)**	**Eluent→**	**Vadose Zone Pore Water**		**Deionized Water**		**Vadose Zone Pore Water**		**Deionized Water**	
		ID →	**CS-T4-VZ-1**	**CS-T4-VZ-2**	**CS-T4-DI-3**	**CS-T4-DI-4**	**CS-T5-VZ-1**	**CS-T5-VZ-2**	**CS-T5-DI-3**	**CS-T5-DI-4**
0.08	0.08		4.05	4.10	0.65	0.94	4.54	4.61	0.67	0.74
.092	1		5.51	5.57	4.28	4.22	5.48	5.48	4.22	4.09
1	2		4.50	4.49	2.37	1.32	4.48	4.45	2.32	2.06
5	7		6.52	6.65	5.65	3.05	6.35	6.30	5.38	5.20
7	14		4.55	4.60	3.67	3.55	4.33	4.28	3.50	3.42
14	28		4.89	4.92	4.33	4.13	4.60	4.61	4.04	3.93
14	42		4.14	4.15	3.14	3.01	3.98	4.00	2.87	2.80
7	49		3.66	3.67	1.47	1.41	3.62	3.60	1.32	1.30
14	63		3.79	3.78	2.13	2.04	3.67	3.66	1.86	1.82
31	94		4.35	4.35	3.60	3.48	4.01	3.95	3.01	3.12
267	361		11.20	11.20	12.80	12.00	9.07	8.90	11.40	10.40
47	408		3.75	3.75	2.74	2.53	3.66	3.66	2.53	2.34
161	569		4.63	4.67	4.60	4.05	4.44	4.46	4.41	3.89
272	841		4.77		5.19		4.74		4.98	

Appendix C

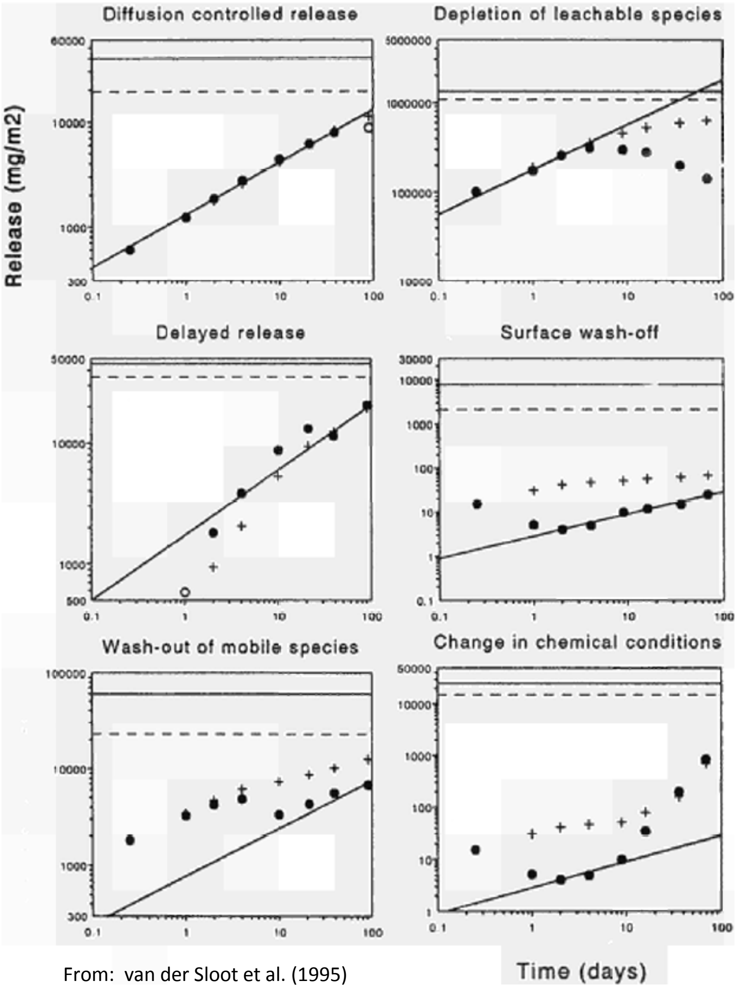
Determination of Leaching Mechanism(s)

Appendix C

Determination of Leaching Mechanism(s)

There are several types of constituent release, and ways of portraying them, that are mentioned in the literature in relation to semi-dynamic tank leaching tests. Figure C.1 is a summary of different plots of Release versus Time which include examples of:

- Diffusion Controlled Release
- Delayed Release
- Wash-Out of Mobile Species
- Depletion of Leachable Species
- Surface Wash-Off
- Change in Chemical Conditions



From: van der Sloot et al. (1995)

Where: Y axis in each plot is log of release of COC (mg/m^2) and X-axis is log of time (days).
horizontal solid line = total concentration
horizontal dotted line = availability
solid dot reflects calculated release based on leached quantity per interval of time
plus sign reflects the cumulative release.

Figure C.1. Different Types of Release as Observed in Tank Leaching Tests on Stabilized Materials

The EPA Method 1315 criteria for determining if a diffusivity is "allowed" to be calculated within a leach test interval using Eq. C.4 (below), is based on Fickian diffusion being the controlling release mechanism. Fickian diffusion requires that the slope for the line within the interval, which is plotted as cumulative log of release versus cumulative log of time be equal to 0.5.

Eq. C.1 is the analytical solution by Crank (1986) for a one-dimensional geometry with an assumption of zero concentration at the solid-liquid bath interface (i.e., the case of a sufficient water renewal and infinite bath assumption) (Kosson et al., 2002).

$$M_t = 2\rho C_0 \left(\frac{D^{obs}\, t}{\pi}\right)^{1/2} \qquad \text{Eq. C.1}$$

where: M_t = cumulative mass released during the leaching interval (mg/m^2)
ρ = density of the "as-tested" sample (kg/m^3)
C_0 = available concentration of constituent of interest in the solid matrix (mg/kg)
D^{obs} = observed diffusivity (m^2/s)
t = leaching time of the test interval (s)

Note that the "available" concentration may be less than the total concentration if some of the mass of the constituent of interest is "inert" to diffusion. The portion of the constituent of interest that is "inert" is sometimes estimated from EPA- Method 1313 (EPA 2012). In order to use Eq. C.1, depletion of the constituent of interest must not occur. Depletion is considered to occur when 20% of the total leachable content has been released (Kosson et al., 2002). Using those values of cumulative release that are less than 20% of the total release ensures the conditions for using Eq. C.2 are met.

A log transform of Eq. C.1, results in a linear equation with a slope of ½ or 0.5.

$$\log M_t = \log\left[\, 2\rho C_0 \left(\frac{D^{obs}}{\pi}\right)^{1/2} \right] + \frac{1}{2}\log t \qquad \text{Eq. C.2}$$

Or, rearranging as Y = mX + b

$$\log M_t = \frac{1}{2}\log t + \log\left[\, 2\rho C_0 \left(\frac{D^{obs}}{\pi}\right)^{1/2} \right] \qquad \text{Eq. C.3}$$

For a diffusion-controlled release mechanism, plotting the logarithm of the cumulative release versus the logarithm of cumulative time will theoretically generate points such that a straight line from each interval's starting point to its ending point will have a slope of 0.5. Per EPA Method 1315, only data from those intervals having slopes of 0.5 ±0.15 are to be used to calculate an observed diffusivity for the interval using Eq. C.4, and those values of D_{obs} are then to be used to determine the overall mean[1] observed diffusivity. If the interval slope is not 0.5 ±0.15 for any given interval, then the interval D_{obs} value calculated from Eq. C.4 cannot be used to determine a mean observed diffusivity.

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[1] In the main text the mean observed D_{obs} value is called the average D_{obs} value. Mean and average are considered equivalent in this report.

$$D_i^{obs} = \pi \left[\frac{M_{t_i}}{2\rho C_0\left(\sqrt{t_i} - \sqrt{t_{i-1}}\right)} \right]^2 \qquad \text{Eq. C.4}$$

where: t_i = cumulative contact time at the end of the current leaching interval, i (s)
t_{i-1} = cumulative contact time at the end of the previous leaching interval, i-1 (s)

It is noted in van der Sloot et al. (1989), de Groot and van der Sloot (1992), and Kosson et al. (2002) that some observed initial releases from laboratory tank leaching tests indicate wash off or dissolution of surface-associated constituents. This apparent constituent release may then be followed by diffusion-controlled releases within subsequent leaching intervals. Per EPA Method 1315, only those data from diffusion-controlled intervals (slope of logarithm of the cumulative release plotted versus the logarithm of time is 0.5 ± 0.15) are to be used to estimate/calculate the observed diffusivity (diffusion coefficient).

As previously discussed in Section 3.3 of this report, under assumed simple diffusion controlled release, both the ANS-16.1 D_e equation and the EPA Method 1315 D_{obs} equation are based on the same diffusion model which assumes (i) a homogeneous solid matrix, (ii) constant release parameters, (iii) constant driving force for diffusion (i.e., zero concentration at the solid's surface boundary), (iv) no depletion of the constituent at the inner core of the solid matrix, and (v) less than 20% of the initial content is leached (which allows the use of the semi-infinite media assumption given in the equations). In addition:

- The ANS-16.1 semi-dynamic leach test method and the EPA Method 1315 semi-dynamic leach test method are both "tests within a test" methods, in that both methods employ a sequence of discrete tests defined by their beginning (initial conditions) times and ending (sampling conditions) times.
- Experimental data are analyzed for each interval test.
- Using an equation to calculate interval diffusivity from either of the methods is predicated on there being some sort of analysis of the experimental data to determine if the equation is valid for use with some or all of the experimental data.
- Equations to calculate an interval diffusivity are used, or not used, based on whether or not diffusion can be verified (not just assumed) as the controlling leach mechanism.
  - For ANS-16.1 method, "calculation of the D_e value requires that diffusion be the controlling leaching mechanism [a straight line must be obtained when plotting ($\Sigma a_n/A_o$) versus $t_n^{1/2}$]" (Cotè and Isabel, 1984).
  - For EPA Method 1315, "An observed diffusivity for each COPC can be determined using the logarithm of the cumulative release plotted versus the logarithm of time. In the case of a diffusion controlled mechanism, this plot is expected to be a straight line with a slope of 0.5. An observed diffusivity can then be determined for each leaching interval where this calculated slope is 0.50 ± 0.15" (EPA, 2013).
- Less than 20% of the COPC has been leached.
  - For ANS-16.1 method, "If more than 20% of a leachable species has been removed by the time, t, the effective diffusivity can only be calculated from a shape-specific solution of the mass transport equations" (ANS, 2003).
- The leachability index value can be interpreted as the negative logarithm of the effective diffusivity only provided long-term leaching data support the hypothesis that diffusivity controls leachability (Cotè and Isabel, 1984)

Previous PNNL and SRNL reports[1] cite EPA Method 1315 (which itself has definitions of D_{obs} and D_{eff}), but use "D_{eff}" or "D_{app}" as the parameter calculated by EPA Method 1315's D_{obs} equation. So, their values of "D_e", "D_{eff}", or "D_{app}" are calculated by the same equation as 1315's D_{obs} equation used in this report. Hence, value of D_{eff} = value of D_{app} = value of D_{obs}.

Also, the cited PNNL and SRNL reports make no mention of whether all the "criteria" checks mentioned above were performed to see whether the interval leaching data satisfies the criteria as thus can be stated to be diffusion controlled. Thus use of the diffusivity values presented in the cited PNNL and SRNL reports requires the assumption that the leaching described in the reports was in fact diffusion controlled. So, unknown to any reader, some of the D_{obs}, D_{app}, or D_{eff} values reported in these cited reports could have been calculated from experimental data collected during times when wash-off, dissolution, or other non-Fickian diffusion-controlled processes dominated the leaching.

The plots that follow in Figure C.2 through Figure C.7 are selected examples, with analysis, from the Tc-Gluconate suite of semi-dynamic leach testing that illustrate analyses that could be used to tell whether leaching in a particular test interval is diffusion controlled. In addition, Table C.1 and Table C.2 show the interval checks for our Extended and Archived Suites of monoliths (see Table 3.1 and Table 4.3 in the main text for specifics).

For each plot of cumulative logarithm of release versus cumulative logarithm of leaching time:

- Starting from the end of the first leaching interval (0.08 day), the dashed black line has a slope of 0.5 and is what is theoretically expected if diffusion is the controlling release mechanism. Each plotted cumulative log release would be on the line if diffusion controlled the release.
- Starting from the end of the first leaching interval (0.08 day), the green dashed lines have slopes of 0.65 and 0.35, which are the upper and lower bounds for diffusion-controlled release per EPA Method 1315. Each cumulative log release plotted within those two bounds would be considered diffusion controlled.

One would expect that any cumulative log release data plotted within the bounding lines would be diffusion controlled. And, any cumulative log release data plotted outside the bounding lines would be controlled by a mechanism other than diffusion.

While not specifically mentioning 20% depletion, EPA Method 1315 simplifying assumptions require <20% depletion to calculate an observed diffusion coefficient. Any cumulative log release data plotted after 20% depletion of the constituent within the monolith would not yield an accurate observed diffusion coefficient.

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[1] Cantrell KJ, JH Westsik, Jr, RJ Serne, W Um and AD Cozzi. 2016. *Secondary Waste Cementitious Waste Form Data Package for the Integrated Disposal Facility Performance Assessment.* PNNL-25194, Pacific Northwest National Laboratory, Richland, WA.

Flach GP, DI Kaplan, RL Nichols, RR Seitz and RJ Serne. 2016. *Solid Secondary Waste Data Package Supporting Hanford Integrated Disposal Facility Performance Assessment.* SRNL-STI-2016-00175, Revision 0, Savannah River National Laboratory, Aiken, SC.

Fox KM, KA Roberts, and TB Edwards. 2014. *Cast Stone Formulations at Higher Sodium Concentrations.* SRNL-STI-2013-00499, Rev. 2, Savannah River Site, Aiken, South Carolina.

Mattigod SV, JH Westsik, Jr., CW Chung, MJ Lindberg, and KE Parker. 2011. *Waste Acceptance Testing of Secondary Waste Forms: Cast Stone, Ceramicrete and DuraLith*. PNNL-20632, Pacific Northwest National Laboratory, Richland, Washington.

Pierce EM, W Um, KJ Cantrell, MM Valenta, JH Westsik, Jr., RJ Serne, and KE Parker. 2010. *Secondary Waste Form Screening Test Results – Cast Stone and Alkali Alumino-Silicate Geopolymer*. PNNL-19505, Pacific Northwest National Laboratory, Richland, Washington.

Serne, RJ, JH Westsik, Jr, BD Williams, HB Jung, and G Wang. 2015. *Extended Leach Testing of Simulated LAW Cast Stone Monoliths.* PNNL-24297, Rev. 0,, Pacific Northwest National Laboratory, Richland, WA.

Sundaram SK, J Chun, W Um, KE Parker, CW Chung, JH Westsik Jr, MM Valenta, ML Kimura, SG Pitman, and CA Burns. 2011. *Secondary Waste Form Development and Optimization—Cast Stone*. PNNL-20159, Rev. 1, Pacific Northwest National Laboratory, Richland, Washington.

Um W, BD Williams, MMV Snyder, and G Wang. 2016. *Liquid Secondary Waste Grout Formulation and Waste Form Qualificatio*n." PNNL-25129, Pacific Northwest National Laboratory, Richland, WA.

Westsik JH, Jr., GF Piepel, MJ Lindberg, PG Heasler, TM Mercier, RL Russell, A Cozzi, WE Daniel, Jr., RE Eibling, EK Hansen, MM Reigel, and DJ Swanberg. 2013. *Supplemental Immobilization of Hanford Low-Activity Waste: Cast Stone Screening Tests*. PNNL-22747, SRNL-STI-2013-00465, Pacific Northwest National Laboratory, Richland, Washington.

Both EPA Method 1315 and ANS-16.1 are designed to be leaching interval-based tests. Several questions arise when attempting to determine which interval data is "allowable" for calculating observed diffusivities and then mean diffusivities and Leachability Indexes.

- How are the mean observed diffusivity and leachability index calculated when there are gaps in the interval mass released (log M_t) vs log t calculated interval slope values that meet the required 0.5 ± 0.15 slope criteria that indicates diffusion-controlled leaching? In Figure C.2 the Tc plot for monolith T4-VZ-1 and in Figure C.7 the plot for monolith T5-VZ-1 are good examples of data sets that have many intervals where the calculated interval slopes do not meet the 0.5 ± 0.15 criteria.
- How are the mean observed diffusivity and leachability index calculated when most of the interval mass released (log M_t) vs log t calculated interval slope values are within the required 0.5 ± 0.15 criteria indicating diffusion-controlled leaching, but the interval data points do not fit within the 0.35 and 0.65 slope boundary lines generated from the end of the 0.08-1 day interval mass released (log M_t) vs log t data point? In Figure C.6 the nitrate plot for monolith T4-DI-3 is an example of a data set where the interval slopes meet the 0.5 ± 0.15 criteria but the interval data points fall outside or in some cases at the upper bound 0.65 slope boundary line generated from the 0.08-1 day starting data point.
- How are the mean observed diffusivity and leachability index calculated when most (or all) of the mass released (log M_t) vs log t data points fall within the calculated 0.35 and 0.65 slope boundary lines generated from the end of the 0.08-1 day interval mass released (log M_t) vs log t data point, which indicates diffusion-controlled leaching, but the interval mass released (log M_t) vs log t interval data points do not have calculated linear slopes within the required 0.5 ± 0.15 criteria? In Figure C.2 the Tc plot for monolith T4-VZ-2 and in Figure C.5 the nitrate plots for both monoliths (T4-VZ-1 and T4-VZ-2) all are good examples where the interval slopes in general don't meet the 0.5 ± 0.15 criteria but the data fall within the 0.35 and 0.65 slope boundary lines generated from the 0.08-1 day starting data point.
- How are the mean diffusivity and leachability index calculated when most (or all) of the mass released (log M_t) vs log t interval data points meet the required 0.5 ± 0.15 interval slope criteria, but they lay outside the calculated 0.35 and 0.65 slope boundary lines generated from the end of the 0.08-1 day interval mass released (log M_t) vs log t data point? In Figure C.3 the Tc plot for monolith T4-DI-3 and in Figure C.4 the Tc plot for monolith T5-DI-3 the interval slopes in general meet the 0.5 ± 0.15 criteria and the data fall outside the 0.35 and 0.65 slope boundary lines generated from the 0.08-1 day starting data point.

Thus selecting only interval D_{obs} values from the Cast Stone monoliths EPA Method 1315 tests that meet the criteria so they can be considered for calculating mean D_{obs} values and leachability indices is problematical. To date we have not seen any of the other published reports containing diffusivity data on Cast Stone, hydrated lime based grouts or saltstone explicitly address this issue.

None of the past reports, cited in foot note 1 on page C.4, alert readers that no "criteria" checks were done on interval leach data before tabulating the data or recommending specific values to get average D_{obs} values to recommend to IDF PA modelers.

For consistency with the cited reports, in Section 4.0 of the main text of this Rev. 1 report, we have provided calculated average interval D_{obs} values without regard to performing "criteria" checks on the data.

This Appendix C is the first attempt at performing such "criteria" checks but we do not censor interval data that fails the "criteria" checks or modify our recommendations for what D_{obs} values to use in the next round of IDF calculations. We leave the decision up to the reader on whether to impose the EPA Method 1315 "criteria" checking procedures on available experimental leach data to remove interval D_{obs} values that do not meet the criteria and thus modify recommendations made in the cited reports. In the future, this EPA Method 1315 “criteria” checking issue should be discussed between the leaching experimentalists, the PA modeling, and the regulatory communities in the hope of developing a consensus path forward.

Tc-Gluconate Suite D_{obs} Equation Criteria Checks and Analyses

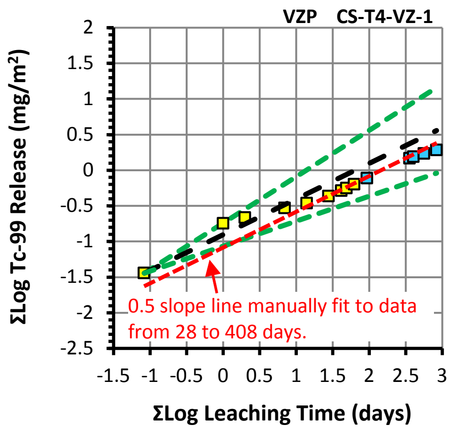
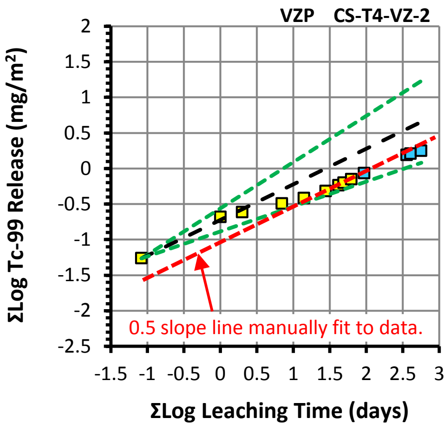
- Black dashed (0.5 slope) line is expected line when COPC depletion does not occur and Fickian diffusion is the dominant release mechanism from 0.08 day interval forward. - Green dashed lines have 0.65 and 0.35 slopes from 0.08 day interval forward. - Yellow markers are from 0.08 to 63 days. - Blue markers are from beyond 63 days.	Calculated Interval Slope (rise/run)	X = Diffusion Controlled ――― Interval Slope = 0.5 ± 0.15	Leach Test Interval (days)	Analysis per EPA Method 1315 COC = Constituent of Concern VZP = Vadose Zone Pore Water
	0.643	X	0.08-1	COC: **Tc-99** Eluent: **VZP** - All points have <20% depletion. - All points are within the theoretical 0.5±0.15 slopes from 0.08 day interval forward. However, slopes for intervals from 1 day to 28 days are not 0.5±0.15. - Release data for intervals from 28 to 408 days could be used for estimating a mean D_{obs} value and Leachability Index.
	0.265		1-2	
	0.248		2-7	
	0.228		7-14	
	0.322		14-28	
	0.455	X	28-42	
	0.527	X	42-49	
	0.474	X	49-63	
	0.511	X	63-94	
	0.474	X	94-361	
	0.398	X	361-408	
	0.319		408-569	
	0.290		569-841	
	0.535	X	0.08-1	COC: **Tc-99** Eluent: **VZP** Duplicate of CS-T4-VZ-1. - All points have <20% depletion. - All points are within the theoretical 0.5±0.15 slopes from 0.08 day interval forward. However, slopes for intervals from 1 day to 28 days are not 0.5±0.15. - Release data of intervals from 28 to 361 days could be used for estimating a mean D_{obs} value and Leachability Index.
	0.233		1-2	
	0.226		2-7	
	0.241		7-14	
	0.330		14-28	
	0.462	X	28-42	
	0.550	X	42-49	
	0.435	X	49-63	
	0.495	X	63-94	
	0.443	X	94-361	
	0.333		361-408	
	0.265		408-569	

Figure C.2. Analysis of Cumulative Log of Tc-99 Release versus Cumulative Log of Leaching Time Plots

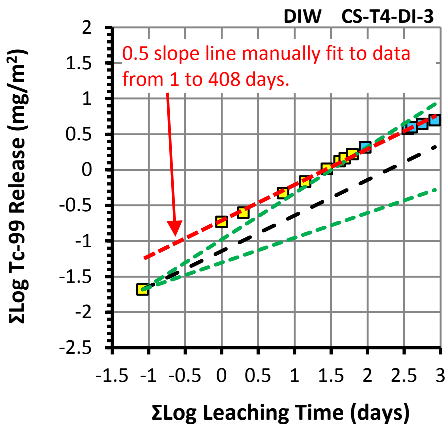
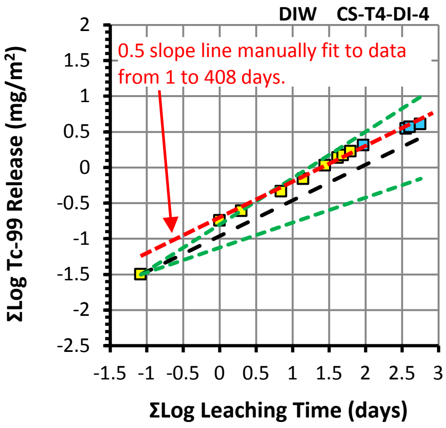
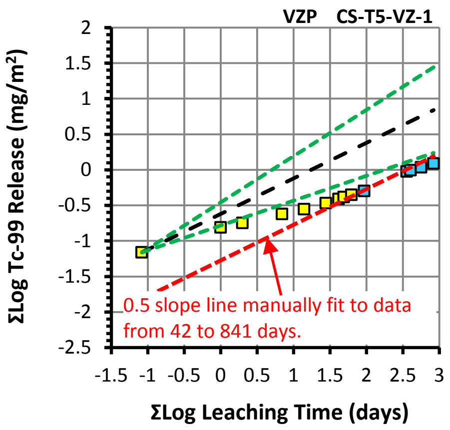
• Black dashed (0.5 slope) line is expected line when COPC depletion does not occur and Fickian diffusion is the dominant release mechanism from 0.08 day interval forward. • Green dashed lines have 0.65 and 0.35 slopes from 0.08 day interval forward. • Yellow markers are from 0.08 to 63 days. • Blue markers are from beyond 63 days.	Calculated Interval Slope (rise/run)	X = Diffusion Controlled ——— Interval Slope = 0.5 ± 0.15	Leach Test Interval (days)	Analysis per EPA Method 1315 COC = Constituent of Concern DIW = De-Ionized Water
	0.873		0.08-1	COC: **Tc-99** Eluent: **DIW** • All points have <20% depletion. • Most points are outside the theoretical 0.5±0.15 slopes from 0.08 day interval forward. However, slopes for intervals from 1 day to 408 days are each within 0.5±0.15. • There may be wash off within the 0.08 to 1 day interval. • Release data for intervals from 1 to 408 days could be used for estimating a mean D_{obs} value and Leachability Index.
	0.437	X	1-2	
	0.503	X	2-7	
	0.541	X	7-14	
	0.602	X	14-28	
	0.621	X	28-42	
	0.579	X	42-49	
	0.552	X	49-63	
	0.540	X	63-94	
	0.442	X	94-361	
	0.438	X	361-408	
	0.304		408-569	
	0.316		569-841	
	0.696		0.08-1	COC: **Tc-99** Eluent: **DIW** Duplicate of CS-T4-DI-3 • All points have <20% depletion. • Most points are within the theoretical 0.5±0.15 slopes from 0.08 day interval forward. Further, slopes for intervals from 1 day to 408 days are each within 0.5±0.15. • There may be wash off within the 0.08 to 1 day interval. • Release data for intervals from 1 to 408 days could be used for estimating a mean D_{obs} value and Leachability Index.
	0.453	X	1-2	
	0.509	X	2-7	
	0.575	X	7-14	
	0.637	X	14-28	
	0.606	X	28-42	
	0.533	X	42-49	
	0.508	X	49-63	
	0.473	X	63-94	
	0.403	X	94-361	
	0.392	X	361-408	
	0.302		408-569	

Figure C.3. Analysis of Cumulative Log of Tc-99 Release versus Cumulative Log of Leaching Time Plots

<table>
<tr>
<td>

- Black dashed (0.5 slope) line is expected line when COPC depletion does not occur and Fickian diffusion is the dominant release mechanism from 0.08 day interval forward.
- Green dashed lines have 0.65 and 0.35 slopes from 0.08 day interval forward.
- Yellow markers are from 0.08 to 63 days.
- Blue markers are from beyond 63 days.

</td>
<td>Calculated Interval Slope (rise/run)</td>
<td>X = Diffusion Controlled
———
Interval Slope = 0.5 ± 0.15</td>
<td>Leach Test Interval (days)</td>
<td>Analysis per EPA Method 1315

COC = Constituent of Concern
VZP = Vadose Zone Pore Water
DIW = De-Ionized Water</td>
</tr>
<tr>
<td>


0.5 slope line manually fit to data from 42 to 841 days.
</td>
<td>0.323
0.209
0.236
0.225
0.280
0.335
0.352
0.297
0.320
0.468
0.337
0.283
0.322</td>
<td>

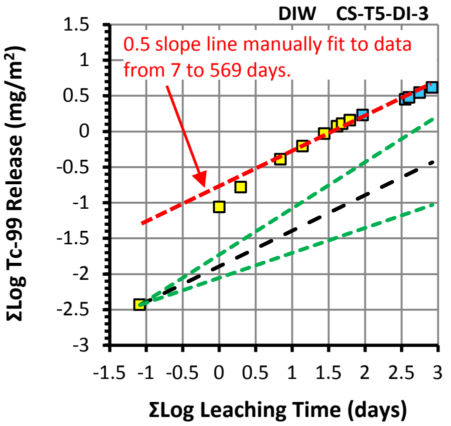
X

X

</td>
<td>0.08-1
1-2
2-7
7-14
14-28
28-42
42-49
49-63
63-94
94-361
361-408
408-569
569-841</td>
<td>

COC: **Tc-99** Eluent: **VZP**

- All points have <20% depletion.
- All points are outside the theoretical 0.5±0.15 slopes from 1 day interval forward.
- No release data from this monolith should be used for estimating a mean D_{obs} value or Leachability Index.
- Even though one can manually generate a line with slope = 0.5 for this data set from 42-841 days, the interval slopes do not indicate diffusion-controlled release.

</td>
</tr>
<tr>
<td>


0.5 slope line manually fit to data from 7 to 569 days.
</td>
<td>1.258
0.946
0.721
0.609
0.582
0.574
0.537
0.473
0.402
0.379
0.516
0.460
0.425</td>
<td>

X
X
X
X
X
X
X
X
X
</td>
<td>0.08-1
1-2
2-7
7-14
14-28
28-42
42-49
49-63
63-94
94-361
361-408
408-569
569-841</td>
<td>

COC: **Tc-99** Eluent: **DIW**
Same monolith formula as CS-T5-VZ-1, different eluent.

- All points have <20% depletion.
- All points are outside the theoretical 0.5±0.15 slopes from 0.08 day interval forward. However, slopes for intervals from 7 days to 569 days are each within 0.5±0.15.
- There may be wash off within the 0.08 to 7 day interval.
- Release data of intervals from 7 to 569 days could be used for estimating a mean D_{obs} value and Leachability Index.

</td>
</tr>
</table>

Figure C.4. Analysis of Cumulative Log of Tc-99 Release versus Cumulative Log of Leaching Time Plots

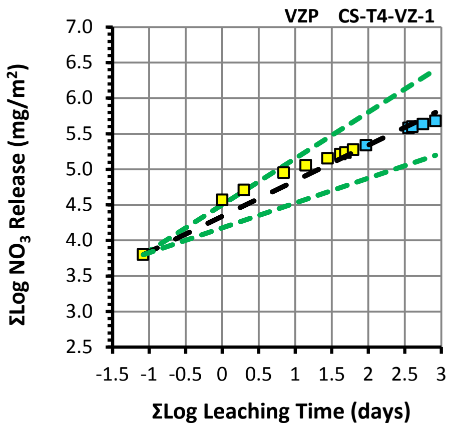
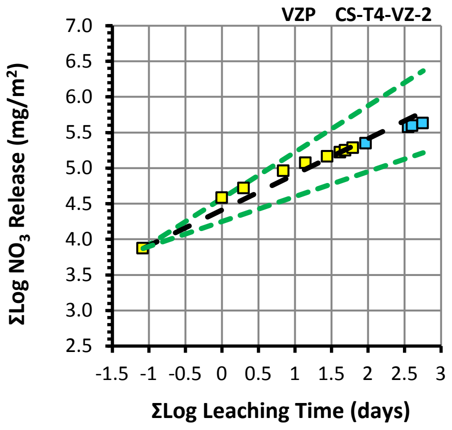
• Black dashed (0.5 slope) line is expected line when COPC depletion does not occur and Fickian diffusion is the dominant release mechanism from 0.08 day interval forward. • Green dashed lines have 0.65 and 0.35 slopes from 0.08 day interval forward. • Yellow markers are from 0.08 to 63 days. • Blue markers are from beyond 63 days.	Calculated Interval Slope (rise/run)	X = Diffusion Controlled ――― Interval Slope = 0.5 ± 0.15	Leach Test Interval (days)	Analysis per EPA Method 1315 COC = Constituent of Concern VZP = Vadose Zone Pore Water
	0.711		0.08-1	COC: **NO_3** Eluent: **VZP** • >20% depletion after 63 days. • Points are outside the theoretical 0.5±0.15 slopes from 1-2 and 2-7 day intervals. However, their interval slopes are 0.5±0.15. • Points are within the theoretical 0.5±0.15 interval slopes from 7-14 day interval forward. However, interval slopes from 7 days to 49 days and beyond 361 days are not 0.5±0.15. • No release data from this monolith should be used for estimating a mean D_{obs} value or Leachability Index.
	0.454	X	1-2	
	0.451	X	2-7	
	0.341		7-14	
	0.323		14-28	
	0.341		28-42	
	0.333		42-49	
	0.351	X	49-63	
	0.383	X	63-94	
	0.411	X	94-361	
	0.343		361-408	
	0.256		408-569	
	0.261		569-841	
	0.656		0.08-1	COC: **NO_3** Eluent: **VZP** Duplicate of CS-T4-VZ-1. • >20% depletion after 63 days. • All points are within the theoretical 0.5±0.15 slope boundaries. However, interval slopes from 14 days to 63 days and beyond 361 days are not 0.5±0.15 • No release data from this monolith should be used for estimating a mean D^{obs} value or Leachability Index.
	0.450	X	1-2	
	0.446	X	2-7	
	0.372	X	7-14	
	0.312		14-28	
	0.333		28-42	
	0.346		42-49	
	0.334		49-63	
	0.372	X	63-94	
	0.389	X	94-361	
	0.342		361-408	
	0.255		408-569	

Figure C.5. Analysis of Cumulative Log of NO_3 Release versus Cumulative Log of Leaching Time Plots

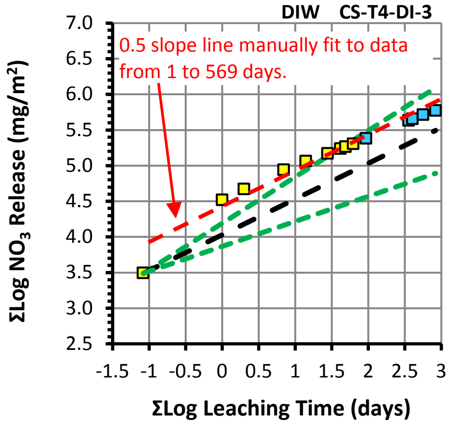
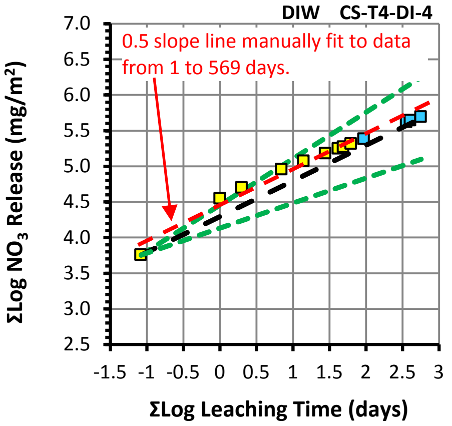
• Black dashed (0.5 slope) line is expected line when COPC depletion does not occur and Fickian diffusion is the dominant release mechanism from 0.08 day interval forward. • Green dashed lines have 0.65 and 0.35 slopes from 0.08 day interval forward. • Yellow markers are from 0.08 to 63 days. • Blue markers are from beyond 63 days.	Calculated Interval Slope (rise/run)	X = Diffusion Controlled ――― Interval Slope = 0.5 ± 0.15	Leach Test Interval (days)	Analysis per EPA Method 1315 COC = Constituent of Concern DIW = De-Ionized Water
	0.946 0.522 0.492 0.408 0.364 0.383 0.375 0.382 0.427 0.431 0.437 0.395 0.349	 X X X X X X X X X X X	0.08-1 1-2 2-7 7-14 14-28 28-42 42-49 49-63 63-94 94-361 361-408 408-569 569-841	COC: **NO_3** Eluent: **DIW** • >20% depletion after 63 days. • Points are outside the theoretical 0.5±0.15 slope boundaries from 0.08 day interval to 28-49 day interval. However, intervals from 1 day to 569 days are each within 0.5±0.15 boundary slopes despite exceeding 20% depletion after 63 days. • There may be wash off within the 0.08 to 1 day interval. • Release data for intervals from 1 to 63 days could be used for estimating a mean D^{obs} value and Leachability Index.
	0.730 0.522 0.465 0.399 0.352 0.378 0.374 0.394 0.401 0.408 0.388 0.353	 X X X X X X X X X X X	0.08-1 1-2 2-7 7-14 14-28 28-42 42-49 49-63 63-94 94-361 361-408 408-569	COC: **NO_3** Eluent: **DIW** Duplicate of CS-T4-DI-3 • >20% depletion after 49 days. • Points are within the theoretical 0.5±0.15 slope boundaries from 14 days forward. However, interval slopes from 1 day to 569 days are each within 0.5±0.15 despite exceeding 20% depletion after 49 days. • There may be wash off within the 0.08 to 1 day interval. • Release data for intervals from 1 to 49 days could be used for estimating a mean D^{obs} value and Leachability Index.

Figure C.6. Analysis of Cumulative Log of NO3 Release versus Cumulative Log of Leaching Time Plots

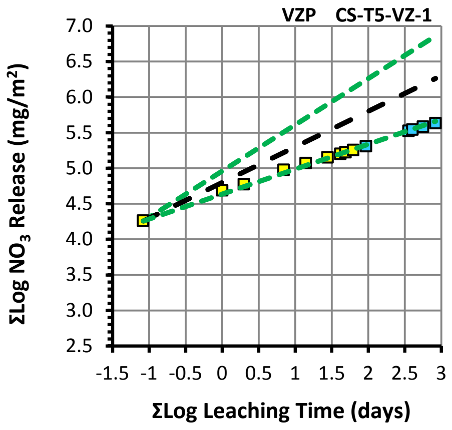
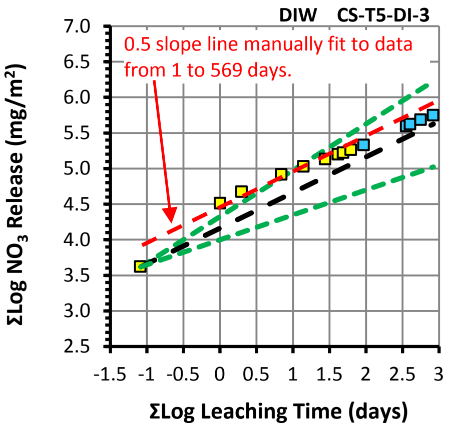
• Black dashed (0.5 slope) line is expected line when COPC depletion does not occur and Fickian diffusion is the dominant release mechanism from 0.08 day interval forward. • Green dashed lines have 0.65 and 0.35 slopes from 0.08 day interval forward. • Yellow markers are from 0.08 to 63 days. • Blue markers are from beyond 63 days.	Calculated Interval Slope (rise/run)	X = Diffusion Controlled ⎯⎯⎯⎯ Interval Slope = 0.5 ± 0.15	Leach Test Interval (days)	Analysis per EPA Method 1315 COC = Constituent of Concern VZP = Vadose Zone Pore Water DIW = De-Ionized Water
	0.394	X	0.08-1	COC: **NO$_3$** Eluent: **VZP** • >20% depletion after 94 days. • All points are along the lower boundary of the 0.35 line emanating from the 0.8 day point from 1 to 841 day intervals. However, only four of the interval slopes are within 0.5±0.15 criteria with two of these intervals exceeding the 20% source depletion. • No release data from this monolith should be used for estimating a mean D^{obs} value or Leachability Index.
	0.289		1-2	
	0.371	X	2-7	
	0.312		7-14	
	0.275		14-28	
	0.284		28-42	
	0.298		42-49	
	0.290		49-63	
	0.304		63-94	
	0.375	X	94-361	
	0.350	X	361-408	
	0.255		408-569	
	0.294		569-841	
	0.817		0.08-1	COC: **NO$_3$** Eluent: **DIW** Same monolith formula as CS-T5-VZ-1, different eluent. • >20% depletion after 63 days. • Points are close to or line emanating from the 0.08 day point. Further, the interval slopes from 1 to 569 days are each within 0.5±0.15 criteria with 4 intervals exceeding the 20% source depletion. • There may be wash off within the 0.08 to 1 day interval. • Release data for intervals from 1 to 63 days could be used for estimating a mean D^{obs} value and Leachability Index.
	0.558	X	1-2	
	0.445	X	2-7	
	0.364	X	7-14	
	0.354	X	14-28	
	0.373	X	28-42	
	0.367	X	42-49	
	0.366	X	49-63	
	0.374	X	63-94	
	0.460	X	94-361	
	0.495	X	361-408	
	0.438	X	408-569	
	0.372		569-841	

Figure C.7. Analysis of Cumulative Log of NO3 Release versus Cumulative Log of Leaching Time Plots

Table C.1. Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
5	T5HCS1-7.8AVG-2	DIW	Cr	0.237		0.08-1
				0.353	X	1-2
				0.229		2-7
				0.269		7-14
				0.295		14-28
				0.372	X	28-42
				0.541	X	42-49
				0.479	X	49-63
				0.456	X	63-77
				0.401	X	77-91
				0.425	X	91-119
				0.437	X	119-152
				0.408	X	152-187
				0.427	X	187-222
				0.420	X	222-257
				0.346		257-562
				0.467	X	562-609
				0.370	X	609-770
5	T5HCS1-7.8AVG-2	DIW	I-127	0.244		0.08-1
				0.326		1-2
				0.320		2-7
				0.301		7-14
				0.279		14-28
				0.238		28-42
				0.211		42-49
				0.198		49-63
				0.169		63-77
				0.133		77-91
				0.164		91-119
				0.174		119-152
				0.171		152-187
				0.163		187-222
				0.154		222-257
				0.149		257-562
				0.187		562-609
				0.183		609-770
5	T5HCS1-7.8AVG-2	DIW	NO_3	0.233		0.08-1
				0.301		1-2
				0.336		2-7
				0.304		7-14
				0.287		14-28
				0.217		28-42
				0.226		42-49
				0.213		49-63
				0.188		63-77
				0.150		77-91
				0.181		91-119
				0.215		119-152
				0.210		152-187
				0.210		187-222
				0.191		222-257
				0.200		257-562
				0.207		562-609
				0.212		609-770

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
5	T5HCS1-7.8AVG-2	DIW	Tc-99	0.302		0.08-1
				0.541	X	1-2
				0.555	X	2-7
				0.696		7-14
				0.608	X	14-28
				0.726		28-42
				0.691		42-49
				0.584	X	49-63
				0.442	X	63-77
				0.343		77-91
				0.384	X	91-119
				0.365	X	119-152
				0.377	X	152-187
				0.399	X	187-222
				0.430	X	222-257
				0.316		257-562
				0.526	X	562-609
				0.595	X	609-770
8	T8LCS1-5RAS-4	DIW	Cr	0.157		0.08-1
				0.112		1-2
				0.080		2-7
				0.097		7-14
				0.116		14-28
				0.131		28-42
				0.167		42-49
				0.173		49-63
				0.169		63-77
				0.130		77-91
				0.138		91-119
				0.178		119-152
				0.180		152-187
				0.184		187-222
				0.185		222-257
				0.141		257-562
				0.228		562-609
				0.161		609-770
				0.166		770-1042
8	T8LCS1-5RAS-4	DIW	I-127	0.240		0.08-1
				0.251		1-2
				0.277		2-7
				0.303		7-14
				0.317		14-28
				0.320		28-42
				0.320		42-49
				0.318		49-63
				0.289		63-77
				0.240		77-91
				0.286		91-119
				0.365	X	119-152
				0.355	X	152-187
				0.354	X	187-222
				0.318		222-257
				0.262		257-562
				0.358		562-609
				0.412	X	609-770
				0.431	X	770-1042

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
8	T8LCS1-5RAS-4	DIW	NO_3	0.134		0.08-1
				0.157		1-2
				0.209		2-7
				0.232		7-14
				0.245		14-28
				0.232		28-42
				0.279		42-49
				0.275		49-63
				0.249		63-77
				0.209		77-91
				0.238		91-119
				0.350	X	119-152
				0.337		152-187
				0.347		187-222
				0.299		222-257
				0.323		257-562
				0.380	X	562-609
				0.354	X	609-770
				0.406		770-1042
8	T8LCS1-5RAS-4	DIW	Tc-99	0.236		0.08-1
				0.346		1-2
				0.414	X	2-7
				0.434	X	7-14
				0.412	X	14-28
				0.429	X	28-42
				0.436	X	42-49
				0.447	X	49-63
				0.386	X	63-77
				0.284		77-91
				0.345		91-119
				0.438	X	119-152
				0.415	X	152-187
				0.399	X	187-222
				0.383	X	222-257
				0.343		257-562
				0.394	X	562-609
				0.327		609-770
				0.395		770-1042
10	CS-T10HCS1-5HIS-6	DIW	Cr	0.331		0.08-1
				0.413	X	1-2
				0.298		2-7
				0.250		7-14
				0.240		14-28
				0.223		28-42
				0.245		42-49
				0.241		49-63
				0.200		63-77
				0.224		77-91
				0.219		91-119
				0.208		119-152
				0.198		152-187
				0.211		187-222
				0.200		222-257
				0.160		257-562
				0.277		562-609
				0.148		609-770

Eluent: DIW = De-ionized Water Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
10	CS-T10HCS1-5HIS-6	DIW	I-127	0.303		0.08-1
				0.360	X	1-2
				0.347		2-7
				0.316		7-14
				0.279		14-28
				0.254		28-42
				0.238		42-49
				0.227		49-63
				0.184		63-77
				0.213		77-91
				0.228		91-119
				0.243		119-152
				0.246		152-187
				0.255		187-222
				0.245		222-257
				0.256		257-562
				0.299		562-609
				0.276		609-770
10	CS-T10HCS1-5HIS-6	DIW	NO_3	0.316		0.08-1
				0.359	X	1-2
				0.363	X	2-7
				0.309		7-14
				0.273		14-28
				0.245		28-42
				0.234		42-49
				0.231		49-63
				0.188		63-77
				0.219		77-91
				0.220		91-119
				0.265		119-152
				0.264		152-187
				0.284		187-222
				0.259		222-257
				0.285		257-562
				0.297		562-609
				0.259		609-770
10	CS-T10HCS1-5HIS-6	DIW	Tc-99	0.340		0.08-1
				0.308		1-2
				0.257		2-7
				0.237		7-14
				0.224		14-28
				0.232		28-42
				0.246		42-49
				0.253		49-63
				0.216		63-77
				0.252		77-91
				0.294		91-119
				0.297		119-152
				0.337		152-187
				0.361		187-222
				0.387	X	222-257
				0.313	X	257-562
				0.459		562-609
				0.422	X	609-770
					X	

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
13	T13LCS2-5AVG-2	DIW	Cr	0.457	X	0.08-1
				0.450	X	1-2
				0.325		2-7
				0.273		7-14
				0.263		14-28
				0.253		28-42
				0.286		42-49
				0.304		49-63
				0.307		63-77
				0.315		77-91
				0.300		91-119
				0.338		119-152
				0.342		152-187
				0.376	X	187-222
				0.391	X	222-257
				0.367	X	257-562
				0.619	X	562-609
				0.092		609-770
13	T13LCS2-5AVG-2	DIW	I-127	0.353	X	0.08-1
				0.363	X	1-2
				0.353	X	2-7
				0.338		7-14
				0.308		14-28
				0.288		28-42
				0.266		42-49
				0.254		49-63
				0.226		63-77
				0.232		77-91
				0.217		91-119
				0.219		119-152
				0.203		152-187
				0.194		187-222
				0.188		222-257
				0.117		257-562
				0.174		562-609
				0.205		609-770
13	T13LCS2-5AVG-2	DIW	NO_3	0.293		0.08-1
				0.327		1-2
				0.349		2-7
				0.319		7-14
				0.295		14-28
				0.260		28-42
				0.268		42-49
				0.237		49-63
				0.215		63-77
				0.218		77-91
				0.198		91-119
				0.231		119-152
				0.215		152-187
				0.218		187-222
				0.199		222-257
				0.191		257-562
				0.203		562-609
				0.203		609-770

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
13	T13LCS2-5AVG-2	DIW	Tc-99	0.492	X	0.08-1
				0.296		1-2
				0.355	X	2-7
				0.566	X	7-14
				0.652		14-28
				0.663		28-42
				0.636	X	42-49
				0.581	X	49-63
				0.489	X	63-77
				0.475	X	77-91
				0.494	X	91-119
				0.523	X	119-152
				0.553	X	152-187
				0.532	X	187-222
				0.542	X	222-257
				0.604	X	257-562
				0.707		562-609
				0.579	X	609-770
14	T14LCS2-7.8HIS-3	DIW	Cr	0.299		0.08-1
				0.327		1-2
				0.227		2-7
				0.276		7-14
				0.313		14-28
				0.322		28-42
				0.408	X	42-49
				0.426	X	49-63
				0.442	X	63-77
				0.414	X	77-91
				0.404	X	91-119
				0.374	X	119-152
				0.404	X	152-187
				0.395	X	187-222
				0.391	X	222-257
				0.326		257-562
				0.573	X	562-609
				0.344		609-770
				0.310		770-1042
14	T14LCS2-7.8HIS-3	DIW	I-127	0.304		0.08-1
				0.345		1-2
				0.332		2-7
				0.307		7-14
				0.252		14-28
				0.234		28-42
				0.221		42-49
				0.202		49-63
				0.167		63-77
				0.160		77-91
				0.165		91-119
				0.173		119-152
				0.173		152-187
				0.179		187-222
				0.196		222-257
				0.176		257-562
				0.213		562-609
				0.241		609-770
				0.241		770-1042

Eluent: DIW = De-ionized Water Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
14	T14LCS2-7.8HIS-3	DIW	NO_3	0.245		0.08-1
				0.318		1-2
				0.332		2-7
				0.300		7-14
				0.258		14-28
				0.194		28-42
				0.254		42-49
				0.216		49-63
				0.178		63-77
				0.167		77-91
				0.166		91-119
				0.189		119-152
				0.183		152-187
				0.195		187-222
				0.188		222-257
				0.220		257-562
				0.226		562-609
				0.214		609-770
				0.222		770-1042
14	CS-T14LCS2-7.8HIS-3	DIW	Tc-99	0.272		0.08-1
				0.378	X	1-2
				0.384	X	2-7
				0.392	X	7-14
				0.400	X	14-28
				0.421	X	28-42
				0.438	X	42-49
				0.416	X	49-63
				0.361	X	63-77
				0.330		77-91
				0.406	X	91-119
				0.469	X	119-152
				0.444	X	152-187
				0.431	X	187-222
				0.441	X	222-257
				0.483	X	257-562
				0.484	X	562-609
				0.395	X	609-770
				0.497		770-1042
15	T15HCS1-7.8HIS-1	DIW	Cr	0.473	X	0.08-1
				0.563	X	1-2
				0.530	X	2-7
				0.417	X	7-14
				0.257		14-28
				0.148		28-42
				0.152		42-49
				0.124		49-63
				0.138		63-77
				0.116		77-91
				0.136		91-119
				0.137		119-152
				0.144		152-187
				0.162		187-222
				0.169		222-257
				0.167		257-562
				0.280		562-609
				0.191		609-770

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
15	T15HCS1-7.8HIS-1	DIW	I-127	0.485	X	0.08-1
				0.462	X	1-2
				0.431	X	2-7
				0.396	X	7-14
				0.349		14-28
				0.318		28-42
				0.309		42-49
				0.283		49-63
				0.213		63-77
				0.214		77-91
				0.251		91-119
				0.236		119-152
				0.249		152-187
				0.238		187-222
				0.218		222-257
				0.196		257-562
				0.233		562-609
				0.206		609-770
15	T15HCS1-7.8HIS-1	DIW	NO_3	0.464	X	0.08-1
				0.428	X	1-2
				0.402	X	2-7
				0.375	X	7-14
				0.339		14-28
				0.235		28-42
				0.352	X	42-49
				0.306		49-63
				0.233		63-77
				0.223		77-91
				0.294		91-119
				0.286		119-152
				0.292		152-187
				0.297		187-222
				0.254		222-257
				0.246		257-562
				0.240		562-609
				0.225		609-770
15	T15HCS1-7.8HIS-1	DIW	Tc-99	0.380	X	0.08-1
				0.265		1-2
				0.259		2-7
				0.271		7-14
				0.224		14-28
				0.220		28-42
				0.240		42-49
				0.224		49-63
				0.170		63-77
				0.185		77-91
				0.235		91-119
				0.237		119-152
				0.282		152-187
				0.295		187-222
				0.320		222-257
				0.269		257-562
				0.494		562-609
				0.405		609-770
					X	
					X	

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
16	T16HCS1-7.8RAS-3	DIW	Cr	0.295		0.08-1
				0.181		1-2
				0.157		2-7
				0.155		7-14
				0.149		14-28
				0.145		28-42
				0.189		42-49
				0.163		49-63
				0.183		63-77
				0.179		77-91
				0.198		91-119
				0.199		119-152
				0.201		152-187
				0.208		187-222
				0.212		222-257
				0.175		257-562
				0.483	X	562-609
				0.196		609-770
16	T16HCS1-7.8RAS-3	DIW	I-127	0.433	X	0.08-1
				0.443	X	1-2
				0.407	X	2-7
				0.421	X	7-14
				0.386	X	14-28
				0.369	X	28-42
				0.341		42-49
				0.333		49-63
				0.270		63-77
				0.268		77-91
				0.316		91-119
				0.321		119-152
				0.341		152-187
				0.329		187-222
				0.322		222-257
				0.285		257-562
				0.309		562-609
				0.262		609-770
16	T16HCS1-7.8RAS-3	DIW	NO_3	0.433	X	0.08-1
				0.443	X	1-2
				0.389	X	2-7
				0.396	X	7-14
				0.382	X	14-28
				0.302		28-42
				0.355	X	42-49
				0.337		49-63
				0.279		63-77
				0.275		77-91
				0.339		91-119
				0.344		119-152
				0.362	X	152-187
				0.369	X	187-222
				0.337		222-257
				0.311		257-562
				0.305		562-609
				0.280		609-770

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
16	T16HCS1-7.8RAS-3	DIW	Tc-99	0.496	X	0.08-1
				0.438	X	1-2
				0.428	X	2-7
				0.446	X	7-14
				0.413	X	14-28
				0.467	X	28-42
				0.500	X	42-49
				0.469	X	49-63
				0.392	X	63-77
				0.389	X	77-91
				0.468	X	91-119
				0.476	X	119-152
				0.511	X	152-187
				0.494	X	187-222
				0.509	X	222-257
				0.395	X	257-562
				0.599	X	562-609
				0.433	X	609-770
17	T17LCS2-5HIA-3	DIW	Cr	0.491	X	0.08-1
				0.559	X	1-2
				0.563	X	2-7
				0.583	X	7-14
				0.553	X	14-28
				0.504	X	28-42
				0.571	X	42-49
				0.488	X	49-63
				0.557	X	63-77
				0.534	X	77-91
				1.142		91-119
				0.411	X	119-152
				0.446	X	152-187
				0.484	X	187-222
				0.470	X	222-257
				0.296		257-562
				0.579	X	562-609
				0.326		609-770
17	T17LCS2-5HIA-3	DIW	I-127	0.480	X	0.08-1
				0.477	X	1-2
				0.392	X	2-7
				0.355	X	7-14
				0.314		14-28
				0.291		28-42
				0.268		42-49
				0.265		49-63
				0.259		63-77
				0.245		77-91
				0.390	X	91-119
				0.211		119-152
				0.228		152-187
				0.240		187-222
				0.245		222-257
				0.178		257-562
				0.243		562-609
				0.274		609-770

Eluent: DIW = De-ionized Water Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
17	T17LCS2-5HIA-3	DIW	NO_3	0.473	X	0.08-1
				0.462	X	1-2
				0.382	X	2-7
				0.341		7-14
				0.305		14-28
				0.239		28-42
				0.284		42-49
				0.272		49-63
				0.263		63-77
				0.245		77-91
				0.530	X	91-119
				0.222		119-152
				0.231		152-187
				0.254		187-222
				0.240		222-257
				0.259		257-562
				0.262		562-609
				0.231		609-770
17	T17LCS2-5HIA-3	DIW	Tc-99	0.298		0.08-1
				0.505	X	1-2
				0.523	X	2-7
				0.571	X	7-14
				0.502	X	14-28
				0.551	X	28-42
				0.584	X	42-49
				0.497	X	49-63
				0.484	X	63-77
				0.449	X	77-91
				1.312		91-119
				0.366	X	119-152
				0.395	X	152-187
				0.410	X	187-222
				0.432	X	222-257
				0.491	X	257-562
				0.583	X	562-609
				0.367	X	609-770
18	T18LCS2-7.8RAS-6	DIW	Cr	0.279		0.08-1
				0.584	X	1-2
				0.375	X	2-7
				0.335		7-14
				0.437	X	14-28
				0.312		28-42
				0.662		42-49
				0.369	X	49-63
				0.625	X	63-77
				0.554	X	77-91
				0.593	X	91-119
				0.527	X	119-147
				0.620	X	147-182
				0.628	X	182-217
				0.676		217-252
				0.607	X	252-557
				0.845		557-604
				0.481	X	604-765
				0.375		765-1037

Eluent: DIW = De-ionized Water Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
18	T18LCS2-7.8RAS-6	DIW	I-127	0.400	X	0.08-1
				0.400	X	1-2
				0.371	X	2-7
				0.361	X	7-14
				0.340		14-28
				0.341		28-42
				0.334		42-49
				0.344		49-63
				0.340		63-77
				0.353	X	77-91
				0.382	X	91-119
				0.384	X	119-147
				0.422	X	147-182
				0.381	X	182-217
				0.382	X	217-252
				0.293		252-557
				0.476	X	557-604
				0.465	X	604-765
				0.463		765-1037
18	T18LCS2-7.8RAS-6	DIW	NO_3	0.388	X	0.08-1
				0.409	X	1-2
				0.365	X	2-7
				0.347		7-14
				0.334		14-28
				0.319		28-42
				0.356	X	42-49
				0.347		49-63
				0.346		63-77
				0.352	X	77-91
				0.394	X	91-119
				0.400	X	119-147
				0.430	X	147-182
				0.407	X	182-217
				0.384	X	217-252
				0.437	X	252-557
				0.502	X	557-604
				0.448	X	604-765
				0.437		765-1037
18	T18LCS2-7.8RAS-6	DIW	Tc-99	0.335		0.08-1
				0.426	X	1-2
				0.426	X	2-7
				0.320		7-14
				0.309		14-28
				0.371	X	28-42
				0.424	X	42-49
				0.437	X	49-63
				0.455	X	63-77
				0.488	X	77-91
				0.606	X	91-119
				0.665		119-147
				0.775		147-182
				0.731		182-217
				0.785		217-252
				0.887		252-557
				0.952		557-604
				0.660		604-765
				0.648		765-1037

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
21	T21LCS1-7.8HIS-6	DIW	Cr	0.412	X	0.08-1
				0.305		1-2
				0.228		2-7
				0.317		7-14
				0.416	X	14-28
				0.495	X	28-42
				0.685		42-49
				0.674		49-63
				0.604	X	63-77
				0.654		77-91
				0.363	X	91-119
				0.865		119-147
				0.774		147-182
				0.692		182-217
				0.609	X	217-252
				0.433	X	252-557
				0.663		557-604
				0.344		604-765
				0.391		765-1037
21	T21LCS1-7.8HIS-6	DIW	I-127	0.475	X	0.08-1
				0.443	X	1-2
				0.382	X	2-7
				0.354	X	7-14
				0.301		14-28
				0.267		28-42
				0.244		42-49
				0.218		49-63
				0.154		63-77
				0.148		77-91
				0.096		91-119
				0.134		119-147
				0.117		147-182
				0.102		182-217
				0.099		217-252
				0.065		252-557
				0.098		557-604
				0.112		604-765
				0.132		765-1037
21	T21LCS1-7.8HIS-6	DIW	NO_3	0.437	X	0.08-1
				0.435	X	1-2
				0.369	X	2-7
				0.333		7-14
				0.296		14-28
				0.241		28-42
				0.276		42-49
				0.247		49-63
				0.174		63-77
				0.164		77-91
				0.089		91-119
				0.168		119-147
				0.146		147-182
				0.135		182-217
				0.117		217-252
				0.116		252-557
				0.136		557-604
				0.130		604-765
				0.153		765-1037

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
21	T21LCS1-7.8HIS-6	DIW	Tc-99	0.352	X	0.08-1
				0.161		1-2
				0.083		2-7
				0.120		7-14
				0.181		14-28
				0.319		28-42
				0.614	X	42-49
				0.703		49-63
				0.673		63-77
				0.804		77-91
				0.318		91-119
				1.053		119-147
				0.882		147-182
				0.697		182-217
				0.625		217-252
				0.418	X	252-557
				0.417	X	557-604
				0.334	X	604-765
				0.402		765-1037
24	T24HCS1-5HIA-1	DIW	Cr	0.463	X	0.08-1
				0.438	X	1-2
				0.306		2-7
				0.232		7-14
				0.226		14-28
				0.190		28-42
				0.238		42-49
				0.211		49-63
				0.233		63-77
				0.221		77-91
				0.256		91-119
				0.237		119-147
				0.254		147-182
				0.252		182-217
				0.243		217-252
				0.230		252-557
				0.406		557-604
				0.229	X	604-765
				0.327		765-1037
24	T24HCS1-5HIA-1	DIW	I-127	0.445	X	0.08-1
				0.439	X	1-2
				0.383	X	2-7
				0.325		7-14
				0.323		14-28
				0.291		28-42
				0.266		42-49
				0.267		49-63
				0.214		63-77
				0.197		77-91
				0.212		91-119
				0.191		119-147
				0.183		147-182
				0.166		182-217
				0.153		217-252
				0.125		252-557
				0.140		557-604
				0.147		604-765
				0.172		765-1037

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.1 (cont). Extended Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
24	T24HCS1-5HIA-1	DIW	NO_3	0.429	X	0.08-1
				0.440	X	1-2
				0.368	X	2-7
				0.308		7-14
				0.319		14-28
				0.250		28-42
				0.293		42-49
				0.287		49-63
				0.219		63-77
				0.201		77-91
				0.238		91-119
				0.225		119-147
				0.215		147-182
				0.210		182-217
				0.184		217-252
				0.168		252-557
				0.165		557-604
				0.157		604-765
				0.181		765-1037
24	T24HCS1-5HIA-1	DIW	Tc-99	0.368	X	0.08-1
				0.243		1-2
				0.240		2-7
				0.226		7-14
				0.271		14-28
				0.319		28-42
				0.354	X	42-49
				0.366	X	49-63
				0.309		63-77
				0.308		77-91
				0.405	X	91-119
				0.410	X	119-147
				0.487	X	147-182
				0.509	X	182-217
				0.547	X	217-252
				0.392	X	252-557
				0.447	X	557-604
				0.493	X	604-765
				0.505		765-1037

Eluent: DIW = De-ionized Water Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2. Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
3	T3HCS2-7.8AVG-5	DIW	Cr	0.378	X	0.08-1
				0.319		1-2
				0.439	X	2-7
				0.474	X	7-14
				0.405	X	14-28
				0.526	X	28-42
				0.548	X	42-49
				0.393	X	49-63
				0.458	X	63-94
				0.352	X	94-361
				2.840		361-408
				0.367	X	408-569
				0.346		569-841
3	T3HCS2-7.8AVG-3	VZP	I-127	0.410	X	0.08-1
				0.453	X	1-2
				0.410	X	2-7
				0.367	X	7-14
				0.348		14-28
				0.393	X	28-42
				0.363	X	42-49
				0.396	X	49-63
				0.374	X	63-94
				0.406	X	94-361
				0.383	X	361-408
				0.329		408-569
				0.276		569-841
3	T3HCS2-7.8AVG-4	VZP	I-127	0.414	X	0.08-1
				0.443	X	1-2
				0.405	X	2-7
				0.377	X	7-14
				0.348		14-28
				0.368	X	28-42
				0.348		42-49
				0.365	X	49-63
				0.365	X	63-94
				0.390	X	94-361
				0.364	X	361-408
				0.326		408-569
				0.254		569-841
3	T3HCS2-7.8AVG-5	DIW	I-127	0.406	X	0.08-1
				0.445	X	1-2
				0.403	X	2-7
				0.379	X	7-14
				0.399	X	14-28
				0.408	X	28-42
				0.359	X	42-49
				0.418	X	49-63
				0.416	X	63-94
				0.384	X	94-361
				0.507	X	361-408
				0.455	X	408-569
				0.348		569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
3	T3HCS2-7.8AVG-3	VZP	NO_3	0.414	X	0.08-1
				0.465	X	1-2
				0.415	X	2-7
				0.380	X	7-14
				0.387	X	14-28
				0.397	X	28-42
				0.351	X	42-49
				0.385	X	49-63
				0.372	X	63-94
				0.409	X	94-361
				0.368	X	361-408
				0.258		408-569
				0.278		569-841
3	T3HCS2-7.8AVG-4	VZP	NO_3	0.417	X	0.08-1
				0.455	X	1-2
				0.406	X	2-7
				0.358	X	7-14
				0.372	X	14-28
				0.370	X	28-42
				0.342		42-49
				0.361	X	49-63
				0.369	X	63-94
				0.396	X	94-361
				0.357	X	361-408
				0.260		408-569
				0.260		569-841
3	T3HCS2-7.8AVG-5	DIW	NO_3	0.410	X	0.08-1
				0.458	X	1-2
				0.410	X	2-7
				0.395	X	7-14
				0.422	X	14-28
				0.425	X	28-42
				0.355	X	42-49
				0.408	X	49-63
				0.422	X	63-94
				0.422	X	94-361
				0.499	X	361-408
				0.419	X	408-569
				0.340		569-841
3	T3HCS2-7.8AVG-3	VZP	Tc-99	0.964		0.08-1
				0.917		1-2
				0.658		2-7
				0.560	X	7-14
				0.517	X	14-28
				0.577	X	28-42
				0.535	X	42-49
				0.610	X	49-63
				0.540	X	63-94
				0.583	X	94-361
				0.419	X	361-408
				0.231		408-569
				0.169		569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
3	T3HCS2-7.8AVG-4	VZP	Tc-99	0.934		0.08-1
				1.108		1-2
				0.809		2-7
				0.586	X	7-14
				0.597	X	14-28
				0.562	X	28-42
				0.598	X	42-49
				0.616	X	49-63
				0.538	X	63-94
				0.560	X	94-361
				0.351	X	361-408
				0.211		408-569
				0.156		569-841
3	T3HCS2-7.8AVG-5	DIW	Tc-99	1.073		0.08-1
				1.077		1-2
				0.908		2-7
				0.882		7-14
				0.795		14-28
				0.744		28-42
				0.613	X	42-49
				0.794		49-63
				0.787		63-94
				0.736		94-361
				0.909		361-408
				0.706		408-569
				0.654		569-841
5	T5HCS1-7.8AVG-6	DIW	Cr	0.330		0.08-1
				0.357	X	1-2
				0.422	X	2-7
				0.527	X	7-14
				0.353	X	14-28
				0.658		28-42
				0.736		42-49
				0.614	X	49-63
				0.468	X	63-94
				0.426	X	94-361
				1.400		361-408
				0.526	X	408-569
				0.400	X	569-841
5	T5HCS1-7.8AVG-3	VZP	I-127	0.443	X	0.08-1
				0.468	X	1-2
				0.451	X	2-7
				0.430	X	7-14
				0.417	X	14-28
				0.391	X	28-42
				0.380	X	42-49
				0.364	X	49-63
				0.328		63-94
				0.328		94-361
				0.362	X	361-408
				0.302		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
5	T5HCS1-7.8AVG-6	DIW	I-127	0.508	X	0.08-1
				0.504	X	1-2
				0.468	X	2-7
				0.459	X	7-14
				0.462	X	14-28
				0.451	X	28-42
				0.441	X	42-49
				0.408	X	49-63
				0.375	X	63-94
				0.360	X	94-361
				0.375	X	361-408
				0.346		408-569
				0.931		569-841
5	T5HCS1-7.8AVG-3	VZP	NO_3	0.412	X	0.08-1
				0.441	X	1-2
				0.418	X	2-7
				0.424	X	7-14
				0.412	X	14-28
				0.410	X	28-42
				0.380	X	42-49
				0.368	X	49-63
				0.344		63-94
				0.367	X	94-361
				0.343		361-408
				0.239		408-569
5	T5HCS1-7.8AVG-6	DIW	NO_3	0.490	X	0.08-1
				0.484	X	1-2
				0.447	X	2-7
				0.445	X	7-14
				0.468	X	14-28
				0.449	X	28-42
				0.426	X	42-49
				0.401	X	49-63
				0.391	X	63-94
				0.375	X	94-361
				0.387	X	361-408
				0.359	X	408-569
				2.040		569-841
5	T5HCS1-7.8AVG-3	VZP	Tc-99	0.944		0.08-1
				0.910		1-2
				0.634	X	2-7
				0.654		7-14
				0.506	X	14-28
				0.516	X	28-42
				0.554	X	42-49
				0.453	X	49-63
				0.421	X	63-94
				0.374	X	94-361
				0.341		361-408
				0.256		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
5	T5HCS1-7.8AVG-6	DIW	Tc-99	1.075		0.08-1
				0.976		1-2
				0.867		2-7
				0.887		7-14
				0.759		14-28
				0.726		28-42
				0.783		42-49
				0.800		49-63
				0.689		63-94
				0.500		94-361
				0.753	X	361-408
				0.619		408-569
				0.245	X	569-841
8	T8LCS1-5RAS-5	DIW	Cr	0.504	X	0.08-1
				0.263		1-2
				0.263		2-7
				0.260		7-14
				0.216		14-28
				0.391	X	28-42
				0.358	X	42-49
				0.483	X	49-63
				0.377	X	63-94
				0.326		94-361
				0.696		361-408
				0.374	X	408-569
				0.329		569-841
8	T8LCS1-5RAS-2	VZP	I-127	0.267		0.08-1
				0.390	X	1-2
				0.425	X	2-7
				0.442	X	7-14
				0.443	X	14-28
				0.428	X	28-42
				0.433	X	42-49
				0.397	X	49-63
				0.367	X	63-94
				0.343		94-361
				0.438	X	361-408
				0.409	X	408-569
				0.376	X	569-841
8	T8LCS1-5RAS-3	VZP	I-127	0.288		0.08-1
				0.403	X	1-2
				0.429	X	2-7
				0.442	X	7-14
				0.426	X	14-28
				0.436	X	28-42
				0.425	X	42-49
				0.396	X	49-63
				0.374	X	63-94
				0.341		94-361
				0.486	X	361-408
				0.410	X	408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
8	T8LCS1-5RAS-5	DIW	I-127	0.267		0.08-1
				0.380	X	1-2
				0.424	X	2-7
				0.454	X	7-14
				0.432	X	14-28
				0.427	X	28-42
				0.375	X	42-49
				0.411	X	49-63
				0.381	X	63-94
				0.302		94-361
				0.413	X	361-408
				0.357	X	408-569
				0.444	X	569-841
8	T8LCS1-5RAS-2	VZP	NO_3	0.214		0.08-1
				0.334		1-2
				0.389	X	2-7
				0.432	X	7-14
				0.448	X	14-28
				0.433	X	28-42
				0.412	X	42-49
				0.376	X	49-63
				0.353	X	63-94
				0.351	X	94-361
				0.411	X	361-408
				0.335		408-569
				0.384	X	569-841
8	T8LCS1-5RAS-3	VZP	NO_3	0.227		0.08-1
				0.364	X	1-2
				0.413	X	2-7
				0.432	X	7-14
				0.458	X	14-28
				0.425	X	28-42
				0.397	X	42-49
				0.377	X	49-63
				0.293		63-94
				0.358	X	94-361
				0.437	X	361-408
				0.280		408-569
8	T8LCS1-5RAS-5	DIW	NO_3	0.223		0.08-1
				0.347		1-2
				0.401	X	2-7
				0.427	X	7-14
				0.441	X	14-28
				0.436	X	28-42
				0.379	X	42-49
				0.391	X	49-63
				0.376	X	63-94
				0.360	X	94-361
				0.408	X	361-408
				0.381	X	408-569
				0.418	X	569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
8	T8LCS1-5RAS-2	VZP	Tc-99	0.267		0.08-1
				0.368	X	1-2
				0.393	X	2-7
				0.369	X	7-14
				0.287		14-28
				0.326		28-42
				0.358	X	42-49
				0.331		49-63
				0.278		63-94
				0.267		94-361
				0.478	X	361-408
				0.692		408-569
				0.614	X	569-841
8	T8LCS1-5RAS-3	VZP	Tc-99	0.270		0.08-1
				0.357	X	1-2
				0.378	X	2-7
				0.357	X	7-14
				0.264		14-28
				0.304		28-42
				0.349		42-49
				0.306		49-63
				0.282		63-94
				0.281		94-361
				0.520	X	361-408
				0.561	X	408-569
8	T8LCS1-5RAS-5	DIW	Tc-99	0.292		0.08-1
				0.419	X	1-2
				0.523	X	2-7
				0.603	X	7-14
				0.604	X	14-28
				0.560	X	28-42
				0.533	X	42-49
				0.570	X	49-63
				0.538	X	63-94
				0.444	X	94-361
				0.475	X	361-408
				0.394	X	408-569
				0.411	X	569-841
10	T10HCS1-5HIS-5	DIW	Cr	0.448	X	0.08-1
				0.393	X	1-2
				0.337		2-7
				0.277		7-14
				0.189		14-28
				0.225		28-42
				0.221		42-49
				0.249		49-63
				0.201		63-94
				0.225		94-361
				0.466	X	361-408
				0.258		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
10	T10HCS1-5HIS-3	VZP	I-127	0.364	X	0.08-1
				0.434	X	1-2
				0.445	X	2-7
				0.441	X	7-14
				0.443	X	14-28
				0.457	X	28-42
				0.435	X	42-49
				0.423	X	49-63
				0.417	X	63-94
				0.411	X	94-361
				0.399	X	361-408
				0.318		408-569
10	T10HCS1-5HIS-4	VZP	I-127	0.371	X	0.08-1
				0.426	X	1-2
				0.431	X	2-7
				0.432	X	7-14
				0.429	X	14-28
				0.413	X	28-42
				0.435	X	42-49
				0.438	X	49-63
				0.410	X	63-94
				0.414	X	94-361
				0.395	X	361-408
				0.333		408-569
10	T10HCS1-5HIS-5	DIW	I-127	0.360	X	0.08-1
				0.436	X	1-2
				0.443	X	2-7
				0.457	X	7-14
				0.471	X	14-28
				0.478	X	28-42
				0.466	X	42-49
				0.487	X	49-63
				0.473	X	63-94
				0.423	X	94-361
				0.508	X	361-408
				0.473	X	408-569
10	T10HCS1-5HIS-3	VZP	NO_3	0.353	X	0.08-1
				0.434	X	1-2
				0.434	X	2-7
				0.431	X	7-14
				0.440	X	14-28
				0.428	X	28-42
				0.403	X	42-49
				0.393	X	49-63
				0.395	X	63-94
				0.404	X	94-361
				0.383	X	361-408
				0.218		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
10	T10HCS1-5HIS-4	VZP	NO_3	0.370	X	0.08-1
				0.429	X	1-2
				0.416	X	2-7
				0.416	X	7-14
				0.419	X	14-28
				0.410	X	28-42
				0.387	X	42-49
				0.380	X	49-63
				0.377	X	63-94
				0.393	X	94-361
				0.379	X	361-408
				0.229		408-569
10	T10HCS1-5HIS-5	DIW	NO_3	0.352	X	0.08-1
				0.434	X	1-2
				0.437	X	2-7
				0.436	X	7-14
				0.460	X	14-28
				0.451	X	28-42
				0.412	X	42-49
				0.436	X	49-63
				0.438	X	63-94
				0.460	X	94-361
				0.469	X	361-408
				0.413	X	408-569
10	T10HCS1-5HIS-3	VZP	Tc-99	0.372	X	0.08-1
				0.496	X	1-2
				0.482	X	2-7
				0.522	X	7-14
				0.457	X	14-28
				0.422	X	28-42
				0.432	X	42-49
				0.407	X	49-63
				0.336		63-94
				0.376	X	94-361
				0.330		361-408
				0.279		408-569
10	T10HCS1-5HIS-4	VZP	Tc-99	0.457	X	0.08-1
				0.594	X	1-2
				0.560	X	2-7
				0.570	X	7-14
				0.476	X	14-28
				0.453	X	28-42
				0.524	X	42-49
				0.427	X	49-63
				0.359	X	63-94
				0.339		94-361
				0.449	X	361-408
				0.311		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
10	T10HCS1-5HIS-5	DIW	Tc-99	0.588	X	0.08-1
				0.846		1-2
				0.791		2-7
				0.706		7-14
				0.681		14-28
				0.607	X	28-42
				0.597	X	42-49
				0.619	X	49-63
				0.585	X	63-94
				0.501	X	94-361
				0.529	X	361-408
				0.491	X	408-569
13	T13LCS2-5AVG-5	DIW	Cr	0.331		0.08-1
				0.525	X	1-2
				0.484	X	2-7
				0.396	X	7-14
				0.241		14-28
				0.488	X	28-42
				0.589	X	42-49
				6.166		49-63
				0.108		63-94
				0.177		94-361
				0.499	X	361-408
				0.229		408-569
13	T13LCS2-5AVG-3	VZP	I-127	0.524	X	0.08-1
				0.520	X	1-2
				0.473	X	2-7
				0.440	X	7-14
				0.398	X	14-28
				0.367	X	28-42
				0.337		42-49
				0.325		49-63
				0.319		63-94
				0.360	X	94-361
				0.513	X	361-408
				0.463	X	408-569
13	T13LCS2-5AVG-4	VZP	I-127	0.429	X	0.08-1
				0.437	X	1-2
				0.427	X	2-7
				0.414	X	7-14
				0.402	X	14-28
				0.341		28-42
				0.345		42-49
				0.332		49-63
				0.320		63-94
				0.340		94-361
				0.521	X	361-408
				0.476	X	408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
13	T13LCS2-5AVG-5	DIW	I-127	0.462	X	0.08-1
				0.488	X	1-2
				0.466	X	2-7
				0.434	X	7-14
				0.416	X	14-28
				0.432	X	28-42
				0.413	X	42-49
				0.434	X	49-63
				0.404	X	63-94
				0.311		94-361
				0.433	X	361-408
				0.478	X	408-569
13	T13LCS2-5AVG-3	VZP	NO_3	0.512	X	0.08-1
				0.769		1-2
				0.382	X	2-7
				0.374	X	7-14
				0.358	X	14-28
				0.336		28-42
				0.310		42-49
				0.301		49-63
				0.298		63-94
				0.359	X	94-361
				0.471	X	361-408
				0.510	X	408-569
13	T13LCS2-5AVG-4	VZP	NO_3	0.428	X	0.08-1
				0.416	X	1-2
				0.392	X	2-7
				0.386	X	7-14
				0.378	X	14-28
				0.349		28-42
				0.340		42-49
				0.311		49-63
				0.298		63-94
				0.348		94-361
				0.490	X	361-408
				0.352	X	408-569
13	T13LCS2-5AVG-5	DIW	NO_3	0.461	X	0.08-1
				0.461	X	1-2
				0.427	X	2-7
				0.407	X	7-14
				0.426	X	14-28
				0.420	X	28-42
				0.384	X	42-49
				0.393	X	49-63
				0.377	X	63-94
				0.367	X	94-361
				0.420	X	361-408
				0.412	X	408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
13	T13LCS2-5AVG-3	VZP	Tc-99	0.522	X	0.08-1
				0.531	X	1-2
				0.516	X	2-7
				0.503	X	7-14
				0.513	X	14-28
				0.521	X	28-42
				0.715		42-49
				0.615	X	49-63
				0.585	X	63-94
				0.776		94-361
				0.931		361-408
				0.669		408-569
13	T13LCS2-5AVG-4	VZP	Tc-99	0.383	X	0.08-1
				0.418	X	1-2
				0.445	X	2-7
				0.455	X	7-14
				0.531	X	14-28
				0.594	X	28-42
				0.762		42-49
				0.638	X	49-63
				0.616	X	63-94
				0.695		94-361
				0.859		361-408
				0.837		408-569
13	T13LCS2-5AVG-5	DIW	Tc-99	0.635	X	0.08-1
				0.662		1-2
				0.701		2-7
				0.787		7-14
				0.812		14-28
				0.842		28-42
				0.832		42-49
				0.873		49-63
				0.837		63-94
				0.916		94-361
				0.997		361-408
				0.729		408-569
14	T14LCS2-7.8HIS-6	DIW	Cr	0.435	X	0.08-1
				0.421	X	1-2
				0.473	X	2-7
				0.317		7-14
				0.157		14-28
				0.329		28-42
				0.548	X	42-49
				0.439	X	49-63
				0.238		63-94
				0.227		94-361
				0.517	X	361-408
				0.291		408-569
				0.202		569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
14	T14LCS2-7.8HIS-4	VZP	I-127	0.497	X	0.08-1
				0.554	X	1-2
				0.504	X	2-7
				0.456	X	7-14
				0.396	X	14-28
				0.465	X	28-42
				0.384	X	42-49
				0.424	X	49-63
				0.411	X	63-94
				0.453	X	94-361
				0.493	X	361-408
				0.423	X	408-569
				0.390	X	569-841
14	T14LCS2-7.8HIS-5	VZP	I-127	0.449	X	0.08-1
				0.558	X	1-2
				0.525	X	2-7
				0.477	X	7-14
				0.474	X	14-28
				0.444	X	28-42
				0.404	X	42-49
				0.406	X	49-63
				0.388	X	63-94
				0.396	X	94-361
				0.451	X	361-408
				0.450	X	408-569
14	T14LCS2-7.8HIS-6	DIW	I-127	0.511	X	0.08-1
				0.619	X	1-2
				0.541	X	2-7
				0.472	X	7-14
				0.437	X	14-28
				0.419	X	28-42
				0.425	X	42-49
				0.414	X	49-63
				0.417	X	63-94
				0.377	X	94-361
				0.485	X	361-408
				0.478	X	408-569
				0.411	X	569-841
14	T14LCS2-7.8HIS-4	VZP	NO_3	0.487	X	0.08-1
				0.546	X	1-2
				0.491	X	2-7
				0.442	X	7-14
				0.387	X	14-28
				0.462	X	28-42
				0.368	X	42-49
				0.400	X	49-63
				0.392	X	63-94
				0.416	X	94-361
				0.441	X	361-408
				0.368	X	408-569
				0.397	X	569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
14	T14LCS2-7.8HIS-5	VZP	NO_3	0.444	X	0.08-1
				0.559	X	1-2
				0.499	X	2-7
				0.466	X	7-14
				0.481	X	14-28
				0.439	X	28-42
				0.401	X	42-49
				0.376	X	49-63
				0.376	X	63-94
				0.392	X	94-361
				0.422	X	361-408
				0.378	X	408-569
14	T14LCS2-7.8HIS-6	DIW	NO_3	0.510	X	0.08-1
				0.601	X	1-2
				0.509	X	2-7
				0.448	X	7-14
				0.443	X	14-28
				0.422	X	28-42
				0.398	X	42-49
				0.388	X	49-63
				0.408	X	63-94
				0.452	X	94-361
				0.456	X	361-408
				0.422	X	408-569
				0.375	X	569-841
14	T14LCS2-7.8HIS-4	VZP	Tc-99	0.837		0.08-1
				0.941		1-2
				0.724		2-7
				0.656		7-14
				0.566	X	14-28
				0.936		28-42
				0.576	X	42-49
				0.679		49-63
				0.570	X	63-94
				0.400	X	94-361
				0.305		361-408
				0.238		408-569
				0.196		569-841
14	T14LCS2-7.8HIS-5	VZP	Tc-99	0.818		0.08-1
				0.984		1-2
				0.781		2-7
				0.730		7-14
				0.634	X	14-28
				0.600	X	28-42
				0.583	X	42-49
				0.538	X	49-63
				0.564	X	63-94
				0.411	X	94-361
				0.373	X	361-408
				0.318		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
14	T14LCS2-7.8HIS-6	DIW	Tc-99	0.640	X	0.08-1
				0.731		1-2
				0.657		2-7
				0.614	X	7-14
				0.561	X	14-28
				0.501	X	28-42
				0.540	X	42-49
				0.539	X	49-63
				0.584	X	63-94
				0.588	X	94-361
				0.452	X	361-408
				0.343		408-569
				0.384	X	408-841
15	T15HCS1-7.8HIS-6	DIW	Cr	0.417	X	0.08-1
				0.391	X	1-2
				0.878		2-7
				0.553	X	7-14
				0.460	X	14-28
				0.526	X	28-42
				0.368	X	42-49
				0.436	X	49-63
				0.295		63-94
				0.238		94-361
				0.570	X	361-408
				0.282		408-569
15	T15HCS1-7.8HIS-3	VZP	I-127	0.419	X	0.08-1
				0.440	X	1-2
				0.427	X	2-7
				0.423	X	7-14
				0.437	X	14-28
				0.434	X	28-42
				0.389	X	42-49
				0.371	X	49-63
				0.344		63-94
				0.334		94-361
				0.312		361-408
				0.283		408-569
15	T15HCS1-7.8HIS-5	VZP	I-127	0.427	X	0.08-1
				0.446	X	1-2
				0.444	X	2-7
				0.421	X	7-14
				0.410	X	14-28
				0.403	X	28-42
				0.392	X	42-49
				0.376	X	49-63
				0.340		63-94
				0.330		94-361
				0.310		361-408
				0.285		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
15	T15HCS1-7.8HIS-6	DIW	I-127	0.441	X	0.08-1
				0.468	X	1-2
				0.460	X	2-7
				0.448	X	7-14
				0.483	X	14-28
				0.480	X	28-42
				0.375	X	42-49
				0.475	X	49-63
				0.421	X	63-94
				0.355	X	94-361
				0.381	X	361-408
				0.367	X	408-569
15	T15HCS1-7.8HIS-3	VZP	NO_3	0.418	X	0.08-1
				0.442	X	1-2
				0.433	X	2-7
				0.426	X	7-14
				0.437	X	14-28
				0.428	X	28-42
				0.391	X	42-49
				0.361	X	49-63
				0.350	X	63-94
				0.355	X	94-361
				0.313		361-408
				0.217		408-569
15	T15HCS1-7.8HIS-5	VZP	NO_3	0.411	X	0.08-1
				0.473	X	1-2
				0.442	X	2-7
				0.426	X	7-14
				0.429	X	14-28
				0.419	X	28-42
				0.384	X	42-49
				0.372	X	49-63
				0.344		63-94
				0.358	X	94-361
				0.309		361-408
				0.219		408-569
15	T15HCS1-7.8HIS-6	DIW	NO_3	0.434	X	0.08-1
				0.453	X	1-2
				0.444	X	2-7
				0.436	X	7-14
				0.485	X	14-28
				0.481	X	28-42
				0.358	X	42-49
				0.435	X	49-63
				0.421	X	63-94
				0.382	X	94-361
				0.379	X	361-408
				0.333		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
15	T15HCS1-7.8HIS--3	VZP	Tc-99	0.906 0.854 0.679 0.648 0.530 0.566 0.626 0.509 0.483 0.424 0.400 0.194	 X X X X X X X X 	0.08-1 1-2 2-7 7-14 14-28 28-42 42-49 49-63 63-94 94-361 361-408 408-569
15	T15HCS1-7.8HIS--5	VZP	Tc-99	0.913 0.895 0.675 0.644 0.533 0.462 0.561 0.528 0.423 0.378 0.416 0.181	 X X X X X X X X 	0.08-1 1-2 2-7 7-14 14-28 28-42 42-49 49-63 63-94 94-361 361-408 408-569
15	T15HCS1-7.8HIS-6	DIW	Tc-99	0.838 0.931 0.910 0.926 0.781 0.737 0.581 0.716 0.636 0.495 0.691 0.528	 X X X X	0.08-1 1-2 2-7 7-14 14-28 28-42 42-49 49-63 63-94 94-361 361-408 408-569
16	T16HCS1-7.8RAS-6	DIW	Cr	0.475 0.210 0.257 0.260 0.218 0.340 0.365 0.382 0.314 0.257 0.844 0.304	X X X 	0.08-1 1-2 2-7 7-14 14-28 28-42 42-49 49-63 63-94 94-361 361-408 408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
16	T16HCS1-7.8RAS-4	VZP	I-127	0.354	X	0.08-1
				0.426	X	1-2
				0.445	X	2-7
				0.443	X	7-14
				0.422	X	14-28
				0.416	X	28-42
				0.400	X	42-49
				0.392	X	49-63
				0.374	X	63-94
				0.349		94-361
				0.405	X	361-408
				0.283		408-569
16	T16HCS1-7.8RAS-5	VZP	I-127	0.367	X	0.08-1
				0.440	X	1-2
				0.449	X	2-7
				0.431	X	7-14
				0.407	X	14-28
				0.400	X	28-42
				0.377	X	42-49
				0.394	X	49-63
				0.390	X	63-94
				0.365	X	94-361
				0.381	X	361-408
				0.304		408-569
16	T16HCS1-7.8RAS-6	DIW	I-127	0.373	X	0.08-1
				0.429	X	1-2
				0.421	X	2-7
				0.424	X	7-14
				0.439	X	14-28
				0.489	X	28-42
				0.401	X	42-49
				0.478	X	49-63
				0.470	X	63-94
				0.408	X	94-361
				0.393	X	361-408
				0.350	X	408-569
16	T16HCS1-7.8RAS-4	VZP	NO_3	0.344		0.08-1
				0.427	X	1-2
				0.433	X	2-7
				0.433	X	7-14
				0.430	X	14-28
				0.422	X	28-42
				0.376	X	42-49
				0.368	X	49-63
				0.369	X	63-94
				0.369	X	94-361
				0.363	X	361-408
				0.242		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
16	T16HCS1-7.8RAS-5	VZP	NO_3	0.362	X	0.08-1
				0.439	X	1-2
				0.428	X	2-7
				0.426	X	7-14
				0.426	X	14-28
				0.411	X	28-42
				0.357	X	42-49
				0.377	X	49-63
				0.381	X	63-94
				0.382	X	94-361
				0.375	X	361-408
				0.266		408-569
16	T16HCS1-7.8RAS-6	DIW	NO_3	0.360	X	0.08-1
				0.416	X	1-2
				0.413	X	2-7
				0.417	X	7-14
				0.450	X	14-28
				0.469	X	28-42
				0.404	X	42-49
				0.448	X	49-63
				0.452	X	63-94
				0.421	X	94-361
				0.395	X	361-408
				0.334		408-569
16	T16HCS1-7.8RAS-4	VZP	Tc-99	0.537	X	0.08-1
				0.551	X	1-2
				0.458	X	2-7
				0.506	X	7-14
				0.480	X	14-28
				0.523	X	28-42
				0.570	X	42-49
				0.532	X	49-63
				0.427	X	63-94
				0.358	X	94-361
				0.342		361-408
				0.199		408-569
16	T16HCS1-7.8RAS-5	VZP	Tc-99	0.686		0.08-1
				0.592	X	1-2
				0.456	X	2-7
				0.525	X	7-14
				0.479	X	14-28
				0.533	X	28-42
				0.509	X	42-49
				0.565	X	49-63
				0.468	X	63-94
				0.350	X	94-361
				0.401	X	361-408
				0.210		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
16	T16HCS1-7.8RAS-6	DIW	Tc-99	0.749		0.08-1
				0.595	X	1-2
				0.500	X	2-7
				0.514	X	7-14
				0.558	X	14-28
				0.578	X	28-42
				0.559	X	42-49
				0.586	X	49-63
				0.561	X	63-94
				0.481	X	94-361
				0.517	X	361-408
				0.377	X	408-569
17	T17LCS2-5HIA-6	DIW	Cr	0.367	X	0.08-1
				0.487	X	1-2
				0.666		2-7
				0.537	X	7-14
				0.381	X	14-28
				0.632	X	28-42
				0.586	X	42-49
				0.643	X	49-63
				0.524	X	63-94
				0.528	X	94-361
				0.774		361-408
				0.414	X	408-569
17	T17LCS2-5HIA-2	VZP	I-127	0.409	X	0.08-1
				0.465	X	1-2
				0.454	X	2-7
				0.434	X	7-14
				0.376	X	14-28
				0.314		28-42
				0.309		42-49
				0.315		49-63
				0.310		63-94
				0.361	X	94-361
				0.474	X	361-408
				0.438	X	408-569
17	T17LCS2-5HIA-4	VZP	I-127	0.395	X	0.08-1
				0.455	X	1-2
				0.453	X	2-7
				0.406	X	7-14
				0.336		14-28
				0.326		28-42
				0.320		42-49
				0.313		49-63
				0.319		63-94
				0.433	X	94-361
				0.537	X	361-408
				0.462	X	408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
17	T17LCS2-5HIA-6	DIW	I-127	0.440	X	0.08-1
				0.456	X	1-2
				0.457	X	2-7
				0.444	X	7-14
				0.420	X	14-28
				0.417	X	28-42
				0.356	X	42-49
				0.387	X	49-63
				0.377	X	63-94
				0.260		94-361
				0.343		361-408
				0.393	X	408-569
17	T17LCS2-5HIA-2	VZP	NO_3	0.385	X	0.08-1
				0.460	X	1-2
				0.442	X	2-7
				0.422	X	7-14
				0.391	X	14-28
				0.331		28-42
				0.306		42-49
				0.305		49-63
				0.293		63-94
				0.360	X	94-361
				0.465	X	361-408
				0.323		408-569
17	T17LCS2-5HIA-4	VZP	NO_3	0.384	X	0.08-1
				0.469	X	1-2
				0.443	X	2-7
				0.397	X	7-14
				0.352	X	14-28
				0.328		28-42
				0.310		42-49
				0.300		49-63
				0.301		63-94
				0.413	X	94-361
				0.529	X	361-408
				0.357	X	408-569
17	T17LCS2-5HIA-6	DIW	NO_3	0.428	X	0.08-1
				0.449	X	1-2
				0.508	X	2-7
				0.394	X	7-14
				0.398	X	14-28
				0.375	X	28-42
				0.314		42-49
				0.349		49-63
				0.351	X	63-94
				0.337		94-361
				0.365	X	361-408
				0.327		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
17	T17LCS2-5HIA-2	VZP	Tc-99	0.686		0.08-1
				0.804		1-2
				0.555	X	2-7
				0.528	X	7-14
				0.479	X	14-28
				0.505	X	28-42
				0.644	X	42-49
				0.594	X	49-63
				0.527	X	63-94
				0.478	X	94-361
				0.617	X	361-408
				0.595	X	408-569
17	T17LCS2-5HIA-4	VZP	Tc-99	0.647	X	0.08-1
				0.768		1-2
				0.572	X	2-7
				0.558	X	7-14
				0.585	X	14-28
				0.564	X	28-42
				0.675		42-49
				0.572	X	49-63
				0.551	X	63-94
				0.524	X	94-361
				0.995		361-408
				0.668		408-569
17	T17LCS2-5HIA-6	DIW	Tc-99	1.096		0.08-1
				0.823		1-2
				0.656		2-7
				0.691		7-14
				0.615	X	14-28
				0.623	X	28-42
				0.614	X	42-49
				0.584	X	49-63
				0.603	X	63-94
				0.625	X	94-361
				0.717		361-408
				0.424	X	408-569
18	T18LCS2-7.8RAS-4	DIW	Cr	0.279		0.08-1
				0.585	X	1-2
				0.383	X	2-7
				0.296		7-14
				0.224		14-28
				0.433	X	28-42
				0.739		42-49
				0.642	X	49-63
				0.452	X	63-94
				0.392	X	94-361
				0.747		361-408
				0.451	X	408-569
				0.419	X	569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
18	T18LCS2-7.8RAS-2	VZP	I-127	0.458	X	0.08-1
				0.548	X	1-2
				0.503	X	2-7
				0.468	X	7-14
				0.416	X	14-28
				0.359	X	28-42
				0.344		42-49
				0.362	X	49-63
				0.337		63-94
				0.366	X	94-361
				0.522	X	361-408
				0.514	X	408-569
				0.476	X	569-841
18	T18LCS2-7.8RAS-3	VZP	I-127	0.455	X	0.08-1
				0.491	X	1-2
				0.482	X	2-7
				0.443	X	7-14
				0.423	X	14-28
				0.387	X	28-42
				0.335		42-49
				0.380	X	49-63
				0.372	X	63-94
				0.391	X	94-361
				0.486	X	361-408
				0.416	X	408-569
18	T18LCS2-7.8RAS-4	DIW	I-127	0.402	X	0.08-1
				0.463	X	1-2
				0.425	X	2-7
				0.413	X	7-14
				0.416	X	14-28
				0.415	X	28-42
				0.369	X	42-49
				0.421	X	49-63
				0.421	X	63-94
				0.354	X	94-361
				0.479	X	361-408
				0.535	X	408-569
				0.531	X	569-841
18	T18LCS2-7.8RAS-2	VZP	NO_3	0.464	X	0.08-1
				0.547	X	1-2
				0.475	X	2-7
				0.444	X	7-14
				0.417	X	14-28
				0.366	X	28-42
				0.330		42-49
				0.335		49-63
				0.337		63-94
				0.397	X	94-361
				0.473	X	361-408
				0.415	X	408-569
				0.466	X	569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
18	T18LCS2-7.8RAS-3	VZP	NO_3	0.461	X	0.08-1
				0.487	X	1-2
				0.454	X	2-7
				0.435	X	7-14
				0.436	X	14-28
				0.389	X	28-42
				0.307		42-49
				0.362	X	49-63
				0.363	X	63-94
				0.397	X	94-361
				0.425	X	361-408
				0.355	X	408-569
18	T18LCS2-7.8RAS-4	DIW	NO_3	0.415	X	0.08-1
				0.459	X	1-2
				0.414	X	2-7
				0.404	X	7-14
				0.425	X	14-28
				0.423	X	28-42
				0.346		42-49
				0.401	X	49-63
				0.405	X	63-94
				0.415	X	94-361
				0.458	X	361-408
				0.456	X	408-569
				0.492	X	569-841
18	T18LCS2-7.8RAS-2	VZP	Tc-99	0.726		0.08-1
				0.815		1-2
				0.645	X	2-7
				0.591	X	7-14
				0.523	X	14-28
				0.526	X	28-42
				0.512	X	42-49
				0.554	X	49-63
				0.490	X	63-94
				0.446	X	94-361
				0.510	X	361-408
				0.458	X	408-569
				0.211		569-841
18	T18LCS2-7.8RAS-3	VZP	Tc-99	1.456		0.08-1
				0.800		1-2
				0.582	X	2-7
				0.549	X	7-14
				0.494	X	14-28
				0.458	X	28-42
				0.409	X	42-49
				0.477	X	49-63
				0.484	X	63-94
				0.420	X	94-361
				0.474	X	361-408
				0.313		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
18	T18LCS2-7.8RAS-4	DIW	Tc-99	0.738		0.08-1
				0.705		1-2
				0.569	X	2-7
				0.580	X	7-14
				0.556	X	14-28
				0.594	X	28-42
				0.546	X	42-49
				0.597	X	49-63
				0.616	X	63-94
				0.641	X	94-361
				0.490	X	361-408
				0.426	X	408-569
				0.678		569-841
21	T21LCS1-7.8HIS-5	DIW	Cr	0.381	X	0.08-1
				0.376	X	1-2
				0.448	X	2-7
				0.467	X	7-14
				0.444	X	14-28
				0.675		28-42
				0.648	X	42-49
				0.770		49-63
				0.697		63-94
				0.531	X	94-361
				0.738		361-408
				0.415	X	408-569
				0.410	X	569-841
21	T21LCS1-7.8HIS-2	VZP	I-127	0.447	X	0.08-1
				0.486	X	1-2
				0.376	X	2-7
				0.440	X	7-14
				0.422	X	14-28
				0.388	X	28-42
				0.326		42-49
				0.357	X	49-63
				0.332		63-94
				0.290		94-361
				0.284		361-408
				0.251		408-569
				0.254		
21	T21LCS1-7.8HIS-4	VZP	I-127	0.443	X	0.08-1
				0.475	X	1-2
				0.449	X	2-7
				0.422	X	7-14
				0.393	X	14-28
				0.367	X	28-42
				0.345		42-49
				0.345		49-63
				0.328		63-94
				0.291		94-361
				0.248		361-408
				0.218		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
21	T21LCS1-7.8HIS-5	DIW	I-127	0.483	X	0.08-1
				0.508	X	1-2
				0.468	X	2-7
				0.459	X	7-14
				0.427	X	14-28
				0.394	X	28-42
				0.310		42-49
				0.381	X	49-63
				0.346		63-94
				0.229		94-361
				0.251		361-408
				0.226		408-569
				0.196		569-841
21	T21LCS1-7.8HIS-2	VZP	NO_3	0.435	X	0.08-1
				0.486	X	1-2
				0.377	X	2-7
				0.449	X	7-14
				0.448	X	14-28
				0.403	X	28-42
				0.317		42-49
				0.361	X	49-63
				0.338		63-94
				0.296		94-361
				0.288		361-408
				0.209		408-569
				0.251		569-841
21	T21LCS1-7.8HIS-4	VZP	NO_3	0.424	X	0.08-1
				0.476	X	1-2
				0.435	X	2-7
				0.422	X	7-14
				0.421	X	14-28
				0.386	X	28-42
				0.326		42-49
				0.345		49-63
				0.334		63-94
				0.297		94-361
				0.248		361-408
				0.179		408-569
21	T21LCS1-7.8HIS-5	DIW	NO_3	0.466	X	0.08-1
				0.495	X	1-2
				0.470	X	2-7
				0.451	X	7-14
				0.442	X	14-28
				0.400	X	28-42
				0.311		42-49
				0.368	X	49-63
				0.348		63-94
				0.280		94-361
				0.262		361-408
				0.208		408-569
				0.189		569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
21	T21LCS1-7.8HIS-2	VZP	Tc-99	0.717		0.08-1
				1.086		1-2
				0.635	X	2-7
				0.959		7-14
				0.803		14-28
				0.823		28-42
				0.761		42-49
				0.930		49-63
				0.795		63-94
				0.707		94-361
				0.472		361-408
				0.325	X	408-569
				0.321		569-841
21	T21LCS1-7.8HIS-4	VZP	Tc-99	0.663		0.08-1
				1.076		1-2
				0.838		2-7
				0.951		7-14
				0.870		14-28
				0.809		28-42
				0.727		42-49
				0.845		49-63
				0.735		63-94
				0.688		94-361
				0.402		361-408
				0.292	X	408-569
21	T21LCS1-7.8HIS-5	DIW	Tc-99	0.987		0.08-1
				1.089		1-2
				1.402		2-7
				1.380		7-14
				1.031		14-28
				0.910		28-42
				0.841		42-49
				0.897		49-63
				0.789		63-94
				0.543		94-361
				0.553	X	361-408
				0.448	X	408-569
				0.419	X	569-841
					X	
24	T24HCS1-5HIA-6	DIW	Cr	0.406	X	0.08-1
				0.362	X	1-2
				0.536	X	2-7
				0.461	X	7-14
				0.375	X	14-28
				0.461	X	28-42
				0.509	X	42-49
				0.492	X	49-63
				0.432	X	63-94
				0.358	X	94-361
				0.681		361-408
				0.390	X	408-569
				0.390	X	569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
24	T24HCS1-5HIA-4	VZP	I-127	0.396	X	0.08-1
				0.422	X	1-2
				0.391	X	2-7
				0.390	X	7-14
				0.365	X	14-28
				0.349		28-42
				0.350	X	42-49
				0.336		49-63
				0.315		63-94
				0.297		94-361
				0.311		361-408
				0.322		408-569
				0.323		569-841
24	T24HCS1-5HIA-5	VZP	I-127	0.395	X	0.08-1
				0.418	X	1-2
				0.384	X	2-7
				0.363	X	7-14
				0.346		14-28
				0.339		28-42
				0.341		42-49
				0.324		49-63
				0.302		63-94
				0.317		94-361
				0.312		361-408
				0.269		408-569
24	T24HCS1-5HIA-6	DIW	I-127	0.350	X	0.08-1
				0.404	X	1-2
				0.380	X	2-7
				0.365	X	7-14
				0.383	X	14-28
				0.367	X	28-42
				0.313		42-49
				0.395	X	49-63
				0.397	X	63-94
				0.344		94-361
				0.400	X	361-408
				0.388	X	408-569
				0.373	X	569-841
24	T24HCS1-5HIA-4	VZP	NO_3	0.394	X	0.08-1
				0.431	X	1-2
				0.376	X	2-7
				0.382	X	7-14
				0.371	X	14-28
				0.342		28-42
				0.315		42-49
				0.313		49-63
				0.303		63-94
				0.327		94-361
				0.331		361-408
				0.183		408-569
				0.318		569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
24	T24HCS1-5HIA-5	VZP	NO_3	0.494	X	0.08-1
				0.327		1-2
				0.314		2-7
				0.311		7-14
				0.326		14-28
				0.301		28-42
				0.284		42-49
				0.289		49-63
				0.276		63-94
				0.314		94-361
				0.297		361-408
				0.142		408-569
24	T24HCS1-5HIA-6	DIW	NO_3	0.349		0.08-1
				0.398	X	1-2
				0.371	X	2-7
				0.359	X	7-14
				0.383	X	14-28
				0.371	X	28-42
				0.314		42-49
				0.353	X	49-63
				0.381	X	63-94
				0.372	X	94-361
				0.367	X	361-408
				0.336		408-569
				0.343		569-841
24	T24HCS1-5HIA-4	VZP	Tc-99	0.484	X	0.08-1
				1.002		1-2
				0.809		2-7
				0.830		7-14
				0.582	X	14-28
				0.533	X	28-42
				0.747		42-49
				0.729		49-63
				0.557	X	63-94
				0.512	X	94-361
				0.612	X	361-408
				0.405	X	408-569
				0.487	X	569-841
24	T24HCS1-5HIA-5	VZP	Tc-99	0.436	X	0.08-1
				0.688		1-2
				0.566	X	2-7
				0.560	X	7-14
				0.440	X	14-28
				0.476	X	28-42
				0.603	X	42-49
				0.577	X	49-63
				0.434	X	63-94
				0.469	X	94-361
				0.347		361-408
				0.242		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.2 (cont). Archive Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
24	T24HCS1-5HIA-6	DIW	Tc-99	0.583	X	0.08-1
				1.098		1-2
				0.902		2-7
				0.785		7-14
				0.839		14-28
				0.735		28-42
				0.671		42-49
				0.702		49-63
				0.810		63-94
				0.728		94-361
				0.778		361-408
				0.750		408-569
				0.856		569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.3. Tc-Gluconate Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
Tc-Gluconate	T4-VZ-1	VZP	Cr	1.017		0.08-1
				0.152		1-2
				0.124		2-7
				0.035		7-14
				0.020		14-28
				0.269		28-42
				0.638	X	42-49
				0.356	X	49-63
				0.206		63-94
				0.073		94-361
				0.021		361-408
				0.207		408-569
				0.038		569-841
Tc-Gluconate	T4-VZ-2	VZP	Cr	0.723		0.08-1
				0.112		1-2
				0.078		2-7
				0.049		7-14
				0.028		14-28
				0.184		28-42
				0.450	X	42-49
				0.258		49-63
				0.152		63-94
				0.046		94-361
				0.002		361-408
				0.163		408-569
Tc-Gluconate	T4-DI-3	DIW	Cr	1.195		0.08-1
				0.366	X	1-2
				0.402	X	2-7
				0.426	X	7-14
				0.360	X	14-28
				0.385	X	28-42
				0.361	X	42-49
				0.303		49-63
				0.295		63-94
				0.227		94-361
				0.812		361-408
				0.232		408-569
				0.207		569-841
Tc-Gluconate	T4-DI-4	DIW	Cr	0.956		0.08-1
				0.383	X	1-2
				0.397	X	2-7
				0.419	X	7-14
				0.354	X	14-28
				0.347		28-42
				0.320		42-49
				0.281		49-63
				0.266		63-94
				0.204		94-361
				0.670		361-408
				0.229		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.3 (cont). Tc-Gluconate Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
Tc-Gluconate	T5-VZ-1	VZP	Cr	0.268		0.08-1
				0.106		1-2
				0.051		2-7
				0.016		7-14
				0.011		14-28
				0.063		28-42
				0.162		42-49
				0.097		49-63
				0.060		63-94
				0.036		94-361
				0.033		361-408
				0.066		408-569
				0.008		569-841
Tc-Gluconate	T5-DI-3	DIW	Cr	0.951		0.08-1
				0.531	X	1-2
				0.501	X	2-7
				0.479	X	7-14
				0.361	X	14-28
				0.403	X	28-42
				0.375	X	42-49
				0.290		49-63
				0.249		63-94
				0.195		94-361
				0.686		361-408
				0.253		408-569
				0.246		569-841
Tc-Gluconate	T4-VZ-1	VZP	NO_3	0.711		0.08-1
				0.454	X	1-2
				0.451	X	2-7
				0.341		7-14
				0.323		14-28
				0.341		28-42
				0.333		42-49
				0.351	X	49-63
				0.383	X	63-94
				0.411	X	94-361
				0.343		361-408
				0.256		408-569
				0.261		569-841
Tc-Gluconate	T4-VZ-2	VZP	NO_3	0.656		0.08-1
				0.450	X	1-2
				0.446	X	2-7
				0.372	X	7-14
				0.312		14-28
				0.333		28-42
				0.346		42-49
				0.334		49-63
				0.372	X	63-94
				0.389	X	94-361
				0.342		361-408
				0.255		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.3 (cont). Tc-Gluconate Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
Tc-Gluconate	T4-DI-3	DIW	NO_3	0.946		0.08-1
				0.522	X	1-2
				0.492	X	2-7
				0.408	X	7-14
				0.364	X	14-28
				0.383	X	28-42
				0.375	X	42-49
				0.382	X	49-63
				0.427	X	63-94
				0.431	X	94-361
				0.437	X	361-408
				0.395	X	408-569
				0.349		569-841
Tc-Gluconate	T4-DI-4	DIW	NO_3	0.730		0.08-1
				0.522	X	1-2
				0.465	X	2-7
				0.399	X	7-14
				0.352	X	14-28
				0.378	X	28-42
				0.374	X	42-49
				0.394	X	49-63
				0.401	X	63-94
				0.408	X	94-361
				0.388	X	361-408
				0.353	X	408-569
Tc-Gluconate	T5-VZ-1	VZP	NO_3	0.394	X	0.08-1
				0.289		1-2
				0.371	X	2-7
				0.312		7-14
				0.275		14-28
				0.284		28-42
				0.298		42-49
				0.290		49-63
				0.304		63-94
				0.375	X	94-361
				0.350	X	361-408
				0.255		408-569
				0.294		569-841
Tc-Gluconate	T5-DI-3	DIW	NO_3	0.817		0.08-1
				0.558	X	1-2
				0.445	X	2-7
				0.364	X	7-14
				0.354	X	14-28
				0.373	X	28-42
				0.367	X	42-49
				0.366	X	49-63
				0.374	X	63-94
				0.460	X	94-361
				0.495	X	361-408
				0.438	X	408-569
				0.372		569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.3 (cont). Tc-Gluconate Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
Tc-Gluconate	T4-VZ-1	VZP	Tc-99	0.643	X	0.08-1
				0.265		1-2
				0.248		2-7
				0.228		7-14
				0.322		14-28
				0.455	X	28-42
				0.527	X	42-49
				0.474	X	49-63
				0.511	X	63-94
				0.474	X	94-361
				0.398	X	361-408
				0.319		408-569
				0.290		569-841
Tc-Gluconate	T4-VZ-2	VZP	Tc-99	0.535	X	0.08-1
				0.233		1-2
				0.226		2-7
				0.241		7-14
				0.330		14-28
				0.462	X	28-42
				0.550	X	42-49
				0.435	X	49-63
				0.495	X	63-94
				0.443	X	94-361
				0.333		361-408
				0.265		408-569
Tc-Gluconate	T4-DI-3	DIW	Tc-99	0.873		0.08-1
				0.437	X	1-2
				0.503	X	2-7
				0.541	X	7-14
				0.602	X	14-28
				0.621	X	28-42
				0.579	X	42-49
				0.552	X	49-63
				0.540	X	63-94
				0.442	X	94-361
				0.438	X	361-408
				0.304		408-569
				0.316		569-841
Tc-Gluconate	T4-DI-4	DIW	Tc-99	0.696		0.08-1
				0.453	X	1-2
				0.509	X	2-7
				0.575	X	7-14
				0.637	X	14-28
				0.606	X	28-42
				0.533	X	42-49
				0.508	X	49-63
				0.473	X	63-94
				0.403	X	94-361
				0.392	X	361-408
				0.302		408-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release
Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.3 (cont). Tc-Gluconate Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
Tc-Gluconate	T5-VZ-1	VZP	Tc-99	0.323		0.08-1
				0.209		1-2
				0.236		2-7
				0.225		7-14
				0.280		14-28
				0.335		28-42
				0.352	X	42-49
				0.297		49-63
				0.320		63-94
				0.468	X	94-361
				0.337		361-408
				0.283		408-569
				0.322		569-841
Tc-Gluconate	T5-DI-3	DIW	Tc-99	1.258		0.08-1
				0.946		1-2
				0.721		2-7
				0.609	X	7-14
				0.582	X	14-28
				0.574	X	28-42
				0.537	X	42-49
				0.473	X	49-63
				0.402	X	63-94
				0.379	X	94-361
				0.516	X	361-408
				0.460	X	408-569
				0.425		569-841

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.4. Iodide Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
Iodide	T1-VZ-2	VZP	NO_3	0.367	X	0.08-1
				0.378	X	1-2
				0.375	X	2-7
				0.294		7-14
				0.289		14-28
				0.307		28-42
				0.301		42-49
				0.284		49-63
				0.296		63-100
				0.358	X	100-369
				0.330		369-414
				0.188		414-569
Iodide	T2-VZ-2	VZP	NO_3	0.370	X	0.08-1
				0.358	X	1-2
				0.380	X	2-7
				0.302		7-14
				0.273		14-28
				0.302		28-42
				0.321		42-49
				0.296		49-63
				0.418	X	63-100
				0.372	X	100-369
				0.320		369-414
				0.231		414-569
Iodide	T3-VZ-1	VZP	NO_3	0.340		0.08-1
				0.543	X	1-2
				0.378	X	2-7
				0.306		7-14
				0.277		14-28
				0.300		28-42
				0.301		42-49
				0.293		49-63
				0.321		63-100
				0.378	X	100-369
				0.332		369-414
				0.230		414-569
Iodide	T1-DI-3	DIW	NO_3	0.336		0.08-1
				0.327		1-2
				0.381	X	2-7
				0.322		7-14
				0.333		14-28
				0.336		28-42
				0.341		42-49
				0.351	X	49-63
				0.377	X	63-100
				0.409	X	100-369
				0.409	X	369-414
				0.322		414-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.4 (cont). Iodide Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	Monolith	Eluent	Constituent of Concern (COC)	Interval Slope	X = Slope is 0.5 ± 0.15	Leach Test Interval (days)
Iodide	T2-DI-3	DIW	NO_3	0.359	X	0.08-1
				0.270		1-2
				0.340		2-7
				0.303		7-14
				0.277		14-28
				0.322		28-42
				0.338		42-49
				0.347		49-63
				0.352	X	63-100
				0.411	X	100-369
				0.424	X	369-414
				0.375	X	414-569
Iodide	T3-DI-3	DIW	NO_3	0.380	X	0.08-1
				0.285		1-2
				0.344		2-7
				0.237		7-14
				0.286		14-28
				0.336		28-42
				0.343		42-49
				0.300		49-63
				0.355	X	63-100
				0.406	X	100-369
				0.416	X	369-414
				0.365	X	414-569
Iodide	T1-VZ-2	VZP	I-127	0.307		0.08-1
				0.354	X	1-2
				0.349		2-7
				0.278		7-14
				0.269		14-28
				0.316		28-42
				0.404	X	42-49
				0.322		49-63
				0.359	X	63-100
				0.374	X	100-369
				0.481	X	369-414
				1.176		414-569
Iodide	T2-VZ-2	VZP	I-127	0.338		0.08-1
				0.348		1-2
				0.355	X	2-7
				0.280		7-14
				0.254		14-28
				0.288		28-42
				0.312		42-49
				0.304		49-63
				0.329		63-100
				0.407	X	100-369
				0.335		369-414
				0.399	X	414-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

Table C.4 (cont). Iodide Suite Method 1315 D_{obs} Equation Criteria Checks for All Monoliths

Test#	**Monolith**	**Eluent**	**Constituent of Concern (COC)**	**Interval Slope**	**X = Slope is 0.5 ± 0.15**	**Leach Test Interval (days)**
Iodide	T3-VZ-1	VZP	I-127	0.300		0.08-1
				0.367	X	1-2
				0.366	X	2-7
				0.322		7-14
				0.261		14-28
				0.306		28-42
				0.314		42-49
				0.309		49-63
				0.334		63-100
				0.383	X	100-369
				0.378	X	369-414
				0.309		414-569
Iodide	T1-DI-3	DIW	I-127	0.316		0.08-1
				0.321		1-2
				0.373	X	2-7
				0.307		7-14
				0.307		14-28
				0.336		28-42
				0.338		42-49
				0.341		49-63
				0.414	X	63-100
				0.412	X	100-369
				0.474	X	369-414
				0.450	X	414-569
Iodide	T2-DI-3	DIW	I-127	0.342		0.08-1
				0.274		1-2
				0.345		2-7
				0.275		7-14
				0.271		14-28
				0.318		28-42
				0.341		42-49
				0.348		49-63
				0.357	X	63-100
				0.416	X	100-369
				0.474	X	369-414
				0.392	X	414-569
Iodide	T3-DI-3	DIW	I-127	0.359	X	0.08-1
				0.287		1-2
				0.340		2-7
				0.224		7-14
				0.280		14-28
				0.328		28-42
				0.338		42-49
				0.294		49-63
				0.353	X	63-100
				0.395	X	100-369
				0.441	X	369-414
				0.402	X	414-569

Eluents: DIW = De-ionized Water VZP = Vadose Zone Porewater Red = >20% Release

Slopes are for each interval of the logarithm of cumulative release versus the logarithm of cumulative time plots.

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