PNNL-24297
RPT-SLAW-001, Rev. 0

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

Extended Leach Testing of Simulated LAW Cast Stone Monoliths

July 2015

RJ Serne
JH Westsik, Jr.
BD Williams
HB Jung
G Wang

U.S. DEPARTMENT OF ENERGY

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

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC05-76RL01830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information,
P.O. Box 62, Oak Ridge, TN 37831-0062;
ph: (865) 576-8401
fax: (865) 576-5728
email: reports@adonis.osti.gov

Available to the public from the National Technical Information Service
5301 Shawnee Rd., Alexandria, VA 22312
ph: (800) 553-NTIS (6847)
email: orders@ntis.gov <http://www.ntis.gov/about/form.aspx>
Online ordering: http://www.ntis.gov

This document was printed on recycled paper.

(8/2010)

PNNL-24297
RPT-SLAW-001, Rev. 0

Extended Leach Testing of Simulated LAW Cast Stone Monoliths

RJ Serne
JH Westsik, Jr.
BD Williams
HB Jung[1]
G Wang

July 2015

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

Pacific Northwest National Laboratory
Richland, Washington 99352

---

[1] Currently located at Department of Geoscience and Geography, New Jersey City University, 2039 Kennedy Boulevard, Jersey City, NJ 07305.

Executive Summary

This report describes the results from long-term laboratory leach tests 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 for selected ongoing EPA-1315 tests on monoliths made with LAW simulants beyond the conventional 63-day time period up to 609 days reported herein (with some tests continuing that will be documented later) in an effort to evaluate long-term leaching properties of Cast Stone to support future performance assessment activities.
2. Starting new EPA-1315 leach tests on archived Cast Stone monoliths made with four LAW simulants using two leachants (deionized water [DIW] and simulated Hanford Integrated Disposal Facility (IDF) Site vadose zone pore water [VZP]).
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 leachants.
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.

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 effective diffusivities and almost an order of magnitude range in effective 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.

Thus monolith leach studies continue with current results documented herein for the same key constituents. Conclusions derived after leaching four different suites of Cast Stone, one suite to address each of the four objectives follow. No leaching data for uranium is reported in the main text because almost all the leachates, regardless of leachant type used (VZP or DIW), had non-detectable uranium concentrations. That is, uranium is sequestered strongly in Cast Stone and leaches so slowly that leachate concentrations are not detectable. 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.

Up through 20 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. However, the most recent DIW leach results for a few monoliths are showing signs of an increase rate of release that might be caused by the formation of new surface micro-cracks visually observed after 790 days of leaching. The cracks are not covered by white precipitate suggesting that they are newly formed. We wonder if this observation is the first signs of some internal degradation of Cast Stone monoliths after a time period between 550 and 790 days of leaching. That is, these "clean" micro-cracks were not observed after 427 days of leaching but were observed and photographed after 790 days of leaching. For monoliths leached in VZP no "late stage" increases in leach rates have been observed, but because of the significant coating of secondary precipitates on the VZP-leached monoliths' surfaces it is not possible to evaluate whether there are new micro-cracks developing.

Tables of interval averaged effective diffusion coefficients (D_e values)---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 leachant is used. The same range of interval averaged D_e 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 3. Chromium interval averaged D_e values for VZP leachants are generally not quantifiable because leachate 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 potential risk to groundwater 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 Na, Cast Stone Mixes 13, 14, and 17 leach less than other mixes and Mixes 3 and 21 leach the most Na. 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 leachates from the various mixes had no detectable Cr such that choosing the best and worst performing Cast Stone mixes is not useful, all mixes retain Cr well. The same is true for uranium.

Regardless of the Cast Stone mix composition, the D_e values for the anions iodide, nitrate and nitrite and the cation sodium are very similar. The range in averaged interval D_e values for these four constituents varies by only a factor of three to six. Cr leaches the slowest especially when VZP leachant 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 leachant is used generally between 49 to 63-d of leaching, we are recommending that the VZP leached averaged 28- to 63-d interval D_e 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 the data from using VZP leachant because it better represents water that will percolate through the IDF facility and when the cumulative mass of a constituent released from the monoliths starts to exceed 20% of their starting inventories, the mass used to calculate the effective diffusion coefficient is not accurate. The accuracy issue arises because the leaching conditions no longer satisfy the assumed boundary conditions; namely semi-infinite source requirement. Further, the early leach periods (0.08 d up to 14 d) are 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_e 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) with some brucite ($Mg(OH)_2$), and perhaps some calcite precipitating. The same minerals are 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 D_e 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 leachate 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 leachates. The earlier literature, including Lockrem et al. (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 reoxidation 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 and other pertechnetate-bearing monoliths from the Archive and Screening Leach-Extended Suite in both VZP and DIW leachants. Whether the Tc in leachates 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 leachates 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 reoxidation process is slow such that the effective 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 3.4.

This report also discusses recommended follow-on leach testing of some of the monoliths that have been leaching for over two 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.

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), and Kevin Fox, Christine Langton, Roger Seitz, Greg Flach (SRNL), and John Mahoney and Felix Miera (YAHGS LLC), and Bob Andrews and Mick Apted (INTERA), and Wooyong Um and Bill Dey (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 leachates 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, 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.

Finally the authors thank EB Baer, WC Dey, KJ Geiszler, DC Lanigan, and MM Valenta-Snyder for performing all the analytical data and calculation QA reviews.

The authors wish to thank Heather Culley and Mary Stover for editorial review and document production.

Acronyms and Abbreviations

ASTM	ASTM International (formerly the American Society for Testing and Materials)
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	contaminants of concern
DIW	deionized water
DOE	U.S. Department of Energy
DOE-ORP	U.S. Department of Energy Office of River Protection
DST	double-shell tanks
EM	DOE Office of Environmental Management
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
LAW	low-activity waste
LI	Leach Index; see Equation 2 in main text for definition
L/S	liquid-to-solid ratio
NMR	nuclear magnetic resonance spectroscopy
OPC	ordinary Portland cement
PA	performance assessment
PNNL	Pacific Northwest National Laboratory
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
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
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 leachant and leachate. Leachant is the starting solution used to interact with the waste form monoliths. We use two leachants 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 leachates. Leachates contain the species that diffuse or dissolve out of the waste forms. The difference between the concentrations of any constituent in the leachate from a container that had a monolith present and the concentration in the leachate from the blank container (contains only the monolith holder) represents the net release from the monolith.

Contents

Figures

Tables

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. The grout that has been selected for solidification of radioactive wastes, including WTP aqueous secondary wastes treated at the ETF at Hanford is similar to Cast Stone. 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.

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. 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 has been underway (albeit with variable funding) since 2012, with several simulated Hanford waste streams to be solidified and tested. One outcome is that a grout 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. In addition, a grout, whose dry blend consists of a mixture of lime, BFS, and fly ash, is currently being developed for solidification of other aqueous wastes currently 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).

[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.

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 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 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.

This report extends 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 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.

Specific objectives of the Cast Stone tests described in this report 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 leachants (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 leachants, 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 leachants. 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 leachates, 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 leachant. DIW is generally considered an aggressive leachant 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 leachate, simulated Hanford formation VZP was used in many of the leach tests described in this progress report.

Each of these objectives and the results are discussed in more detail in Section 3.0. Section 2.0 describes the LAW simulant and VZP recipes, Cast Stone preparation details, and leach test methodology. Section 4.0 summarizes the key findings and describes some future studies that would be useful to gain more understanding on Cast Stone long-term performance. Section 5.0 lists the references cited in the report.

1.1 Quality Assurance

The multi-year work described in 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. As mentioned above, four main objectives were conceived and four distinct suites of leach tests are ongoing.

The extended leach tests beyond the conventional 63-day time period are composed of four sets of leach tests that we are labeling as suites of tests:

- Screening Leach Tests –Extended Suite that used Cast Stone monoliths from the original Screening Test activities which leached only in DIW.
- Archive Leach Test Suite that used Cast Stone monoliths from the original Screening Test activities that had been stored at room temperature under 100% relative humidity conditions.
- I-Loading Suite that used newly made Cast Stone monoliths prepared with a new batch of 7.8 M Na Average simulant spiked with varying initial iodide concentrations. These newly made monoliths used the same mix formulation as the original Screening Test replicate Mixes #3 and #22, except no Tc or uranium was spiked into the simulant.
- Tc-Gluconate Spiked Suite that used newly made Cast Stone monoliths using newly prepared 7.8 M Na Average simulant spiked with either the Tc(I)-gluconate or pertechnetate and the traditional Cast Stone dry blend at a water-to-dry blend ratio of 0.6. These newly made monoliths used the same mix formulation as the original Screening Test replicate Mixes #3 and #22, except no iodide or uranium was added to the newly prepared simulant. Mixes #3 and #22 used fly ash from a NW source and BFS from a SE source. Details are found in Westsik et al. (2013a).

1.1.1 Screening Leach Tests –Extended Suite

There are 11 monoliths being leached in DIW in this set of leach tests. The monoliths were prepared as part of the Supplemental Immobilization of Hanford Low-Activity Waste project funded by WRPS (PNNL project 62745, WRPS contract releases 36437-122 and 36437-134). Leach testing of the 11 monoliths through the first 91 days of testing was conducted under project 62745. This work was done under the PNNL Quality Assurance (QA) Plan *WRPS Waste Form Testing Program Quality Assurance Plan* (QA-WWFTP-001). Preparation of the monoliths and the initial leach testing was conducted under test plan TP-62745-001. Leach testing of the monoliths began in January 2013. Along with the 11 monoliths, two containers, called blanks, with no Cast Stone (containing only the monolith holder and DIW) are processed at each sampling time to be certain that the containers and monolith holders to do not leach constituents into solution.

The leach testing continued beyond 91 days to a total of 252-257 days with funding from the DOE EM-31 Support Program (EMSP) “Production and Long-term Performance of Low Temperature Waste Forms”. The work was conducted under the EMSP Project Test Plan TP-EMSP-0011 and under the Environmental Management Support Program Quality Assurance Plan (QA-EMSP-001). Leach testing continued to September 2013.

Beyond September 2013, the leach testing continued again with funding from WRPS as part of the Supplemental Immobilization of Hanford Low-Activity Waste (LAW) project (PNNL project 66596, WRPS contract release 36437-166). This work was done under the PNNL QA Plan *WRPS Waste Form Testing Program Quality Assurance Plan* (QA-WWFTP-001).

In February 2015, the 11 monoliths (leaching in DIW) were sampled and 6 of the monoliths were archived in their respective leachates for future solid-phase characterization. Five of the monoliths are being continued in long-term leach tests until final decisions on their fate are made (sometime around July 2015).

1.1.2 Archive Leach Test Suite

For the Archive Leach Test Suite, there are 35 monoliths and 5 blanks (containing only the monolith holder) being leached either in DIW or simulated VZP. The monoliths were prepared as part of the Supplemental Immobilization of Hanford Low-Activity Waste project funded by WRPS (PNNL project 62745, WRPS contract releases 36437-122 and 36437-134). Preparation of the monoliths is described in test plan TP-62745-001. The leach testing itself was conducted with funding from the DOE EMSP “Production and Long-term Performance of Low Temperature Waste Forms”. The work was conducted under the EMSP Project Test Plan TP-EMSP-0011 and under the Environmental Management Support Program Quality Assurance Plan (QA-EMSP-001). Leach testing was started in July 2013 and continued to September 2013.

Beyond September 2013, the leach testing of the Archived Suite of monoliths continued with funding from WRPS as part of the Supplemental Immobilization of Hanford LAW project (PNNL project 66596, WRPS contract release 36437-166). This work was done under the PNNL QA Plan *WRPS Waste Form Testing Program Quality Assurance Plan* (QA-WWFTP-001). In late February 2015, all 35 monolith leachates and the 5 blanks were sampled. Of the 35 monoliths, 13 continue to be leached in their respective leachants in long-term leach tests until final decisions on their fate will be made (sometime around July 2015). Of the “Archive” monoliths leach tests, 17 were stopped, and the monoliths are currently stored in their respective leachates for future solid-phase characterization. Five of the “Archive” monoliths were discarded as waste, since they are duplicates of the 17 monoliths being stored.

1.1.3 I-Loading Suite

In the I-Loading Suite, there are 12 monoliths being leached in DIW or VZP and 2 blanks (containing only the monolith holder), one for each leachant. The waste simulant used to prepare the I-spiked Cast Stone monoliths contained the short-lived radiotracer ^{125}I, which required special radiation and safety precautions, and stable ^{127}I. Unfortunately, soon after commencing the leach tests, the auto-gamma counter used to measure the ^{125}I became inoperative. Because of the age and overall condition of the instrument, it was economically unfeasible to repair or replace the instrument. Thus, the leach tests had to rely on measuring only the stable ^{127}I, which fortunately was successfully measured at very low concentrations by inductively coupled plasma mass spectrometry (ICP-MS) such that defensible data were produced.

Preparation of the I-spiked monoliths and the leach testing was initially funded by the DOE EMSP "Production and Long-term Performance of Low Temperature Waste Forms". The work was conducted under the EMSP Project Test Plan TP-EMSP-0011 and under the Environmental Management Support Program Quality Assurance Plan (QA-EMSP-001). Leach testing was started in July 2013 and continued to September 2013.

Beyond September 2013, the leach testing continued with funding from WRPS as part of the Supplemental Immobilization of Hanford LAW project (PNNL project 66596, WRPS contract release 36437-166). This work was done under the PNNL QA Plan *WRPS Waste Form Testing Program Quality Assurance Plan* (QA-WWFTP-001).

In late February 2015, the 12 iodide monolith leachates and the 2 blanks were sampled. All 12 of the iodide monoliths were disposed of after the February 2015 sampling. The Iodide Suite of monoliths used the same waste simulant/dry blend mix and free-water-to-dry blend ratio as the Tc-Gluconate suite of monoliths. The ^{125}I spiked monoliths and all their leachates have required special handling and segregation of all waste based on ^{125}I being categorized as a "hard to detect" nuclide and the expenses of continuing the segregation and special handling was deemed to be excessive compared to the value of the data being generated; thus the ^{125}I suite of tests has been terminated.

1.1.4 Tc-Gluconate Spiked Suite

In the Tc-Gluconate Spiked Suite, there are eight monoliths and two blanks (containing only the monolith holder) being leached in DIW or a simulated VZP. Preparation of the monoliths and the leach testing were funded by the DOE EMSP "Production and Long-term Performance of Low Temperature Waste Forms." The work was conducted under the EMSP Project Test Plan TP-EMSP-0011 and under the Environmental Management Support Program Quality Assurance Plan (QA-EMSP-001). Leach testing was started in July 2013 and continued to September 2013.

Beyond September 2013, the leach testing continued with funding from WRPS as part of the Supplemental Immobilization of Hanford LAW project (PNNL project 66596, WRPS contract release 36437-166). This work was done under the PNNL QA Plan *WRPS Waste Form Testing Program Quality Assurance Plan* (QA-WWFTP-001).

In late February 2015 the eight monolith leachates and the two blanks were sampled. Four of the eight monoliths continue to be leached in their respective leachants in long-term leach tests until final decisions on their fate will be made (sometime around July 2015). The other four "Tc-Gluconate" monoliths leach tests were stopped and the monoliths are currently stored in their respective leachates for future solid-phase characterization.

1.1.5 Final Report and Records

This work was conducted with funding from WRPS under contract 36437-166, Supplemental Immobilization of Hanford LAW. The work was conducted as part of PNNL Project 66596.

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* (NQA-1-2000) to R&D activities, and the NQA-1a-2009 addendum (NQA-1a-2009 addendum). To ensure that all client QA expectations were addressed, the QA controls of the WRPS Waste Form Testing Program (WWFTP) QA program were also implemented for this work. The WWFTP QA program consists of the WWFTP Quality Assurance Plan (QA-WWFTP-001)[1] and associated QA-NSLW-numbered procedures that provide detailed instructions for implementing NQA-1 requirements for R&D work. The WWFTP QA program implements the requirements of ASME 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."

The work described in this report was assigned the technology level "Applied Research," and was planned, performed, documented, and reported in accordance with Procedure QA-NSLW-1102, *Scientific Investigation for Applied Research*. All staff members contributing to the work received proper technical and QA training prior to performing quality-affecting work.

Records of all of the experimental work will be filed with the records for the Supplemental Immobilization of Hanford LAW project (PNNL project 66596, WRPS contract release 36437-166).

[1] MacPherson DB. 2013. WRPS Waste Form Testing Program Quality Assurance Plan, Pacific Northwest National Laboratory.

2.0 LAW Waste Simulants and Cast Stone Monolith Preparation

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) standards.

[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 Metals	HTWOS Overall Avg. moles/mole Na	HTWOS Maximum moles/mole Na	Other Considerations 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 chromium nitrate 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^{3}in 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)	xxx	**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**

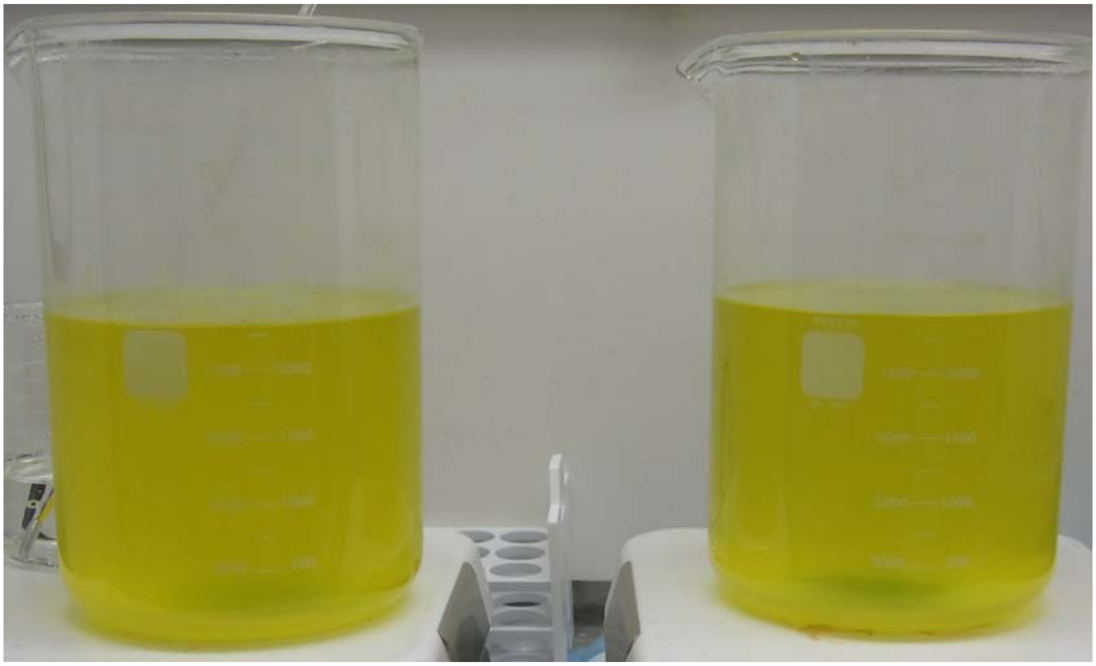


Figure 2.1. 7.8 M Na Average Simulant

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,

[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.

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.

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 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 in 100% 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 100% relative humidity. 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.

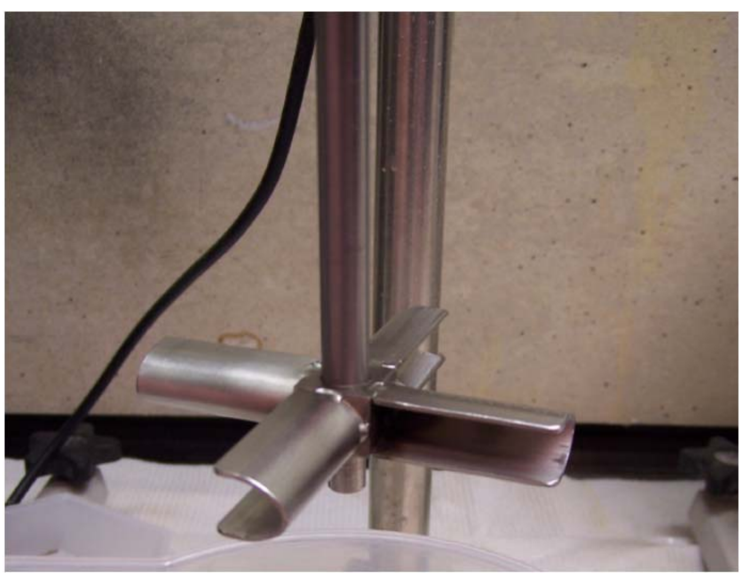


Figure 2.2. Mixer Blade on End of Mixer Shaft (Direction of Rotation is Clockwise)

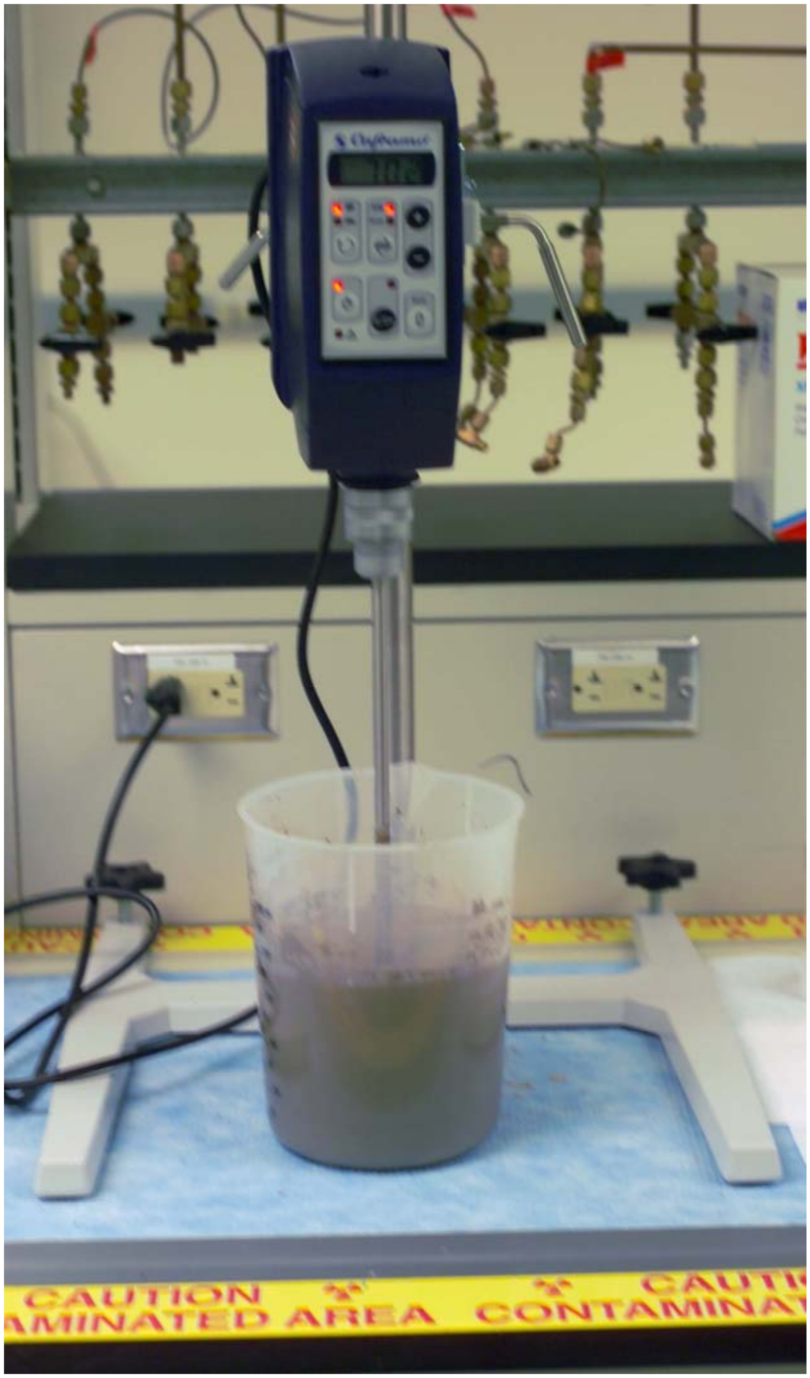


Figure 2.3. Mixer Pump & Assembly Immersed in Mixing Container

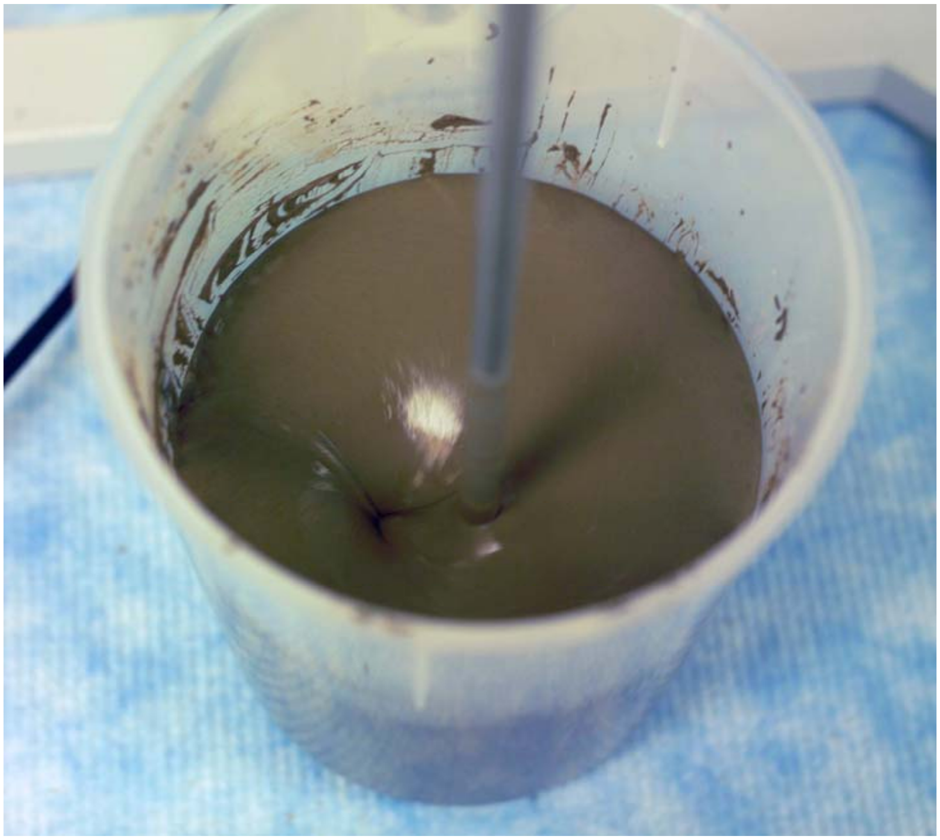


Figure 2.4. Close Up of Well Mixed Cast Stone Slurry Ready for Molding

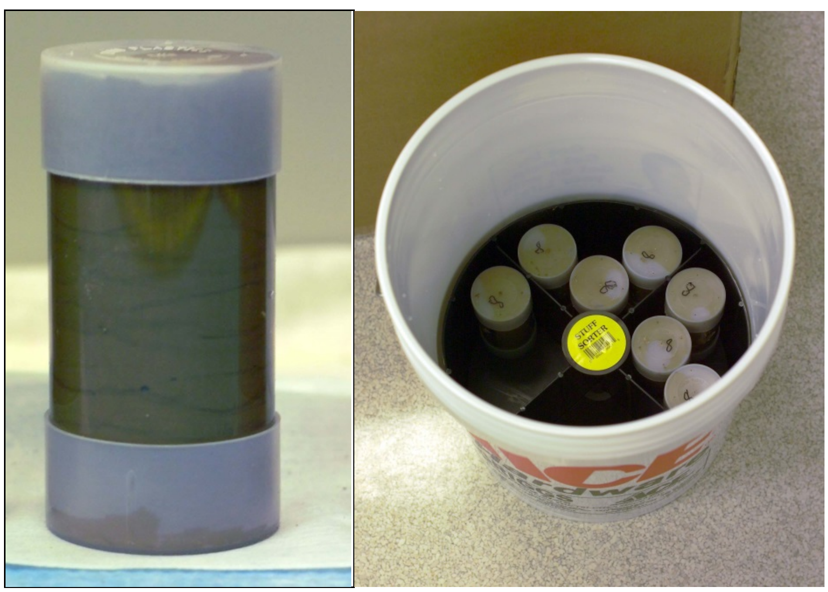


Figure 2.5. Curing Molds Filled with Cast Stone Slurry & Placement in Curing Containers

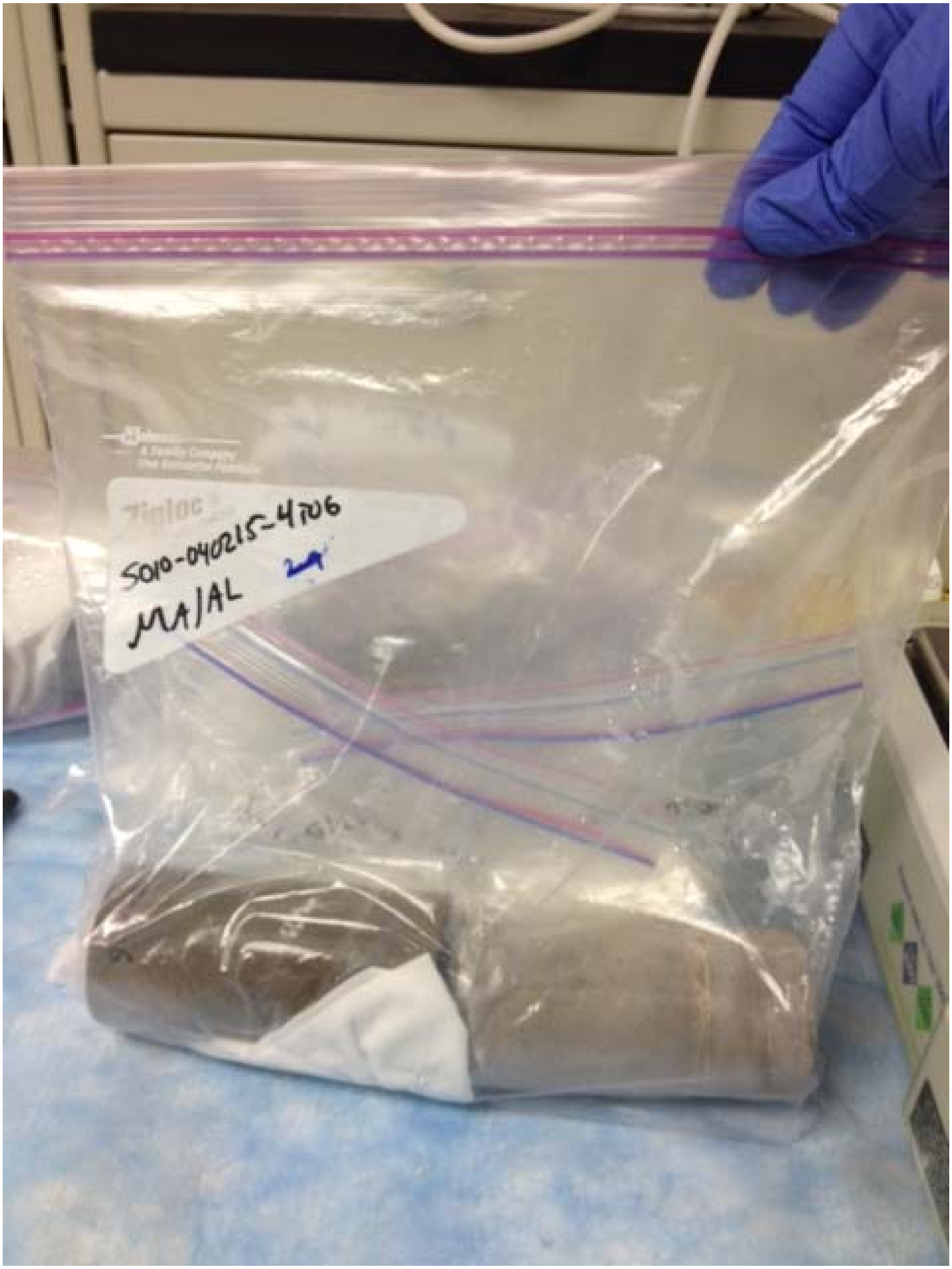


Figure 2.6. Storage Method-Monoliths in Double Lined Bags with Wet Paper Towel

2.3 Leach Test Details

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 2012)[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 leachants were used to leach the Cast Stone monoliths. As customary, DIW was one of the leachants, and a simulated Hanford formation sediment VZP was the second leachant. Second, the time intervals prescribed in the EPA-1315 method were extended, albeit with some large intervals, beyond 63 days up to 609 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 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 2.5 (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 100% relative humidity conditions). The

[1]The shape of the monoliths was right cylinders with dimensions of 2-in diameter by 4-in length. EPA-1315 leach method states 'The geometry of monolithic samples may be rectangular (e.g., bricks or tiles), cubes, wafers or cylinders. A minimum sample size of 5 cm in the direction of mass transfer must be employed and the liquid (leachant)-to- monolith 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_e 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 1315 method became a formally accepted EPA method in January 2015 with no significant changes in methodology between the draft and final version. The final version is available at http://www.epa.gov/epawaste/hazard/testmethods/sw846/new_meth.htm#1315.

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 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 moisture content value (0.254) for the newly prepared monoliths was calculated by difference from the measured solids content, 0.746.

Table 2.5. Monoliths from Screening Test that had Leach Testing Extended

Number	Mix #	Monoliths 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 1315 leach testing for the original screening tests).

2.3.1 EPA Method 1315

The EPA Method 1315 (EPA 2015) is a semi-dynamic leach test that consists of submerging a monolithic sample in leachant 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, leachates were collected at longer times, listed in Table 2.6. At each sampling interval, all the leaching fluid was removed and replaced with fresh fluid. A schematic of this process is shown in Figure 2.7.

Table 2.6. List of Sampling Times (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						
35	187	35	182						
35	222	35	217						
35	257	35	252						
305	562	305	557						
47	609	47	604						

Note: most of these leach tests were continued through February 2015 when ~half were terminated. Results for the February 2015 sampling will be added to a revision to this report by the end of calendar year 2015.

At each of the leaching intervals, the monolith mass is recorded, and the leaching solution is 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 the EPA 1315 method 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.

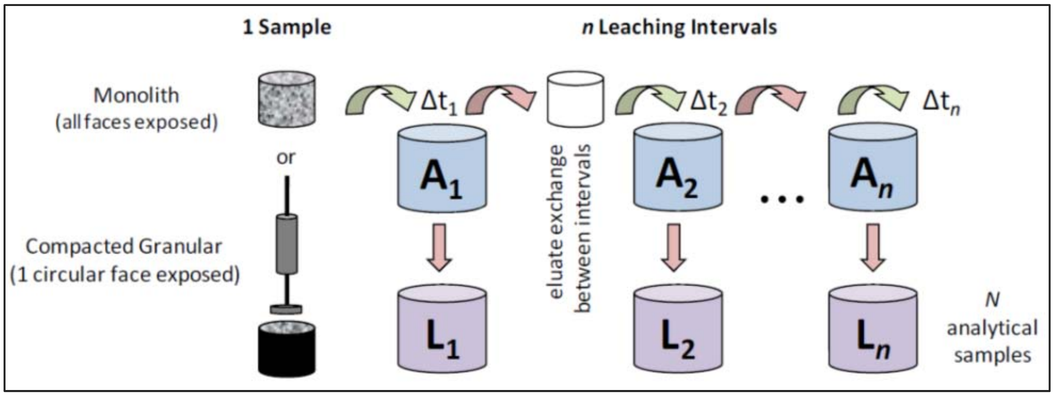


Figure 2.7. Schematic of the EPA-1315 Leach Method

The cylindrical monoliths were placed into the centers of leaching vessels containing sufficient leachant to maintain a solution-to-solid surface area ratio of 9 ± 1 mL of leachant per square centimeter of sample geometric surface area. Sample stands and holders (see Figure 2.8 and Figure 2.9) were used to maximize the contact area of the monolith sample with the leaching solution. Figure 2.10 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 leachate at each leaching interval. Then a larger aliquot of leachate was removed from the leach container and split into various analytical aliquots, some preserved as noted in Table 2.7, and submitted for chemical analyses using the analytical method designated.

Figure 2.8. Photo of Monoliths on Stands and Leach Containers Prior to Start of Leach Tests

Figure 2.9. Close Up of Monoliths on Stands Prior to Leaching

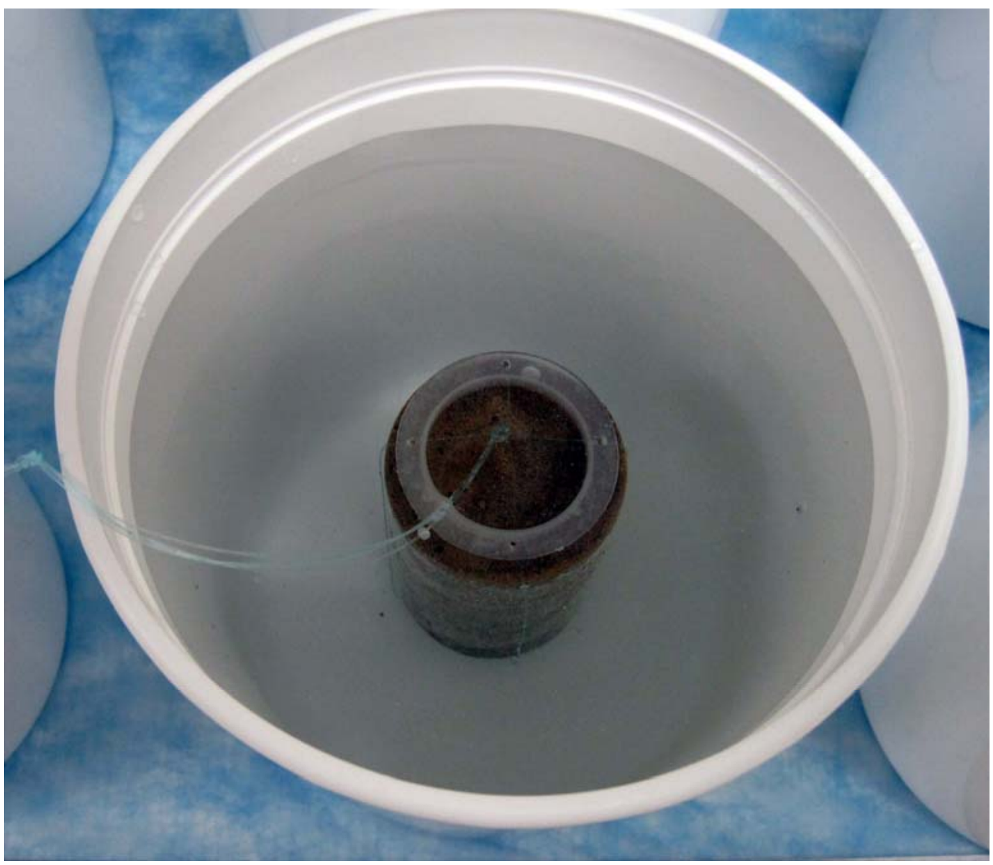


Figure 2.10. Monolith in Leach Container During Leach Testing

Table 2.7. Chemical Analyses of Leachates

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

2.4 Description of the Vadose Zone Pore Water Simulant

The VZP simulant recipe is shown in Table 2.8. The recipe is based on several direct measurements of actual VZP removed from Hanford formation sediments from boreholes in 200 E where the IDF is located. Several hundred grams of field moist sediment collected from boreholes 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 depths (48.5 and 82.5 ft below ground surface [bgs]) from borehole C4124 (299-E27-22) was averaged and then charge balanced (see Brown et al. 2008). 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.

Table 2.8. 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	$CaSO_4 \cdot 2H_2O$	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	$MgCl_2 \cdot 6H_2O$	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.2 compares the "theoretical compositions based on masses of chemical reagents added" with the actual measured composition of the VZP.

2.5 Effective Diffusion Coefficient Calculations

The observed effective diffusivity for each constituent of interest is calculated using the analytical solution for Fick's 2nd Law using Equation 1, for simple radial diffusion from a cylinder into an infinite bath as presented by Crank (1986) and also used in EPA Method 1315. For this report, the constituents of interest include Cr, I, Na, Tc, U, nitrate, and nitrite.

$$D_e = \pi \left[\frac{M_{t_i}}{2\rho C_o \left(\sqrt{t_i} - \sqrt{t_{i-1}}\right)} \right]^2 \quad \text{Equation 1}$$

where
- D_e = observed or effective 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 after leaching interval, *i* (s),
- t_{i-1} = cumulative contact time after leaching interval, *i-1* (s),
- C_o = initial concentration of constituent in the dry Cast Stone (mg/kg-dry), and
- ρ = Cast Stone dry bulk density ($kg\text{-}dry/m^3$).

Note that the most common units for effective diffusion coefficients when used in transport modeling activities is cm^2/s such that the value calculated in Equation 1 is multiplied by 10,000 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.

The leachability index (LI), a unitless parameter derived from the interval effective diffusion coefficient values, is calculated using Equation 2:

$$LI_i = -\log[De] \quad \text{Equation 2}$$

where LI_i = the LI for each leach interval (*i*) and D_e is the effective diffusivity for the element of interest (cm^2/s) during the leach interval *i*. In general, the LIs for each time interval are averaged for each COC to calculate an average LI.

The EPA Method 1315 is similar to ANSI/ANS16.1 (see 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 effective diffusion coefficient over time periods of 91 days for Cast Stone monoliths containing several secondary waste simulants.

In the ANS16.1 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). The fraction leached for all COCs in our tests as a function of cumulative leach time are shown in Appendix B.

ASTM-1308 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.

The next section describes the results of the leach testing on the LAW Cast Stone monoliths within this "extended" leach testing task.

2.6 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:

$$D_i = D_f \frac{\varepsilon\delta}{\tau^2} \qquad \text{Equation 3}$$

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 2.11 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 2.12. In practice, the constrictivity, tortuosity, and porosity are often used in models as purely empirical parameters to establish the effective diffusivities in porous media.

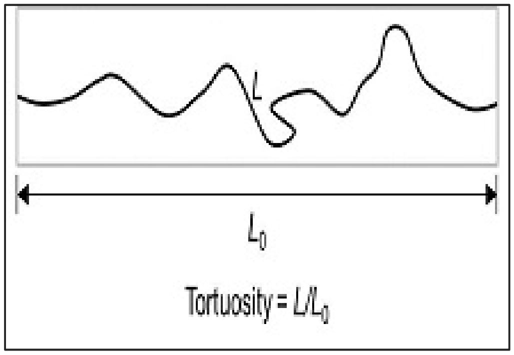


Figure 2.11. Conceptual Representation of Tortuosity (τ)

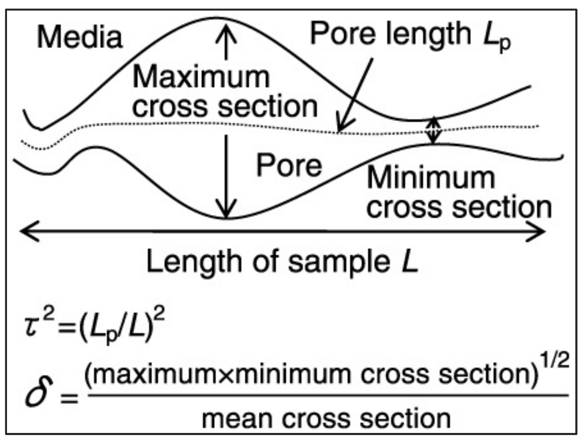


Figure 2.12. Conceptual Representation of Constrictivity (δ)[1]

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:

$$D_e = \frac{D_i}{\alpha} \qquad \text{Equation 4}$$

[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).

where the modified diffusion coefficient is called the effective diffusion coefficient, D_e[1], 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:

$$\alpha = \varepsilon + \rho K_d \qquad \text{Equation 5}$$

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 effective 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_e values for a companion species that is assumed to 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_e value for the non-sorbing constituent allows one to calculate the intrinsic (D_i) coefficient for the specific waste form using Equation 4 and the known porosity because Equation 5 reduces to the porosity (ε) when K_d is zero.

For Cast Stone, which contains the reductant BFS in its dry blend, we caution against assuming the measured D_e 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_e value for the mobile COC (often nitrate) is used in Equations 4 and 5 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_e 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 Equation 4. From the independently measured values for porosity and dry bulk density one can calculate the K_d for the constituent. 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

[1] The term effective diffusion coefficient used in this report is called observed diffusion coefficient in EPA Method 1315 documents. ANSI/ANS16.1 uses the symbol D with no subscript and names D =effective diffusion coefficient. ASTM C1308 method uses the symbol D_e and name effective diffusion coefficient. In some literature this parameter, the D_e value in Equations 1 and 4 is called apparent diffusion coefficient, D_a (see for example Grathwohl (1998). All three names are equivalent and are "quantified" in standard leach tests by Equations 2 and 4.

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.

3.0 Results of the Long-Term Leach Tests—Through FY 2014

The results of the extended leach testing on the Cast Stone monoliths are presented for the four suites of tests that were performed to gather data on the objectives delineated in Section 1.0. The order of presentation is Screening Leach Tests –Extended Suite, Archive Leach Test Suite, I-Loading Suite, and finally, the Tc-Gluconate Spiked Suite.

3.1 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 2.5 shows the eight 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 8 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_e value) monoliths, one of the worst performing (i.e., having nearly the highest leach rate or D_e value) monoliths, and one of the median performing (i.e., having a leach rate or D_e 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 any 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 3 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 3.1 shows the average of interval D_e values for the constituents of interest for the eight monoliths being leached in DIW up through either 604 or 609 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 eight 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_e values used the interval leaching data from day 7 through the last sampling interval, either 604 days or 609 days. The second average of interval D_e 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_e values provided in Table 3.1 uses the interval leach data from 28 days out through the last sampling (either 604 days or 609 days).

Table 3.1. Average of Interval D_e Values (cm^2/s) for Designated Sampling Periods

Monolith	De 7d to 604/609 d Avg.	σ	De 28 d to 63 d Avg.	σ	De 28 d to 604/609 Avg.	σ
	Tc(VII)					
T18LCS2-7.8RAS	1.65E-11	2.01E-11	5.69E-12	1.05E-12	1.73E-11	2.19E-11
T14LCS2-7.8HIS	9.51E-12	2.32E-12	9.60E-12	1.79E-12	8.92E-12	1.86E-12
T15HCS1-7.8HIS	5.59E-11	5.13E-11	4.62E-11	1.04E-11	3.37E-11	1.41E-11
T24HCS1-5HIA	6.91E-11	2.14E-11	6.52E-11	6.74E-12	7.08E-11	1.97E-11
T5HCS1-7.8AVG	4.54E-11	2.44E-11	7.16E-11	1.47E-11	4.43E-11	2.60E-11
T13LCS2-5AVG	7.22E-11	3.36E-11	8.27E-11	1.09E-11	7.92E-11	2.46E-11
T21LCS1-7.8HIS	2.19E-10	1.98E-10	1.13E-10	9.70E-11	2.51E-10	1.94E-10
T10HCS1-5HIS	1.40E-10	2.20E-11	1.20E-10	1.42E-11	1.10E-10	2.20E-11
T16HCS1-7.8RAS	1.47E-10	3.27E-11	1.57E-10	1.65E-11	1.43E-10	3.41E-11
T17LCS2-5HIA	2.20E-10	2.54E-10	1.76E-10	2.81E-11	2.30E-10	2.79E-10
T8LCS1-5RAS	1.93E-10	8.43E-11	2.16E-10	9.48E-12	1.65E-10	5.16E-11
	Iodide					
T8LCS1-5RAS	2.29E-09	1.06E-09	2.55E-09	3.87E-10	1.91E-09	6.74E-10
T18LCS2-7.8RAS	3.39E-09	1.53E-09	3.15E-09	4.39E-10	2.78E-09	5.87E-10
T15HCS1-7.8HIS	5.53E-09	5.52E-09	4.27E-09	3.80E-09	3.32E-09	2.46E-09
T10HCS1-5HIS	4.88E-09	5.03E-09	4.44E-09	1.59E-09	2.84E-09	1.73E-09
T5HCS1-7.8AVG	5.60E-09	6.98E-09	5.30E-09	7.89E-10	8.20E-10	2.78E-09
T14LCS2-7.8HIS	6.68E-09	8.42E-09	5.70E-09	2.26E-09	3.22E-09	2.61E-09
T17LCS2-5HIA	8.24E-09	7.86E-09	7.58E-09	2.52E-09	5.08E-09	3.12E-09
T16HCS1-7.8RAS	6.97E-09	4.57E-09	7.86E-09	1.81E-09	5.24E-09	2.63E-09
T24HCS1-5HIA	7.93E-09	8.62E-09	8.13E-09	2.92E-09	4.56E-09	3.75E-09
T13LCS2-5AVG	8.24E-09	8.31E-09	8.47E-09	2.89E-09	4.93E-09	3.71E-09
T21LCS1-7.8HIS	1.34E-08	2.12E-08	1.67E-08	8.12E-09	6.17E-09	8.45E-09
	Na					
T8LCS1-5RAS	2.21E-09	9.47E-10	2.40E-09	2.84E-10	1.86E-09	5.78E-10
T18LCS2-7.8RAS	2.85E-09	1.08E-09	2.85E-09	2.27E-10	2.44E-09	6.28E-10
T14LCS2-7.8HIS	4.24E-09	4.04E-09	4.45E-09	1.08E-09	2.64E-09	1.78E-09
T5HCS1-7.8AVG	5.47E-09	5.24E-09	5.58E-09	2.04E-09	3.45E-09	2.31E-09
T13LCS2-5AVG	6.04E-09	5.58E-09	6.43E-09	1.79E-09	3.86E-09	2.61E-09
T17LCS2-5HIA	6.29E-09	5.25E-09	6.67E-09	1.87E-09	4.24E-09	2.70E-09
T16HCS1-7.8RAS	6.12E-09	3.15E-09	7.07E-09	1.11E-09	4.97E-09	2.13E-09
T15HCS1-7.8HIS	6.25E-09	4.29E-09	7.35E-09	1.55E-09	4.66E-09	2.62E-09
T10HCS1-5HIS	7.50E-09	6.65E-09	7.50E-09	1.99E-09	2.50E-09	2.73E-09
T24HCS1-5HIA	8.52E-09	7.29E-09	9.79E-09	2.53E-09	5.75E-09	4.01E-09
T21LCS1-7.8HIS	8.19E-09	1.17E-08	1.04E-08	3.39E-09	4.08E-09	4.74E-09

	De 7d to 604/609 d Avg.	σ	De 28 d to 63 d Avg.	σ	De 28 d to 604/609 Avg.	σ
	NO_2					
T8LCS1-5RAS	2.44E-09	1.39E-09	2.44E-09	4.29E-10	1.91E-09	6.31E-10
T18LCS2-7.8RAS	3.22E-09	1.48E-09	2.95E-09	2.93E-10	2.62E-09	5.50E-10
T10HCS1-5HIS	6.59E-09	7.80E-09	5.57E-09	1.84E-09	3.54E-09	7.16E-10
T14LCS2-7.8HIS	7.12E-09	9.73E-09	5.93E-09	2.11E-09	3.29E-09	2.71E-09
T5HCS1-7.8AVG	6.78E-09	8.38E-09	6.23E-09	3.08E-09	3.52E-09	3.10E-09
T15HCS1-7.8HIS	7.22E-09	6.59E-09	7.18E-09	2.73E-09	4.54E-09	3.02E-09
T16HCS1-7.8RAS	6.86E-09	4.60E-09	7.36E-09	1.71E-09	5.07E-09	2.49E-09
T17LCS2-5HIA	8.00E-09	7.74E-09	7.36E-09	2.24E-09	4.76E-09	2.91E-09
T13LCS2-5AVG	8.10E-09	9.48E-09	7.46E-09	2.52E-09	4.33E-09	3.26E-09
T24HCS1-5HIA	7.93E-09	7.89E-09	7.96E-09	1.64E-09	4.81E-09	3.18E-09
T21LCS1-7.8HIS	1.07E-08	1.48E-08	1.33E-08	4.02E-09	5.54E-09	5.96E-09
	NO_3					
T8LCS1-5RAS	2.65E-09	1.23E-09	2.60E-09	5.21E-10	2.18E-09	1.23E-09
T18LCS2-7.8RAS	3.71E-09	1.46E-09	3.24E-09	4.55E-10	3.13E-09	5.37E-10
T10HCS1-5HIS	7.09E-09	7.53E-09	6.10E-09	2.25E-09	4.06E-09	2.30E-09
T5HCS1-7.8AVG	6.90E-09	8.37E-09	6.20E-09	3.11E-09	3.50E-09	3.05E-09
T14LCS2-7.8HIS	7.22E-09	8.90E-09	6.26E-09	2.47E-09	3.63E-09	2.80E-09
T15HCS1-7.8HIS	7.12E-09	6.44E-09	7.02E-09	2.61E-09	4.53E-09	2.81E-09
T17LCS2-5HIA	9.03E-09	8.23E-09	7.71E-09	2.60E-09	5.87E-09	3.97E-09
T16HCS1-7.8RAS	7.48E-09	4.53E-09	8.04E-09	2.03E-09	5.75E-09	2.50E-09
T13LCS2-5AVG	8.86E-09	9.50E-09	8.40E-09	3.23E-09	5.03E-09	3.65E-09
T24HCS1-5HIA	9.42E-09	9.63E-09	9.61E-09	2.92E-09	5.69E-09	2.91E-09
T21LCS1-7.8HIS	1.15E-08	1.71E-08	1.46E-08	5.79E-09	5.73E-09	6.91E-09
	Cr					
T14LCS2-7.8HIS	1.02E-14	3.03E-15	9.91E-15	2.25E-15	9.80E-15	2.64E-15
T18LCS2-7.8RAS	<2.76E-14	1.79E-14	<1.84E-14	1.19E-14	<2.98E-14	1.89E-14
T13LCS2-5AVG	6.68E-14	3.72E-14	5.50E-14	9.00E-15	5.46E-14	1.63E-14
T5HCS1-7.8AVG	9.10E-14	1.05E-13	8.00E-14	2.82E-14	5.70E-14	2.28E-14
T24HCS1-5HIA	1.27E-13	1.37E-13	9.41E-14	3.68E-14	8.36E-14	2.95E-14
T21LCS1-7.8HIS	2.05E-13	1.22E-13	1.56E-13	7.19E-14	2.29E-13	1.13E-13
T17LCS2-5HIA	2.07E-13	1.81E-13	1.65E-13	2.52E-14	2.14E-13	2.00E-13
T16HCS1-7.8RAS	3.00E-13	2.49E-13	2.44E-13	5.39E-14	2.06E-13	7.75E-14
T15HCS1-7.8HIS	7.87E-13	1.43E-12	3.16E-13	3.07E-13	1.79E-13	2.26E-13
T10HCS1-5HIS	8.18E-13	1.01E-12	6.99E-13	3.99E-13	4.80E-13	3.32E-13
T8LCS1-5RAS	6.10E-13	3.05E-13	7.15E-13	1.02E-13	5.14E-13	2.19E-13

Note: None of the interval averaged D_e values has been corrected for inventory depletion.

The average interval D_e 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_e 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_e 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 reoxidation, to higher solubility species from the initially reduced (low solubility) species, followed by diffusion through the tortuous Cast Stone matrix. Table 3.1 also shows the standard deviation for each of the three specified interval D_e averages. In general, the standard deviations for the more leachable nitrate, nitrite, iodide, and sodium are large because the interval D_e values are, in most instances, continually decreasing with increasing time. A key reason for the decrease in D_e 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 3.1), nitrite, nitrate, and iodide reach >20% depletion between 7 and 14 days of cumulative leaching. Recall that ANS16.1 protocol states that when a monolith releases >20% of its starting inventory for a constituent, that the observed D_e calculated using Equation 1 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% 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_e values in Table 3.1 (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_e values for monoliths leached in VZP that are our recommended "base-case," or most probable, D_e values (most representative of the IDF subsurface burial environment). Tables 3.3, 3.4, 3.6, and 3.8 interval averaged D_e values for monoliths leached in VZP have not been corrected for inventory depletion. Bear in mind that when the recommended D_e 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% for long time periods. 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_e values should be considered in future IDF PA activities.

A second plausible reason for the decrease in D_e values versus time (shown in Table 3.1) is the long leaching intervals used in the extended leach tests out beyond 91 days. Such long intervals between refreshing the leachant (30 to 47 days with one 305 day interval when the project was shut off) could have allowed the leachate concentrations of leachable constituents to increase enough to significantly lower the concentration gradient as a driving force for diffusion. However, as shown in Appendix B for each monolith leach test, the concentration of mobile constituents in the leachates from the longer leach intervals beyond 91 days, are significantly lower than leachates 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 leachates to reduce the diffusion gradient. Therefore, because the longer leach intervals used in the extended leach testing resulted in lower leachate concentrations, this second cause for the observed decrease in D_e values is likely not the main cause.

Photographs of some of the most leach resistant and least leach resistant monoliths after ~555 days of leaching are shown in Figure 3.1. The upper two photographs are monoliths from Mix 8 and Mix 18 that showed the lowest D_e values for the mobile constituents, such as nitrate, nitrite, iodide, and sodium, as shown in the rankings in Table 3.1. The bottom two photographs are monoliths from Mixes 21 and 24, which showed the highest D_e values for the mobile constituents. All four photographs show that the monoliths have remained intact throughout the 550 to 555 days of leaching.[1] Two of the photographs show some white precipitate either clinging to the monolith 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 3.2). 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 the 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 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 3.1 suggest that the monoliths have little signs of significant degradation or extensive secondary mineral formation on the outer surfaces after 20 months of leaching in DIW.

[1] Note that as this report was being finalized photographs of selected Extended Suite monoliths after 790 days of leaching became available and some monoliths are showing more cracks has developed and some monoliths are showing more mass of white precipitate. Analytical data for leachates are not yet available to assess what impact more cracking may have on interval D_e values.

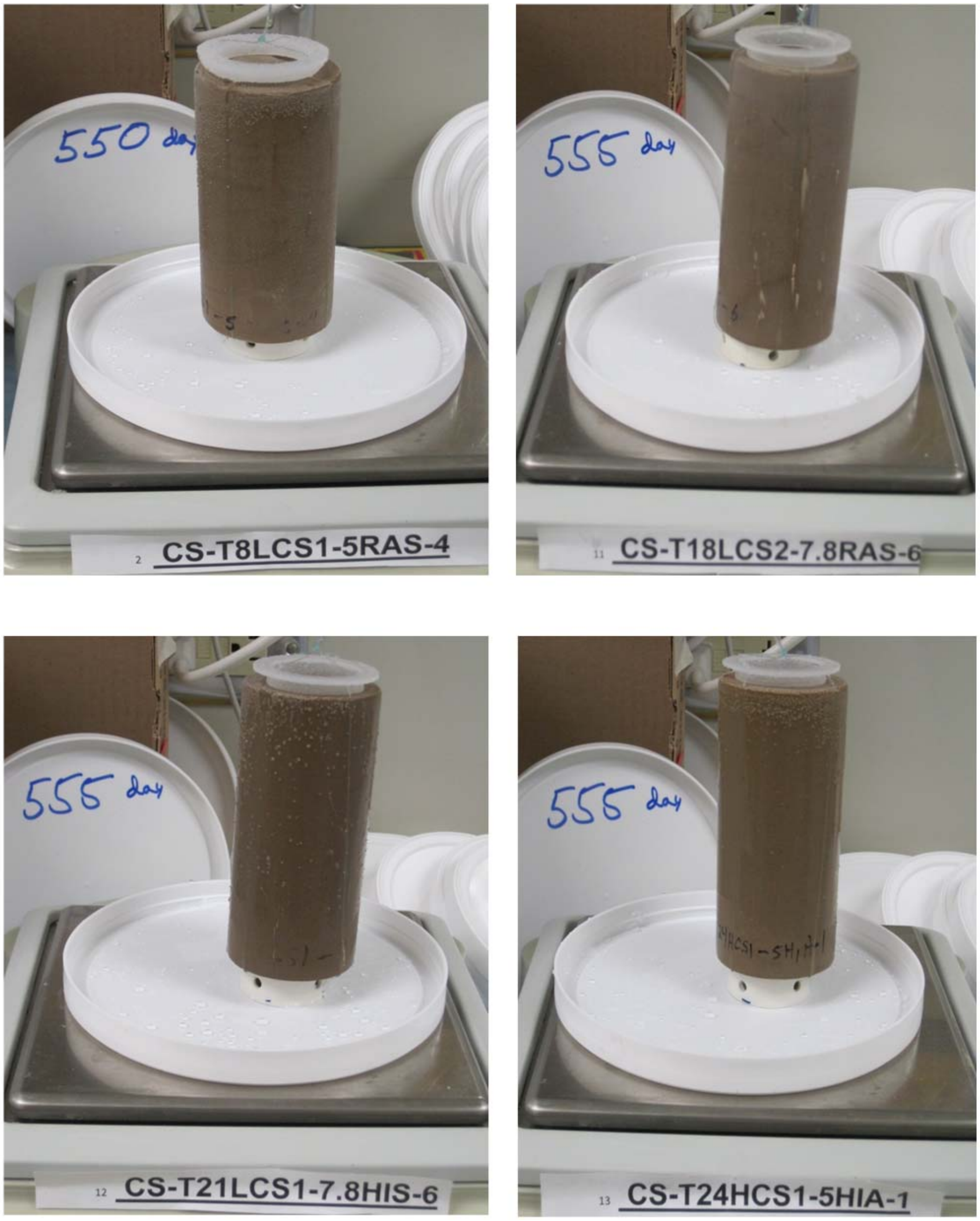


Figure 3.1. Photographs of •Screening Leach Tests –Extended Suite Monoliths after ~555 days of Leaching in DIW

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 effective diffusivities and almost an order of magnitude range in effective 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.

3.2 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 than the DIW leachant used throughout the screening tests. The more realistic leachant is the VZP described in Section 2.4. 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 100% relative humidity. These stored monoliths are called archived monoliths for this report.

Table 3.2 lists the monoliths and leachant 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. Because two monoliths of each mix were already leached in DIW, 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 3.3 presents average interval D_e values for the Archive Leach Test Suite monoliths similar to the results shown in Table 3.1 for the Extended Suite of monoliths, which were leached solely in DIW. The Archive Suite leach test results are available through 427 days of leaching in comparison to the Extended Suite, which has leach data through 604-609 days, dependent on the Cast Stone mix. Again, the interval D_e values for the two replicates leached in VZP for the three different sampling periods were averaged (28-, 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 3.3 suggests that there is more leaching occurring from monoliths of a particular mix composition when the leachant is DIW than when using VZP as the leachant. This is especially evident for the leaching of ^{99}Tc and Cr. Figure 3.2 shows bar charts for the averaged interval D_e values for the 28-day to 63-day data. When leached in VZP, most of the averaged interval D_e values for Cr are not readily determinable. Because many of the VZP leachates did not contain detectable concentrations of Cr, we resorted to assuming that these leachates contained Cr concentrations at the sample detection limit. This assumption led to designating the calculated D_e values as “less than values”, e.g., $< x$ where x is the numerical value of D_e in units of cm^2/sec. In contrast, when leached in DIW, only one averaged interval Cr D_e data set contained leachates with Cr concentrations below detection. Table 3.4 presents the average interval D_e values for each COC sorted from low to high leaching.

Table 3.2. Listing of Archived Monoliths and Leachant Type Used in Leach Tests

Test or Mix #	VPZ Leachant	DIW Leachant
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-5 T21LCS1-7.8HIS-5	T21LCS1-7.8HIS-5
Test 24	T24HCS1-5HIA-4 T24HCS1-5HIA-5	T24HCS1-5HIA-6

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_e 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_e values. The sodium n 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_e 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_e 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_e values. The Cr in the Archive Leach Test Suite monoliths had many values less than the lower quantification limit for the VZP leachates; thus, no quantitative ranking of Cast Stone mixes is possible.

Table 3.3. Interval Averaged D_e Values (cm^2/s) for Archived Monoliths Leached in Either VZP or DIW

	Leachant = VZP							Leachant = DIW					
	D_e 7d to 427d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	Σ		D_e 7d to 427d Avg.	Σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	σ
Monolith	**Tc**						**Monolith**	**Tc**					
T3H2-7.8AVG	2.57E-12	6.19E-13	2.70E-12	5.94E-13	2.61E-12	6.67E-13	T3H2-7.8AVG	7.93E-11	7.27E-11	6.31E-11	3.10E-11	9.47E-11	7.61E-11
T5H-7.8AVG	1.40E-12	6.01E-13	1.60E-12	2.63E-13	1.23E-12	5.30E-13	T5H-7.8AVG	3.65E-11	1.54E-11	4.00E-11	1.10E-11	4.16E-11	1.28E-11
T8L-5RAS	6.36E-12	4.28E-12	5.17E-12	8.70E-13	4.33E-12	1.43E-12	T8L-5RAS	1.21E-10	2.57E-11	1.41E-10	9.71E-12	1.26E-10	2.53E-11
T10H-5HIS	6.31E-12	2.76E-12	6.90E-12	1.08E-12	5.28E-12	2.14E-12	T10H-5HIS	7.93E-11	1.19E-11	8.73E-11	5.55E-12	8.35E-11	8.81E-12
T13L-5AVG-5	1.22E-11	1.17E-11	7.86E-12	2.72E-12	1.44E-11	1.24E-11	T13L-5AVG-5	7.46E-11	9.68E-11	3.50E-11	1.13E-11	9.31E-11	1.03E-10
T14L-7.8HIS	2.22E-12	1.71E-12	2.97E-12	2.17E-12	2.34E-12	1.91E-12	T14L-7.8HIS	7.86E-12	1.37E-12	7.13E-12	5.94E-13	7.95E-12	1.55E-12
T15H-7.8HIS	1.34E-12	4.80E-13	1.54E-12	2.95E-13	1.23E-12	4.47E-13	T15H-7.8HIS	3.12E-11	1.03E-11	3.39E-11	6.93E-12	3.44E-11	8.22E-12
T16H-7.8RAS	2.40E-11	9.06E-12	3.02E-11	5.88E-12	2.33E-11	1.00E-11	T16H-7.8RAS	1.42E-10	2.52E-11	1.58E-10	1.56E-11	1.52E-10	1.87E-11
T17L-5HIA	2.02E-11	1.36E-11	1.84E-11	4.65E-12	2.19E-11	1.50E-11	T17L-5HIA	2.24E-10	1.01E-10	1.89E-10	1.50E-11	2.47E-10	1.03E-10
T18L2-7.8RAS	6.00E-12	1.24E-12	6.03E-12	7.74E-13	5.56E-12	9.40E-13	T18L2-7.8RAS	2.63E-11	8.17E-12	2.36E-11	3.10E-12	2.85E-11	7.88E-12
T21L-7.8HIS	5.44E-12	2.70E-12	5.73E-12	1.85E-12	6.43E-12	2.12E-12	T21L-7.8HIS	2.51E-10	1.15E-10	3.05E-10	7.48E-11	2.96E-10	7.59E-11
T24H-5HIA	6.89E-13	2.17E-13	7.55E-13	2.38E-13	6.77E-13	2.27E-13	T24H-5HIA	8.33E-12	6.04E-12	5.96E-12	4.70E-13	9.93E-12	5.93E-12
	I							**I**					
T3H2-7.8AVG	5.36E-09	2.23E-09	4.90E-09	4.66E-10	4.36E-09	8.07E-10	T3H2-7.8AVG	6.96E-09	1.85E-09	6.63E-09	1.03E-09	6.25E-09	1.11E-09
T5H-7.8AVG	2.65E-09	1.37E-09	2.73E-09	5.77E-10	2.10E-09	8.95E-10	T5H-7.8AVG	3.89E-09	1.47E-09	4.35E-09	6.31E-10	3.41E-09	1.29E-09
T8L-5RAS	2.04E-09	6.86E-10	2.20E-09	2.85E-10	1.80E-09	5.59E-10	T8L-5RAS	2.44E-09	1.02E-09	2.62E-09	4.60E-10	2.07E-09	8.20E-10
T10H-5HIS	1.80E-09	4.57E-10	1.88E-09	1.77E-10	1.63E-09	3.58E-10	T10H-5HIS	2.36E-09	2.85E-10	2.48E-09	9.00E-11	2.31E-09	3.08E-10
T13L-5AVG-5	3.06E-09	1.54E-09	2.68E-09	7.80E-10	2.36E-09	7.77E-10	T13L-5AVG-5	3.57E-09	1.41E-09	3.76E-09	2.37E-10	3.05E-09	1.04E-09
T14L-7.8HIS	3.85E-09	1.21E-09	3.68E-09	7.97E-10	3.34E-09	7.39E-10	T14L-7.8HIS	4.48E-09	1.63E-09	4.25E-09	4.80E-10	3.77E-09	7.92E-10
T15H-7.8HIS	2.15E-09	1.02E-09	2.34E-09	4.26E-10	1.77E-09	7.78E-10	T15H-7.8HIS	3.64E-09	1.28E-09	4.15E-09	9.42E-10	3.34E-09	1.31E-09
T16H-7.8RAS	2.90E-09	1.14E-09	2.91E-09	4.23E-10	2.41E-09	7.15E-10	T16H-7.8RAS	4.51E-09	1.08E-09	4.84E-09	8.18E-10	4.30E-09	1.13E-09
T17L-5HIA	4.19E-09	2.42E-09	3.10E-09	8.72E-10	3.04E-09	9.27E-10	T17L-5HIA	6.01E-09	3.23E-09	6.30E-09	1.37E-09	4.78E-09	2.38E-09

	Leachant = VZP						Leachant = DIW						
	D_e 7d to 427d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	Σ		D_e 7d to 427d Avg.	Σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	σ
T18L2-7.8RAS	3.30E-09	1.540E-09	2.88E-09	7.55E-10	2.61E-09	7.3E-10	T18L2-7.8RAS	3.07E-09	9.52E-10	3.04E-09	4.76E-10	2.71E-09	6.54E-10
T21L-7.8HIS	1.08E-08	6.44E-09	1.11E-08	2.93E-09	8.20E-09	4.29E-09	T21L-7.8HIS	1.53E-08	1.01E-08	1.58E-08	5.14E-09	1.13E-08	7.21E-09
T24H-5HIA	2.14E-09	1.25E-09	2.06E-09	3.75E-10	1.59E-09	6.59E-10	T24H-5HIA	2.58E-09	1.05E-09	2.48E-09	5.96E-10	2.17E-09	6.56E-10
	Na							Na					
T3H2-7.8AVG	4.38E-09	1.82E-09	4.37E-09	4.96E-10	3.60E-09	1.04E-09	T3H2-7.8AVG	5.84E-09	2.05E-09	5.52E-09	5.89E-10	5.18E-09	1.76E-09
T5H-7.8AVG	2.83E-09	1.33E-09	2.77E-09	3.23E-10	2.25E-09	7.10E-10	T5H-7.8AVG	4.22E-09	1.26E-09	4.45E-09	5.15E-10	3.85E-09	1.16E-09
T8L-5RAS	1.97E-09	7.16E-10	2.21E-09	2.92E-10	1.73E-09	6.29E-10	T8L-5RAS	2.39E-09	7.26E-10	2.60E-09	2.42E-10	2.16E-09	6.53E-10
T10H-5HIS	2.99E-09	1.11E-09	3.14E-09	2.48E-10	2.55E-09	7.62E-10	T10H-5HIS	4.11E-09	1.14E-09	4.09E-09	3.97E-10	3.77E-09	1.06E-09
T13L-5AVG-5	2.21E-09	1.27E-09	1.93E-09	3.56E-10	1.63E-09	5.15E-10	T13L-5AVG-5	2.95E-09	1.07E-09	3.03E-09	2.85E-10	2.60E-09	9.12E-10
T14L-7.8HIS	1.83E-09	7.71E-10	1.83E-09	3.32E-10	1.51E-09	4.62E-10	T14L-7.8HIS	2.57E-09	8.45E-10	2.47E-09	1.84E-10	2.24E-09	5.72E-10
T15H-7.8HIS	3.01E-09	1.26E-09	3.19E-09	3.32E-10	2.52E-09	8.62E-10	T15H-7.8HIS	4.59E-09	1.29E-09	4.94E-09	7.51E-10	4.23E-09	1.22E-09
T16H-7.8RAS	2.65E-09	1.14E-09	2.65E-09	3.51E-10	2.15E-09	6.60E-10	T16H-7.8RAS	4.48E-09	1.05E-09	4.74E-09	5.76E-10	4.21E-09	9.95E-10
T17L-5HIA	2.71E-09	1.68E-09	2.15E-09	3.64E-10	1.93E-09	6.55E-10	T17L-5HIA	4.35E-09	1.93E-09	4.38E-09	4.13E-10	3.57E-09	1.33E-09
T18L2-7.8RAS	2.39E-09	1.03E-09	2.21E-09	4.16E-10	1.93E-09	4.97E-10	T18L2-7.8RAS	2.58E-09	7.06E-10	2.37E-09	6.34E-10	2.33E-09	5.65E-10
T21L-7.8HIS	4.69E-09	2.48E-09	4.92E-09	1.07E-09	3.71E-09	1.75E-09	T21L-7.8HIS	7.74E-09	4.10E-09	8.15E-09	1.08E-09	6.10E-09	2..83E-09
T24H-5HIA	3.18E-09	1.90E-09	3.14E-09	5.13E-10	2.35E-09	1.05E-09	T24H-5HIA	3.96E-09	1.65E-09	3.70E-09	5.11E-10	3.27E-09	9.88E-10
	NO_2							NO_2					
T3H2-7.8AVG	3.92E-09	1.83E-09	4.03E-09	3.44E-10	3.17E-09	1.21E-09	T3H2-7.8AVG	5.31E-09	1.40E-09	5.39E-09	3.63E-10	4.81E-09	1.03E-09
T5H-7.8AVG	2.60E-09	1.18E-09	2.97E-09	6.21E-10	2.24E-09	1.07E-09	T5H-7.8AVG	4.04E-09	1.46E-09	4.78E-09	7.30E-10	3.69E-09	1.49E-09
T8L-5RAS	1.76E-09	8.16E-10	2.14E-09	3.12E-10	1.55E-09	7.96E-10	T8L-5RAS	2.23E-09	7.64E-10	2.47E-09	3.17E-10	1.98E-09	6.81E-10
T10H-5HIS	1.98E-09	8.13E-10	2.32E-09	3.55E-10	1.78E-09	8.04E-10	T10H-5HIS	2.72E-09	4.52E-10	2.85E-09	1.65E-10	2.57E-09	4.01E-10
T13L-5AVG-5	2.21E-09	1.10E-09	2.26E-09	4.43E-10	1.75E-09	7.17E-10	T13L-5AVG-5	2.97E-09	1.13E-09	3.26E-09	3.54E-10	2.58E-09	9.26E-10
T14L-7.8HIS	2.48E-09	1.08E-09	2.69E-09	6.30E-10	2.11E-09	8.98E-10	T14L-7.8HIS	3.21E-09	1.14E-09	3.15E-09	4.14E-10	2.72E-09	6.36E-10
T15H-7.8HIS	2.50E-09	1.17E-09	2.90E-09	4.69E-10	2.14E-09	1.07E-09	T15H-7.8HIS	4.03E-09	1.40E-09	4.82E-09	8.56E-10	3.80E-09	1.52E-09
T16H-7.8RAS	2.22E-09	9.51E-10	2.47E-09	1.82E-10	1.90E-09	8.19E-10	T16H-7.8RAS	3.88E-09	9.64E-10	4.33E-09	2.88E-10	3.66E-09	9.86E-10
T17L-5HIA	2.88E-09	1.68E-09	2.52E-09	6.26E-10	2.08E-09	7.16E-10	T17L-5HIA	4.35E-09	2.09E-09	4.53E-09	6.85E-10	3.51E-09	1.43E-09

	Leachant = VZP							**Leachant = DIW**					
	D_e 7d to 427d Avg.	**σ**	**D_e 28d to 63d Avg.**	**σ**	**D_e 28d to 427d Avg.**	**Σ**		**D_e 7d to 427d Avg.**	**Σ**	**D_e 28d to 63d Avg.**	**σ**	**D_e 28d to 427d Avg.**	**σ**
T18L2-7.8RAS	2.09E-09	1.02E-09	2.04E-09	4.32E-10	1.65E-09	6.25E-10	T18L2-7.8RAS	2.35E-09	6.91E-10	2.40E-09	2.91E-10	2.09E-09	4.88E-10
T21L-7.8HIS	7.88E-09	4.71E-09	8.79E-09	1.96E-09	6.16E-09	3.69E-09	T21L-7.8HIS	1.07E-08	6.41E-09	1..16E-08	2.56E-09	8.24E-09	4.87E-09
T24H-5HIA	2.23E-09	1.33E-09	2.28E-09	4.40E-10	1.67E-09	8.49E-10	T24H-5HIA	2.79E-09	1.18E-09	2.76E-09	3.26E-10	2.30E-09	7.02E-10
	NO_3							**NO_3**					
T3H2-7.8AVG	4.90E-09	1.97E-09	4.62E-09	7.49E-10	4.06E-09	9.39E-10	T3H2-7.8AVG	6.70E-09	1.51E-09	6.32E-09	1.29E-09	6.14E-09	1.02E-09
T5H-7.8AVG	3.52E-09	1.53E-09	3.66E-09	7.03E-10	2.91E-09	1.08E-09	T5H-7.8AVG	4.61E-09	1.63E-09	4.93E-09	8.88E-10	4.02E-09	1.30E-09
T8L-5RAS	2.48E-09	9.42E-10	2.80E-09	5.84E-10	2.19E-09	8.51E-10	T8L-5RAS	3.15E-09	1.08E-09	3.41E-09	7.14E-10	2.79E-09	9.43E-10
T10H-5HIS	2.98E-09	9.56E-10	3.05E-09	5.10E-10	2.60E-09	6.93E-10	T10H-5HIS	3.82E-09	5.35E-10	3.80E-09	5.03E-10	3.63E-09	4.09E-10
T13L-5AVG-5	3.61E-09	1.80E-09	3.19E-09	9.05E-10	2.79E-09	8.87E-10	T13L-5AVG-5	4.31E-09	1.51E-09	4.47E-09	7.64E-10	3.75E-09	1.09E-09
T14L-7.8HIS	3.85E-09	1.41E-09	3.79E-09	9.83E-10	3.28E-09	9.48E-10	T14L-7.8HIS	4.67E-09	1.43E-09	4.27E-09	8.13E-10	4.06E-09	6.42E-10
T15H-7.8HIS	3.68E-09	1.65E-09	3.97E-09	8.21E-10	3.06E-09	1.28E-09	T15H-7.8HIS	5.74E-09	1.93E-09	6.38E-09	1.78E-09	5.30E-09	1.97E-09
T16H-7.8RAS	3.63E-09	1.41E-09	3.69E-09	8.07E-10	3.06E-09	9.96E-10	T16H-7.8RAS	5.71E-09	1.22E-09	6.09E-09	8.56E-10	5.44E-09	1.23E-09
T17L-5HIA	5.11E-09	2.93E-09	4.02E-09	1.35E-09	3.75E-09	1.28E-09	T17L-5HIA	7.88E-09	5.12E-09	7.03E-09	2.13E-09	5.76E-09	2.27E-09
T18L2-7.8RAS	3.65E-09	1.65E-09	3.26E-09	1.11E-09	2.92E-09	9.29E-10	T18L2-7.8RAS	3.77E-09	1.01E-09	3.63E-09	8.39E-10	3.38E-09	6.81E-10
T21L-7.8HIS	1.02E-08	5.80E-09	1.09E-08	3.53E-09	8.03E-09	4.47E-09	T21L-7.8HIS	1.44E-08	8.94E-09	1.49E-08	5.40E-09	1.10E-08	6.60E-09
T24H-5HIA	3.25E-09	1.91E-09	3.06E-09	8.85E-10	2.41E-09	1.03E-09	T24H-5HIA	4.05E-09	1.66E-09	3.85E-09	1.02E-09	3.38E-09	1.01E-09
	Cr							**Cr**					
T3H2-7.8AVG	*<1.66E-15*	2.40E-15	*<2.95E-15*	3.16E-15	*<1.80E-15*	2.70E-15	T3H2-7.8AVG	2.14E-15	7.05E-16	2.30E-15	9.08E-16	2.14E-15	7.05E-1
T5H-7.8AVG	*<1.92E-15*	3.25E-15	*<4.07E-15*	4.10E-15	*<2.46E-15*	3.54E-15	T5H-7.8AVG	4.37E-14	5.11E-14	3.36E-14	1.65E-14	4.98E-14	5.73E-14
T8L-5RAS	*<3.16E-14*	3.90E-14	*<4.67E-14*	4.36E-14	*<2.83E-14*	3.90E-14	T8L-5RAS	2.45E-13	1.09E-13	2.30E-13	1.09E-13	2.38E-13	1.20E-13
T10H-5HIS	3.95E-14	7.22E-14	1.09E-14	1.36E-14	6.39E-15	1.14E-14	T10H-5HIS	2.37E-13	2.69E-13	1.32E-13	1.02E-14	1.16E-13	3.90E-14
T13L-5AVG-5	*<1.12E-14*	1.64E-14	*<7.85E-15*	8.23E-15	*<4.77E-15*	7.11E-15	T13L-5AVG-5	1.49E-12	4.39E-12	3.31E-12	6.59E-12	1.91E-12	4.98E-12
T14L-7.8HIS	*<2.13E-15*	3.21E-15	*<4.21E-15*	3.93E-15	*<2.55E-15*	3.51E-15	T14L-7.8HIS	6.19E-15	5.88E-15	5.70E-15	4.02E-15	4.15E-15	3.52E-15
T15H-7.8HIS	*<2.58E-15*	4.01E-15	*<3.45E-15*	4.28E-15	*<2.12E-15*	3.54E-15	T15H-7.8HIS	1.65E-13	1.14E-13	1.62E-13	4.79E-14	1.19E-13	6.71E-14
T16H-7.8RAS	*<7.35E-15*	1.04E-14	*<1.38E-14*	1.29E-14	*<8.39E-15*	1.15E-14	T16H-7.8RAS	1.25E-13	7.23E-14	1.07E-13	3.06E-14	1.16E-13	7.83E-14
T17L-5HIA	*<3.87E-15*	5.97E-15	*<6.54E-15*	8.26E-15	*<4.01E-15*	6.79E-15	T17L-5HIA	7.09E-14	3.12E-14	6.20E-14	2.47E-14	7.30E-14	3.55E-14

	Leachant = VZP						Leachant = DIW						
	D_e 7d to 427d Avg.	**σ**	**D_e 28d to 63d Avg.**	**σ**	**D_e 28d to 427d Avg.**	**Σ**		**D_e 7d to 427d Avg.**	**Σ**	**D_e 28d to 63d Avg.**	**σ**	**D_e 28d to 427d Avg.**	**σ**
T18L2-7.8RAS	*<8.01E-15*	1.11E-14	*<1.48E-14*	1.38E-14	*<9.06E-15*	1.22E-14	T18L2-7.8RAS	*<1.88E-14*	1.13E-14	*<2.06E-14*	1.50E-14	*<1.96E-14*	1.24E-14
T21L-7.8HIS	*<1.66E-15*	2.41E-15	*<3.06E-15*	3.07E-15	*<1.85E-15*	2.68E-15	T21L-7.8HIS	3.21E-14	1.68E-14	3.06E-14	1.43E-14	3.70E-14	1.58E-14
T24H-5HIA	*<5.29E-15*	6.98E-15	*<6.66E-15*	7.39E-15	*<4.07E-15*	6.27E-15	T24H-5HIA	5.90E-14	2.00E-14	5.58E-14	1.14E-14	5.30E-14	1.74E-14

Italicized numbers = interval D_e values included leachates where Cr concentrations were less than detection values. **Note that none of the interval averaged D_e values have been corrected for inventory depletion.**

Table 3.4. Interval Averaged D_e Values (cm2/s) for Archived Monoliths Leached in Either VZP or DIW—Sorted Based on 28 to 63 Day Interval Average

	Leachant = VZP							Leachant = DIW					
	D_e 7d to 427d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	Σ		D_e 7d to 427d Avg.	Σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	σ
Monolith	**Tc**						**Monolith**	**Tc**					
T24H-5HIA	6.89E-13	2.17E-13	7.55E-13	2.38E-13	6.77E-13	2.27E-13	T24H-5HIA	8.33E-12	6.04E-12	5.96E-12	4.70E-13	9.93E-12	5.93E-12
T15H-7.8HIS	1.34E-12	4.80E-13	1.54E-12	2.95E-13	1.23E-12	4.47E-13	T14L-7.8HIS	7.86E-12	1.37E-12	7.13E-12	5.94E-13	7.95E-12	1.55E-12
T5H-7.8AVG	1.40E-12	6.01E-13	1.60E-12	2.63E-13	1.23E-12	5.30E-13	T18L2-7.8RAS	2.63E-11	8.17E-12	2.36E-11	3.10E-12	2.85E-11	7.88E-12
T3H2-7.8AVG	2.57E-12	6.19E-13	2.70E-12	5.94E-13	2.61E-12	6.67E-13	T15H-7.8HIS	3.12E-11	1.03E-11	3.39E-11	6.93E-12	3.44E-11	8.22E-12
T14L-7.8HIS	2.22E-12	1.71E-12	2.97E-12	2.17E-12	2.34E-12	1.91E-12	T13L-5AVG-5	7.46E-11	9.68E-11	3.50E-11	1.13E-11	9.31E-11	1.03E-10
T8L-5RAS	6.36E-12	4.28E-12	5.17E-12	8.70E-13	4.33E-12	1.43E-12	T5H-7.8AVG	3.65E-11	1.54E-11	4.00E-11	1.10E-11	4.16E-11	1.28E-11
T21L-7.8HIS	5.44E-12	2.70E-12	5.73E-12	1.85E-12	6.43E-12	2.12E-12	T3H2-7.8AVG	7.93E-11	7.27E-11	6.31E-11	3.10E-11	9.47E-11	7.61E-11
T18L2-7.8RAS	6.00E-12	1.24E-12	6.03E-12	7.74E-13	5.56E-12	9.40E-13	T10H-5HIS	7.93E-11	1.19E-11	8.73E-11	5.55E-12	8.35E-11	8.81E-12
T10H-5HIS	6.31E-12	2.76E-12	6.90E-12	1.08E-12	5.28E-12	2.14E-12	T8L-5RAS	1.21E-10	2.57E-11	1.41E-10	9.71E-12	1.26E-10	2.53E-11
T13L-5AVG-5	1.22E-11	1.17E-11	7.86E-12	2.72E-12	1.44E-11	1.24E-11	T16H-7.8RAS	1.42E-10	2.52E-11	1.58E-10	1.56E-11	1.52E-10	1.87E-11
T17L-5HIA	2.02E-11	1.36E-11	1.84E-11	4.65E-12	2.19E-11	1.50E-11	T17L-5HIA	2.24E-10	1.01E-10	1.89E-10	1.50E-11	2.47E-10	1.03E-10
T16H-7.8RAS	2.40E-11	9.06E-12	3.02E-11	5.88E-12	2.33E-11	1.00E-11	T21L-7.8HIS	2.51E-10	1.15E-10	3.05E-10	7.48E-11	2.96E-10	7.59E-11
	I							**I**					
T10H-5HIS	1.80E-09	4.57E-10	1.88E-09	1.77E-10	1.63E-09	3.58E-10	T10H-5HIS	2.36E-09	2.85E-10	2.48E-09	9.00E-11	2.31E-09	3.08E-10
T24H-5HIA	2.14E-09	1.25E-09	2.06E-09	3.75E-10	1.59E-09	6.59E-10	T24H-5HIA	2.58E-09	1.05E-09	2.48E-09	5.96E-10	2.17E-09	6.56E-10
T8L-5RAS	2.04E-09	6.86E-10	2.20E-09	2.85E-10	1.80E-09	5.59E-10	T8L-5RAS	2.44E-09	1.02E-09	2.62E-09	4.60E-10	2.07E-09	8.20E-10
T15H-7.8HIS	2.15E-09	1.02E-09	2.34E-09	4.26E-10	1.77E-09	7.78E-10	T18L2-7.8RAS	3.07E-09	9.52E-10	3.04E-09	4.76E-10	2.71E-09	6.54E-10
T13L-5AVG-5	3.06E-09	1.54E-09	2.68E-09	7.80E-10	2.36E-09	7.77E-10	T13L-5AVG-5	3.57E-09	1.41E-09	3.76E-09	2.37E-10	3.05E-09	1.04E-09
T5H-7.8AVG	2.65E-09	1.37E-09	2.73E-09	5.77E-10	2.10E-09	8.95E-10	T15H-7.8HIS	3.64E-09	1.28E-09	4.15E-09	9.42E-10	3.34E-09	1.31E-09
T18L2-7.8RAS	3.30E-09	1.54E-09	2.88E-09	7.55E-10	2.61E-09	7.30E-10	T14L-7.8HIS	4.48E-09	1.63E-09	4.25E-09	4.80E-10	3.77E-09	7.92E-10
T16H-7.8RAS	2.90E-09	1.14E-09	2.91E-09	4.23E-10	2.41E-09	7.15E-10	T5H-7.8AVG	3.89E-09	1.47E-09	4.35E-09	6.31E-10	3.41E-09	1.29E-09
T17L-5HIA	4.19E-09	2.42E-09	3.10E-09	8.72E-10	3.04E-09	9.27E-10	T16H-7.8RAS	4.51E-09	1.08E-09	4.84E-09	8.18E-10	4.30E-09	1.13E-09

	Leachant = VZP							Leachant = DIW					
	D_e 7d to 427d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	Σ		D_e 7d to 427d Avg.	Σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	σ
T14L-7.8HIS	3.85E-09	1.21E-09	3.68E-09	7.97E-10	3.34E-09	7.39E-10	T17L-5HIA	6.01E-09	3.23E-09	6.30E-09	1.37E-09	4.78E-09	2.38E-09
T3H2-7.8AVG	5.36E-09	2.23E-09	4.90E-09	4.66E-10	4.36E-09	8.07E-10	T3H2-7.8AVG	6.96E-09	1.85E-09	6.63E-09	1.03E-09	6.25E-09	1.11E-09
T21L-7.8HIS	1.08E-08	6.44E-09	1.11E-08	2.93E-09	8.20E-09	4.29E-09	T21L-7.8HIS	1.53E-08	1.01E-08	1.58E-08	5.14E-09	1.13E-08	7.21E-09
	Na							Na					
T14L-7.8HIS	1.83E-09	7.71E-10	1.83E-09	3.32E-10	1.51E-09	4.62E-10	T18L2-7.8RAS	2.58E-09	7.06E-10	2.37E-09	6.34E-10	2.33E-09	5.65E-10
T13L-5AVG-5	2.21E-09	1.27E-09	1.93E-09	3.56E-10	1.63E-09	5.15E-10	T14L-7.8HIS	2.57E-09	8.45E-10	2.47E-09	1.84E-10	2.24E-09	5.72E-10
T17L-5HIA	2.71E-09	1.68E-09	2.15E-09	3.64E-10	1.93E-09	6.55E-10	T8L-5RAS	2.39E-09	7.26E-10	2.60E-09	2.42E-10	2.16E-09	6.53E-10
T8L-5RAS	1.97E-09	7.16E-10	2.21E-09	2.92E-10	1.73E-09	6.29E-10	T13L-5AVG-5	2.95E-09	1.07E-09	3.03E-09	2.85E-10	2.60E-09	9.12E-10
T18L2-7.8RAS	2.39E-09	1.03E-09	2.21E-09	4.16E-10	1.93E-09	4.97E-10	T24H-5HIA	3.96E-09	1.65E-09	3.70E-09	5.11E-10	3.27E-09	9.88E-10
T16H-7.8RAS	2.65E-09	1.14E-09	2.65E-09	3.51E-10	2.15E-09	6.60E-10	T10H-5HIS	4.11E-09	1.14E-09	4.09E-09	3.97E-10	3.77E-09	1.06E-09
T5H-7.8AVG	2.83E-09	1.33E-09	2.77E-09	3.23E-10	2.25E-09	7.10E-10	T17L-5HIA	4.35E-09	1.93E-09	4.38E-09	4.13E-10	3.57E-09	1.33E-09
T10H-5HIS	2.99E-09	1.11E-09	3.14E-09	2.48E-10	2.55E-09	7.62E-10	T5H-7.8AVG	4.22E-09	1.26E-09	4.45E-09	5.15E-10	3.85E-09	1.16E-09
T24H-5HIA	3.18E-09	1.90E-09	3.14E-09	5.13E-10	2.35E-09	1.05E-09	T16H-7.8RAS	4.48E-09	1.05E-09	4.74E-09	5.76E-10	4.21E-09	9.95E-10
T15H-7.8HIS	3.01E-09	1.26E-09	3.19E-09	3.32E-10	2.52E-09	8.62E-10	T15H-7.8HIS	4.59E-09	1.29E-09	4.94E-09	7.51E-10	4.23E-09	1.22E-09
T3H2-7.8AVG	4.38E-09	1.82E-09	4.37E-09	4.96E-10	3.60E-09	1.04E-09	T3H2-7.8AVG	5.84E-09	2.05E-09	5.52E-09	5.89E-10	5.18E-09	1.76E-09
T21L-7.8HIS	4.69E-09	2.48E-09	4.92E-09	1.07E-09	3.71E-09	1.75E-09	T21L-7.8HIS	7.74E-09	4.10E-09	8.15E-09	1.08E-09	6.10E-09	2..83E-09
	NO_2							NO_2					
T18L2-7.8RAS	2.09E-09	1.02E-09	2.04E-09	4.32E-10	1.65E-09	6.25E-10	T18L2-7.8RAS	2.35E-09	6.91E-10	2.40E-09	2.91E-10	2.09E-09	4.88E-10
T8L-5RAS	1.76E-09	8.16E-10	2.14E-09	3.12E-10	1.55E-09	7.96E-10	T8L-5RAS	2.23E-09	7.64E-10	2.47E-09	3.17E-10	1.98E-09	6.81E-10
T13L-5AVG-5	2.21E-09	1.10E-09	2.26E-09	4.43E-10	1.75E-09	7.17E-10	T24H-5HIA	2.79E-09	1.18E-09	2.76E-09	3.26E-10	2.30E-09	7.02E-10
T24H-5HIA	2.23E-09	1.33E-09	2.28E-09	4.40E-10	1.67E-09	8.49E-10	T10H-5HIS	2.72E-09	4.52E-10	2.85E-09	1.65E-10	2.57E-09	4.01E-10
T10H-5HIS	1.98E-09	8.13E-10	2.32E-09	3.55E-10	1.78E-09	8.04E-10	T14L-7.8HIS	3.21E-09	1.14E-09	3.15E-09	4.14E-10	2.72E-09	6.36E-10
T16H-7.8RAS	2.22E-09	9.51E-10	2.47E-09	1.82E-10	1.90E-09	8.19E-10	T13L-5AVG-5	2.97E-09	1.13E-09	3.26E-09	3.54E-10	2.58E-09	9.26E-10
T17L-5HIA	2.88E-09	1.68E-09	2.52E-09	6.26E-10	2.08E-09	7.16E-10	T16H-7.8RAS	3.88E-09	9.64E-10	4.33E-09	2.88E-10	3.66E-09	9.86E-10
T14L-7.8HIS	2.48E-09	1.08E-09	2.69E-09	6.30E-10	2.11E-09	8.98E-10	T17L-5HIA	4.35E-09	2.09E-09	4.53E-09	6.85E-10	3.51E-09	1.43E-09

	Leachant = VZP							Leachant = DIW					
	D_e 7d to 427d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	Σ		D_e 7d to 427d Avg.	Σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	σ
T15H-7.8HIS	2.50E-09	1.17E-09	2.90E-09	4.69E-10	2.14E-09	1.07E-09	T5H-7.8AVG	4.04E-09	1.46E-09	4.78E-09	7.30E-10	3.69E-09	1.49E-09
T5H-7.8AVG	2.60E-09	1.18E-09	2.97E-09	6.21E-10	2.24E-09	1.07E-09	T15H-7.8HIS	4.03E-09	1.40E-09	4.82E-09	8.56E-10	3.80E-09	1.52E-09
T3H2-7.8AVG	3.92E-09	1.83E-09	4.03E-09	3.44E-10	3.17E-09	1.21E-09	T3H2-7.8AVG	5.31E-09	1.40E-09	5.39E-09	3.63E-10	4.81E-09	1.03E-09
T21L-7.8HIS	7.88E-09	4.71E-09	8.79E-09	1.96E-09	6.16E-09	3.69E-09	T21L-7.8HIS	1.07E-08	6.41E-09	1..16E-08	2.56E-09	8.24E-09	4.87E-09
	NO_3							NO_3					
T8L-5RAS	2.48E-09	9.42E-10	2.80E-09	5.84E-10	2.19E-09	8.51E-10	T8L-5RAS	3.15E-09	1.08E-09	3.41E-09	7.14E-10	2.79E-09	9.43E-10
T10H-5HIS	2.98E-09	9.56E-10	3.05E-09	5.10E-10	2.60E-09	6.93E-10	T18L2-7.8RAS	3.77E-09	1.01E-09	3.63E-09	8.39E-10	3.38E-09	6.81E-10
T24H-5HIA	3.25E-09	1.91E-09	3.06E-09	8.85E-10	2.41E-09	1.03E-09	T10H-5HIS	3.82E-09	5.35E-10	3.80E-09	5.03E-10	3.63E-09	4.09E-10
T13L-5AVG-5	3.61E-09	1.80E-09	3.19E-09	9.05E-10	2.79E-09	8.87E-10	T24H-5HIA	4.05E-09	1.66E-09	3.85E-09	1.02E-09	3.38E-09	1.01E-09
T18L2-7.8RAS	3.65E-09	1.65E-09	3.26E-09	1.11E-09	2.92E-09	9.29E-10	T14L-7.8HIS	4.67E-09	1.43E-09	4.27E-09	8.13E-10	4.06E-09	6.42E-10
T5H-7.8AVG	3.52E-09	1.53E-09	3.66E-09	7.03E-10	2.91E-09	1.08E-09	T13L-5AVG-5	4.31E-09	1.51E-09	4.47E-09	7.64E-10	3.75E-09	1.09E-09
T16H-7.8RAS	3.63E-09	1.41E-09	3.69E-09	8.07E-10	3.06E-09	9.96E-10	T5H-7.8AVG	4.61E-09	1.63E-09	4.93E-09	8.88E-10	4.02E-09	1.30E-09
T14L-7.8HIS	3.85E-09	1.41E-09	3.79E-09	9.83E-10	3.28E-09	9.48E-10	T16H-7.8RAS	5.71E-09	1.22E-09	6.09E-09	8.56E-10	5.44E-09	1.23E-09
T15H-7.8HIS	3.68E-09	1.65E-09	3.97E-09	8.21E-10	3.06E-09	1.28E-09	T3H2-7.8AVG	6.70E-09	1.51E-09	6.32E-09	1.29E-09	6.14E-09	1.02E-09
T17L-5HIA	5.11E-09	2.93E-09	4.02E-09	1.35E-09	3.75E-09	1.28E-09	T15H-7.8HIS	5.74E-09	1.93E-09	6.38E-09	1.78E-09	5.30E-09	1.97E-09
T3H2-7.8AVG	4.90E-09	1.97E-09	4.62E-09	7.49E-10	4.06E-09	9.39E-10	T17L-5HIA	7.88E-09	5.12E-09	7.03E-09	2.13E-09	5.76E-09	2.27E-09
T21L-7.8HIS	1.02E-08	5.80E-09	1.09E-08	3.53E-09	8.03E-09	4.47E-09	T21L-7.8HIS	1.44E-08	8.94E-09	1.49E-08	5.40E-09	1.10E-08	6.60E-09
	Cr							Cr					
T3H2-7.8AVG	*<1.66E-15*	2.40E-15	*<2.95E-15*	3.16E-15	*<1.80E-15*	2.70E-15	T3H2-7.8AVG	2.14E-15	7.05E-16	2.30E-15	9.08E-16	2.14E-15	7.05E-01
T21L-7.8HIS	*<1.66E-15*	2.41E-15	*<3.06E-15*	3.07E-15	*<1.85E-15*	2.68E-15	T14L-7.8HIS	6.19E-15	5.88E-15	5.70E-15	4.02E-15	4.15E-15	3.52E-15
T15H-7.8HIS	*<2.58E-15*	4.01E-15	*<3.45E-15*	4.28E-15	*<2.12E-15*	3.54E-15	T18L2-7.8RAS	*<1.88E-14*	1.13E-14	*<2.06E-14*	1.50E-14	*<1.96E-14*	1.24E-14
T5H-7.8AVG	*<1.92E-15*	3.25E-15	*<4.07E-15*	4.10E-15	*<2.46E-15*	3.54E-15	T21L-7.8HIS	3.21E-14	1.68E-14	3.06E-14	1.43E-14	3.70E-14	1.58E-14
T14L-7.8HIS	*<2.13E-15*	3.21E-15	*<4.21E-15*	3.93E-15	*<2.55E-15*	3.51E-15	T5H-7.8AVG	4.37E-14	5.11E-14	3.36E-14	1.65E-14	4.98E-14	5.73E-14
T17L-5HIA	*<3.87E-15*	5.97E-15	*<6.54E-15*	8.26E-15	*<4.01E-15*	6.79E-15	T24H-5HIA	5.90E-14	2.00E-14	5.58E-14	1.14E-14	5.30E-14	1.74E-14
T24H-5HIA	*<5.29E-15*	6.98E-15	*<6.66E-15*	7.39E-15	*<4.07E-15*	6.27E-15	T17L-5HIA	7.09E-14	3.12E-14	6.20E-14	2.47E-14	7.30E-14	3.55E-14

	Leachant = VZP							Leachant = DIW					
	D_e 7d to 427d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	Σ		D_e 7d to 427d Avg.	Σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	σ
T13L-5AVG-5	*<1.12E-14*	1.64E-14	*<7.85E-15*	8.23E-15	*<4.77E-15*	7.11E-15	T16H-7.8RAS	1.25E-13	7.23E-14	1.07E-13	3.06E-14	1.16E-13	7.83E-14
T10H-5HIS	3.95E-14	7.22E-14	1.09E-14	1.36E-14	6.39E-15	1.14E-14	T10H-5HIS	2.37E-13	2.69E-13	1.32E-13	1.02E-14	1.16E-13	3.90E-14
T16H-7.8RAS	*<7.35E-15*	1.04E-14	*<1.38E-14*	1.29E-14	*<8.39E-15*	1.15E-14	T15H-7.8HIS	1.65E-13	1.14E-13	1.62E-13	4.79E-14	1.19E-13	6.71E-14
T18L2-7.8RAS	*<8.01E-15*	1.11E-14	*<1.48E-14*	1.38E-14	*<9.06E-15*	1.22E-14	T8L-5RAS	2.45E-13	1.09E-13	2.30E-13	1.09E-13	2.38E-13	1.20E-13
T8L-5RAS	*<3.16E-14*	3.90E-14	*<4.67E-14*	4.36E-14	*<2.83E-14*	3.90E-14	T13L-5AVG-5	1.49E-12	4.39E-12	3.31E-12	6.59E-12	1.91E-12	4.98E-12

Italicized numbers for Cr interval D_e values included leachates where Cr concentrations were less than detection values. **Note that none of the interval averaged D_e 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 3.4 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 18 are among the worst performing. For iodide, regardless of leachant 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 leachant used. For nitrate and nitrite, Cast Stone Mixes 18 and 8 leach less and Mixes 21 and 3 leach the most regardless of leachant used. As found for the Screening Leach Tests –extended results in the previous subsection, the Archive Leach Test Suite D_e 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_e values for these four constituents varies by only a factor of three to six.

Cr leaches the slowest, especially when the VZP leachant is used, followed by ^{99}Tc, and then the four more mobile constituents. 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_e values for the VZP leachant are recommended as the most probable values, if an empirical diffusivity conceptual model is chosen for the release from LAW Cast Stone. The D_e values for the COCs from earlier leaching periods (0.08 to 14 days) and the D_e values for tests that used DIW as the leachant could be used to create a range of D_e 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_e values; often an order of magnitude and in a few cases up to two orders of magnitude higher than interval D_e 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 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_e 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_e values to obtain a range to perform sensitivity analyses, the early leaching data and D_e 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. It is the solidified secondary waste simulants that will be addressed in the 2017 IDF PA. Detailed results 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_e values from using VZP leachant 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 (around day 93 of leaching in our data), the mass used to calculate the effective diffusion coefficient become less accurate. The accuracy issue arises because the leaching conditions no longer satisfy the assumed boundary conditions. Further, the D_e 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 interval D_e values as most representative of IDF burial conditions.

One possible reason for the observed lower interval D_e 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 3.3). We cannot confirm that there is enough surface precipitate on these monoliths leached in VZP to state objectively that the lower interval D_e 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_e 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 3.4 (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 3.4 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_e value compared to the similar test in the presence of carbon dioxide/carbonate. In our Cast Stone monolith leach tests, those performed in VZP leachant contained larger amounts of bicarbonate/carbonate than the tests performed in DIW; the measured D_e values follow the same trend as shown in Figure 3.4. Further, monolith weight measurements taken after each sampling and before placing the monoliths in the next batch of fresh leachant 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 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 3.15 in the Tc-Gluconate section shows an example of the monolith mass changes with time for one set of monoliths.

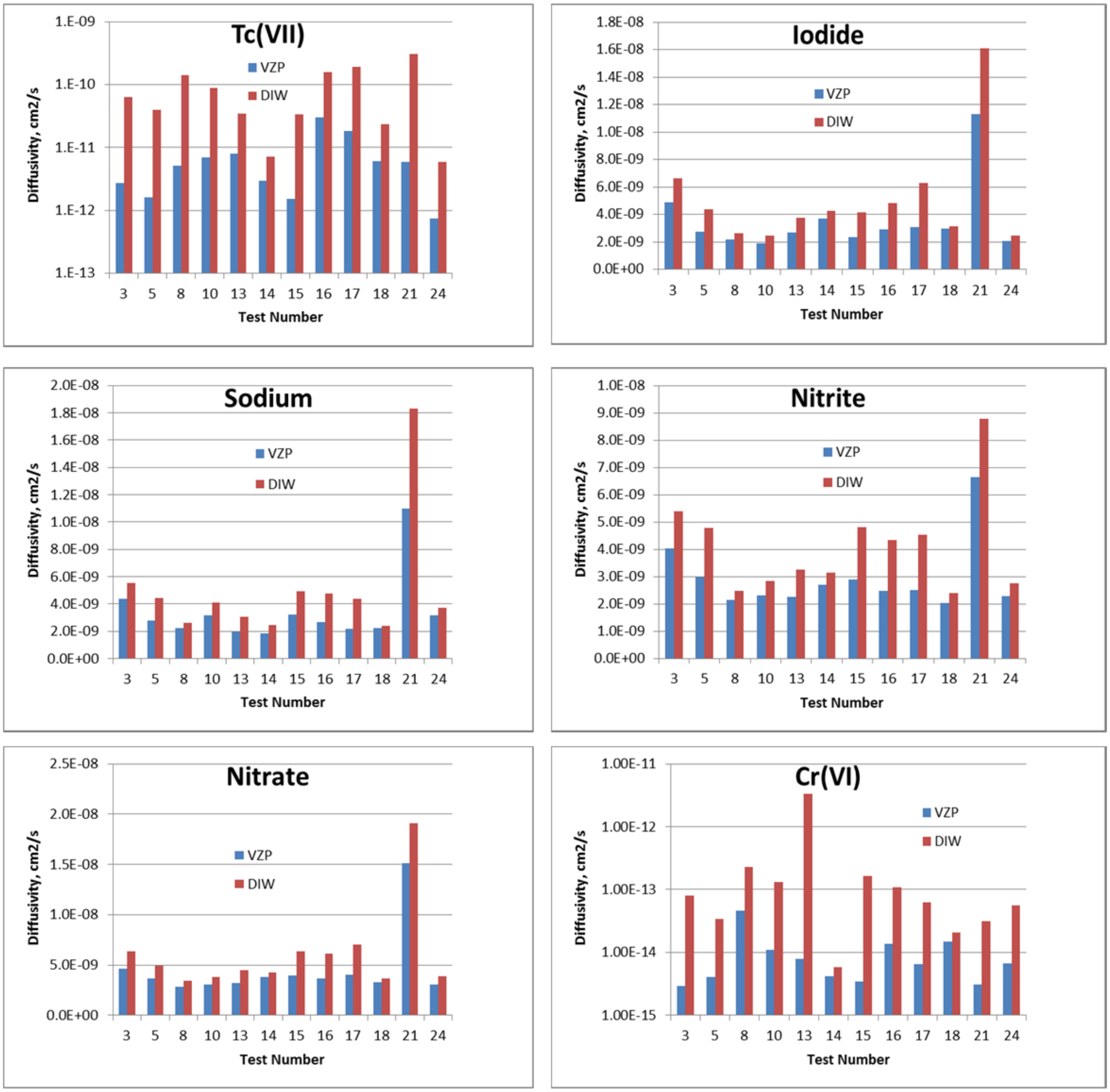
Another parameter that is consistently different for the VZP versus DIW leachates is pH. The VZP leachates' pH values are consistently lower than the DIW leachates' pH values. An example is shown in Figure 3.5 for Mix 5 monoliths. There is an increase in pH between the 93- and 380- day samplings for both leachates because of the long interval between samplings. Essentially, over this 287-day interval, more material diffused out of the monoliths from both leachates. Because the simulants themselves and the Cast Stone internal pore water are inherently caustic, as the time between changing out the leachant increased the leachates' pH increases. Increased pH in the leachates 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' leachates show a similar pH pattern as Mix 5, (see Appendix B.2 Table B.2.13 for leachate pH values for all the Extended Suite Leachates, Appendix B.2

Table B.2.13 for Archive Suite leachate pH values, Appendix B.3 Table B.3.9 for Iodide Loading Suite leachate pH values and Appendix B.4 Table B.4.11 for Tc-Gluconate Suite leachate pH values).

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 leachates 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 3.3). The cracks 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 after about 427 days of leaching. None of the Archive Leach Test Suite monoliths leached in VZP shows signs of fresh surface cracks, although the “fuzzy” white surface precipitates mask getting good looks at the monolith surface. Figure 3.6 shows a plot of interval D_e values that hint at an uptick in D_e between the 380- and 427-day sampling events. Not all the Archive Leach Test Suite monoliths’ DIW leachates show this trend, nor do all the constituents that we are tracking show this trend. Therefore, future leachates from monoliths leached in DIW at longer times will be scrutinized, along with doing more attentive visual study of monoliths between sampling events.

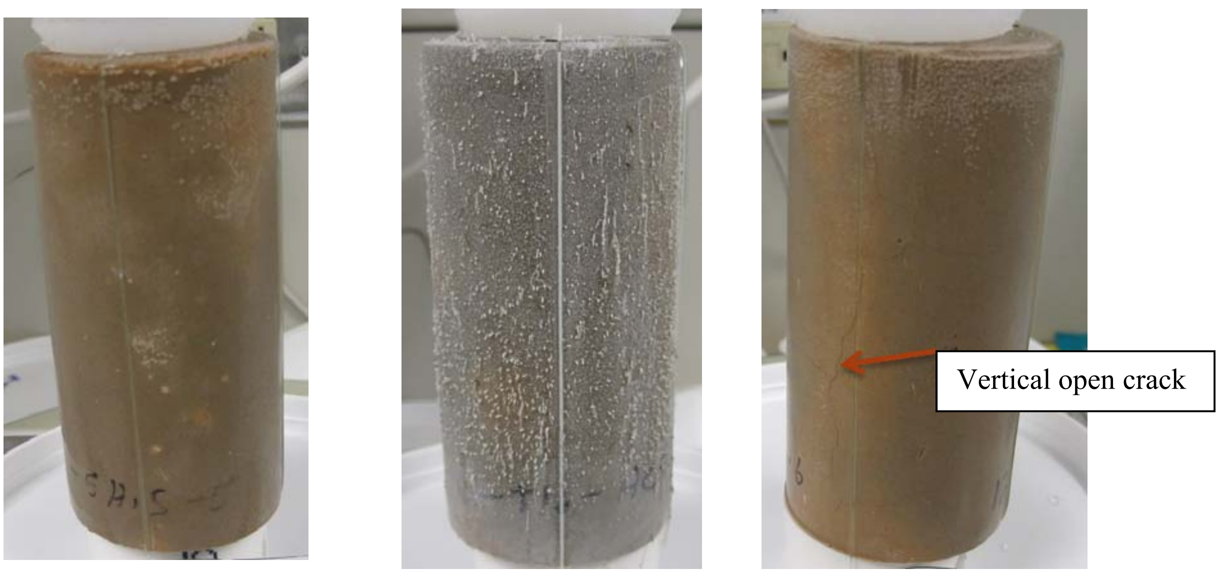
Observing surface cracks in monoliths submerged in water could lead to increased leaching of constituents from within the monoliths. However, in the IDF burial environment once the metal container is compromised and recharge water has the opportunity to contact Cast Stone solid waste forms, the presence 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.



Note: All average interval D_e Values for Cr in VZP except for test 10 are less than values and all average interval D_e Values for Cr in DIW except Test 18 are real values. Test 18 D_e value in DIW is less than.

Figure 3.2. Archive Interval D_e Values (cm^2/s) for 28-day to 63-day Samplings in DIW vs VZP Leachant

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

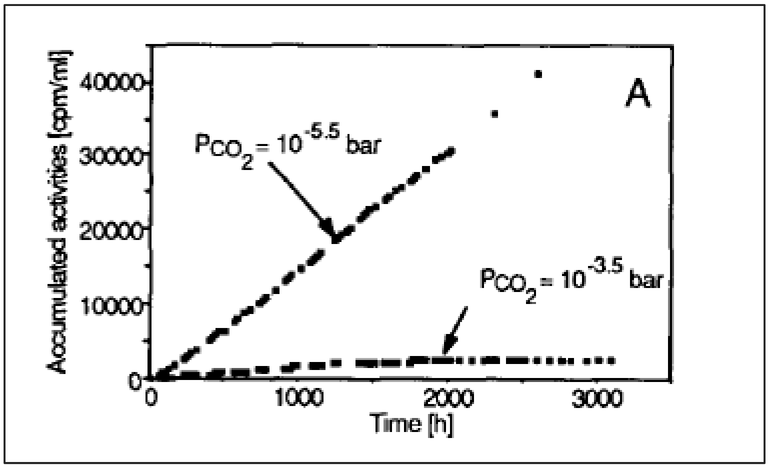


Figure 3.4. Cumulative Fraction of ^{125}I Passing through Cement Paste Disk Under Carbonate-Free vs Carbonate Containing Conditions
(figure 3A in Sarott et al. (1992))

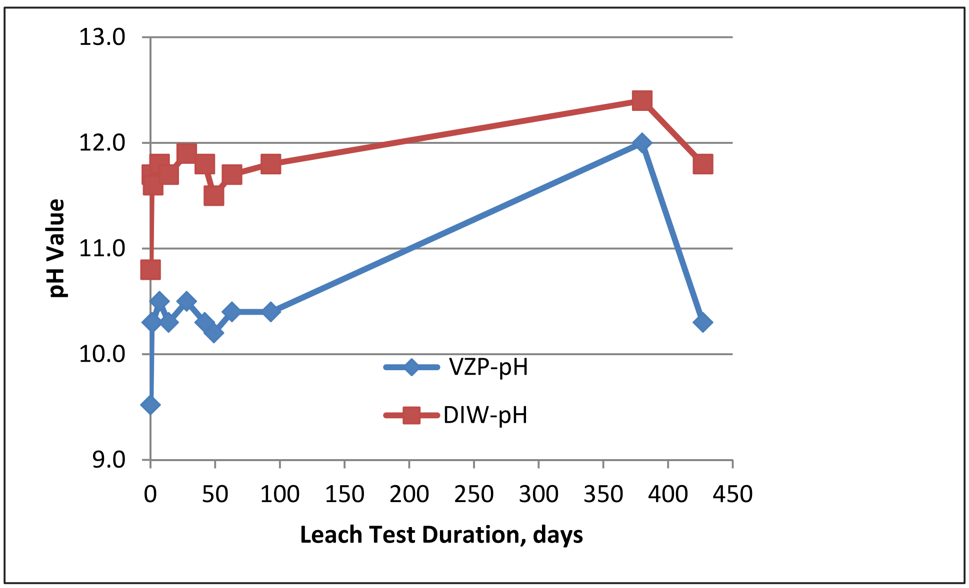
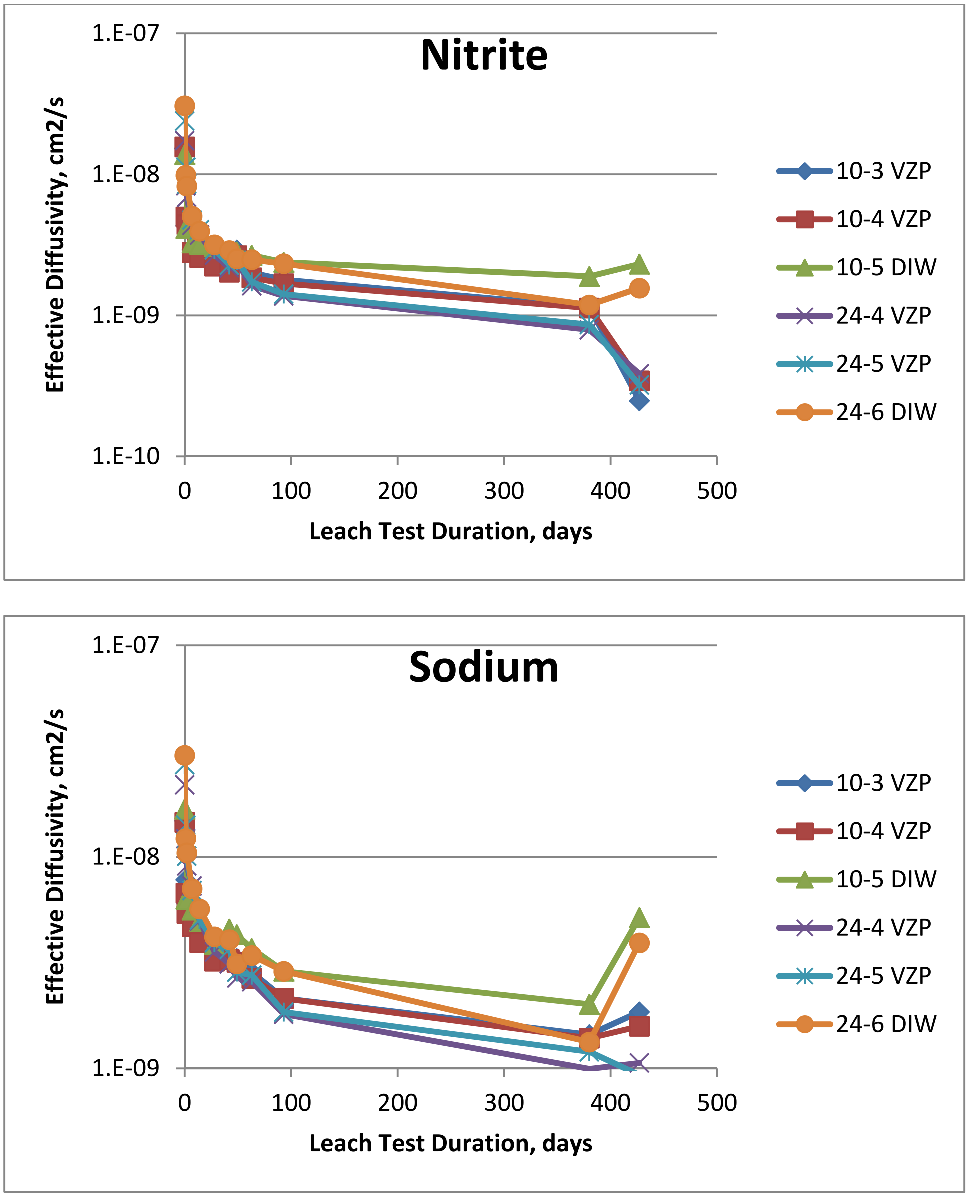


Figure 3.5. Typical pH Trend for Archive Suite Leachates vs. Leach Time



Note: uptick in Nitrite and Sodium D_e's for Mix 10 and Mix 24.

Figure 3.6. Uptick in D_e between 380 and 427 Days for Monoliths Leached in DIW

3.3 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 leachants. 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 leachates to facilitate calculation of interval D_e 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 et al. (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 3.7 shows that the iodide leach index (see Equation 2 for definition) in the Lockrem et al. (2005) study does in fact decrease (i.e., D_e increase by a factor of 10 between loadings of 0.14 to 1.4 wt% iodide.

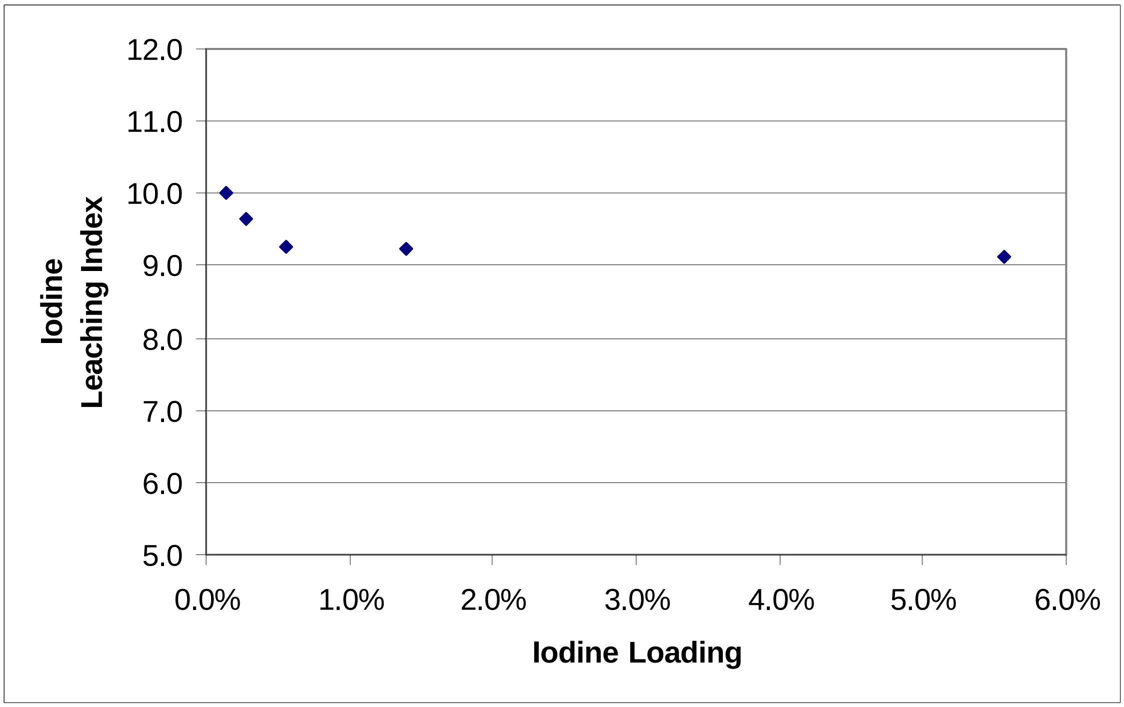


Figure 3.7. Leach Index of Iodide vs. Iodide Loading in Cast Stone (data from Lockrem et al. 2005)

Given the observed iodide leach sensitivity to loading and the Screening Test results that showed large 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_e values or higher LI values).

Table 3.5 lists the Iodide Suite of monoliths sample IDs, the iodide loading values, and the leachant 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 3.5 are at least a factor of 10 lower than the lowest iodide loading used by Lockrem et al. (2005), and more than 100 times below 1 wt% loading.

Table 3.5. Details on Iodide Suite of Monoliths

Sample ID	Iodide Spiking Level	Iodide Loading (mg/kg-dry)	Iodide Loading wt%	Leachant
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

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 3.6 provides the average interval D_e values for the same three sampling time intervals as used previously, when available. Data from Table 3.6 for the iodide average interval D_e values for the sampling period from 28- through 63- days are plotted as a bar chart in Figure 3.8. 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_e values, despite being replicates. When bar charts similar to Figure 3.8 are prepared for the other constituent averaged interval D_e values, the same trend (that Mix 3 shows the largest averaged interval D_e value and Mix 22 shows a lower averaged interval D_e value) is found (see Figure 3.9). 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_e 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 interval D_e 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.

Further, the standard deviations shown in Table 3.6 are large enough to preclude showing statistical differences in the averaged interval D_e 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_e values for other constituents.

Thus, we conclude that at iodide loading well below 0.14% (the lowest loading used by Lockrem et al. 2005)—as would be expected for all future Hanford waste streams based on HTWOS predictions—it should not matter what iodide loading below ~0.01 wt% is used in future simulant tests. 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 are used in future Cast Stone formulations.

As observed for the Archive Leach Test Suite of monoliths, the averaged interval D_e values for iodide from the Iodide Suite of monoliths suggests slightly lower leaching when VZP is the leachant 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. Recent photographs[1] of four of the I-Loading Suite monoliths after 569 days of leaching are shown in Figure 3.10. 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.

Similar to the Archive Leach Test Suite leachates, the I-Loading Suite leachates from DIW show higher pH values than the VZP leachates. A plot for the Iodide Suite leachates' pH is shown in Figure 3.11. Figure 3.11 is very similar to Figure 3.5, 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 leachate pH data for all Suites in Appendix B). All I-Loading Suite leachates 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 leachates. Between the 369-day and 414-day samplings, the pH of all I-Loading Suite leachates dropped.

[1] Photographs taken in late Feb 2015 after 569 days of leaching; leachate analyses are underway but not completed.

Table 3.6. Averaged Interval D_e Values (cm^2/s) for Iodide Suite of Monoliths and Similar Screening Test Monoliths

	Leachant = VZP							Leachant = DIW					
	D_e 7d to 414d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 414d Avg.	σ		D_e 7d to 414d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 414d Avg.	σ
	I							**I**					
T3HCS2-7.8AVG	5.36E-09	2.23E-09	4.90E-09	4.66E-10	4.36E-09	8.07E-10	T3HCS2-7.8AVG	6.96E-09	1.85E-09	6.63E-09	1.03E-09	6.25E-09	1.11E-09
Low I	5.47E-09	2.70E-09	4.59E-09	1.04E-09	4.42E-09	9.01E-10	Low I	6.05E-09	2.41E-09	4.69E-09	4.58E-10	5.08E-09	7.15E-10
Med I	4.32E-09	2.79E-09	3.26E-09	1.91E-10	3.14E-09	4.23E-10	Med I	6.01E-09	2.89E-09	4.66E-09	2.57E-10	4.82E-09	4.94E-10
High I	4.59E-09	2.88E-09	3.41E-09	2.98E-10	3.26E-09	3.11E-10	High I	5.96E-09	3.20E-09	5.09E-09	7.78E-10	5.04E-09	6.00E-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	4.38E-09	1.82E-09	4.37E-09	4.96E-10	3.60E-09	1.04E-09	T3HCS2-7.8AVG	5.84E-09	2.05E-09	5.52E-09	5.89E-10	5.18E-09	1.76E-09
Low I	3.29E-09	2.58E-09	2.80E-09	9.78E-10	2.18E-09	1.06E-09	Low I	4.46E-09	2.21E-09	4.12E-09	5.06E-10	3.52E-09	1.03E-09
Med I	3.59E-09	2.81E-09	2.94E-09	9.16E-10	2.30E-09	1.04E-09	Med I	4.58E-09	2.76E-09	3.83E-09	6.29E-10	3.37E-09	9.96E-10
High I	3.61E-09	3.02E-09	2.83E-09	1.01E-09	2.27E-09	1.04E-09	High I	4.23E-09	2.53E-09	4.07E-09	8.95E-10	3.43E-09	1.15E-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	4.82E-09	3.53E-09	4.03E-09	3.44E-10	4.34E-09	3.87E-09	T3HCS2-7.8AVG	5.31E-09	1.40E-09	5.39E-09	3.63E-10	4.81E-09	1.03E-09
Low I	2.08E-09	1.42E-09	1.82E-09	2.38E-10	1.47E-09	5.30E-10	Low I	2.83E-09	1.21E-09	2.48E-09	2.48E-10	2.30E-09	3.18E-10
Med I	2.04E-09	1.34E-09	1.76E-09	1.26E-10	1.45E-09	4.88E-10	Med I	2.75E-09	1.28E-09	2.32E-09	1.08E-10	2.17E-09	2.18E-10
High I	1.99E-09	1.36E-09	1.66E-09	1.47E-10	1.38E-09	4.64E-10	High I	2.57E-09	1.31E-09	2.36E-09	3.76E-10	2.18E-09	3.65E-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	4.90E-09	1.97E-09	4.62E-09	7.49E-10	4.06E-09	4.90E-09	T3HCS2-7.8AVG	6.70E-09	1.51E-09	6.32E-09	1.29E-09	6.14E-09	1.02E-09
Low I	3.24E-09	2.27E-09	2.63E-09	4.84E-10	2.26E-09	3.24E-09	Low I	4.45E-09	1.85E-09	3.77E-09	4.10E-10	3.65E-09	3.52E-10
Med I	3.14E-09	2.03E-09	2.40E-09	2.58E-10	2.29E-09	3.14E-09	Med I	4.42E-09	1.96E-09	3.49E-09	1.22E-10	3.56E-09	3.24E-10
High I	3.01E-09	2.07E-09	2.27E-09	2.55E-10	2.08E-09	3.01E-09	High I	4.06E-09	2.06E-09	3.55E-09	5.54E-10	3.47E-09	4.34E-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 leachates							Cr was not measured in Iodide Suite leachates					

Note that none of the interval averaged D_e values have been corrected for inventory depletion.

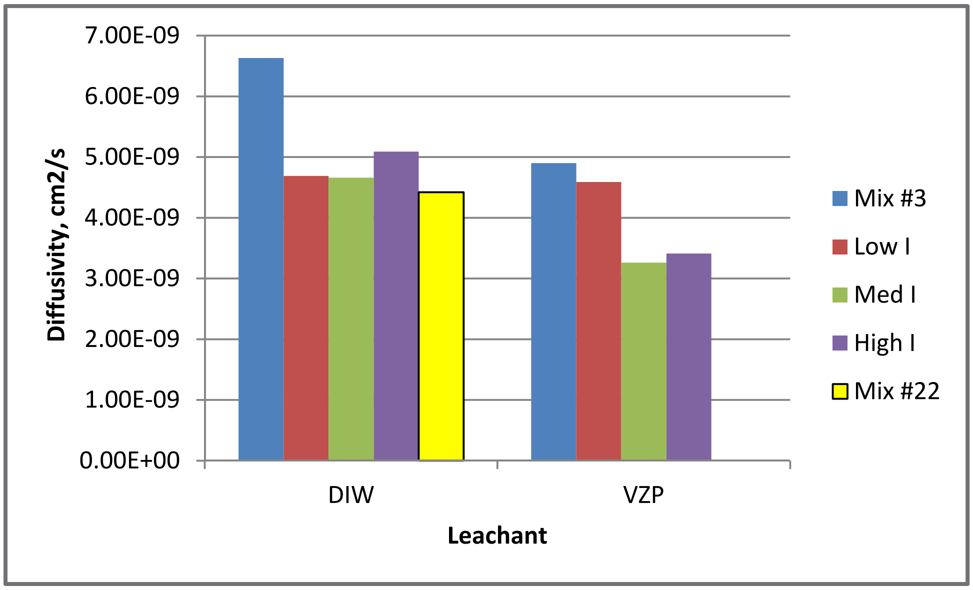


Figure 3.8. Comparison of the Iodide Averaged Interval D_e values (28 to 63-D) for Cast Stone Loaded with Varying Iodide Concentrations and Leachant Type

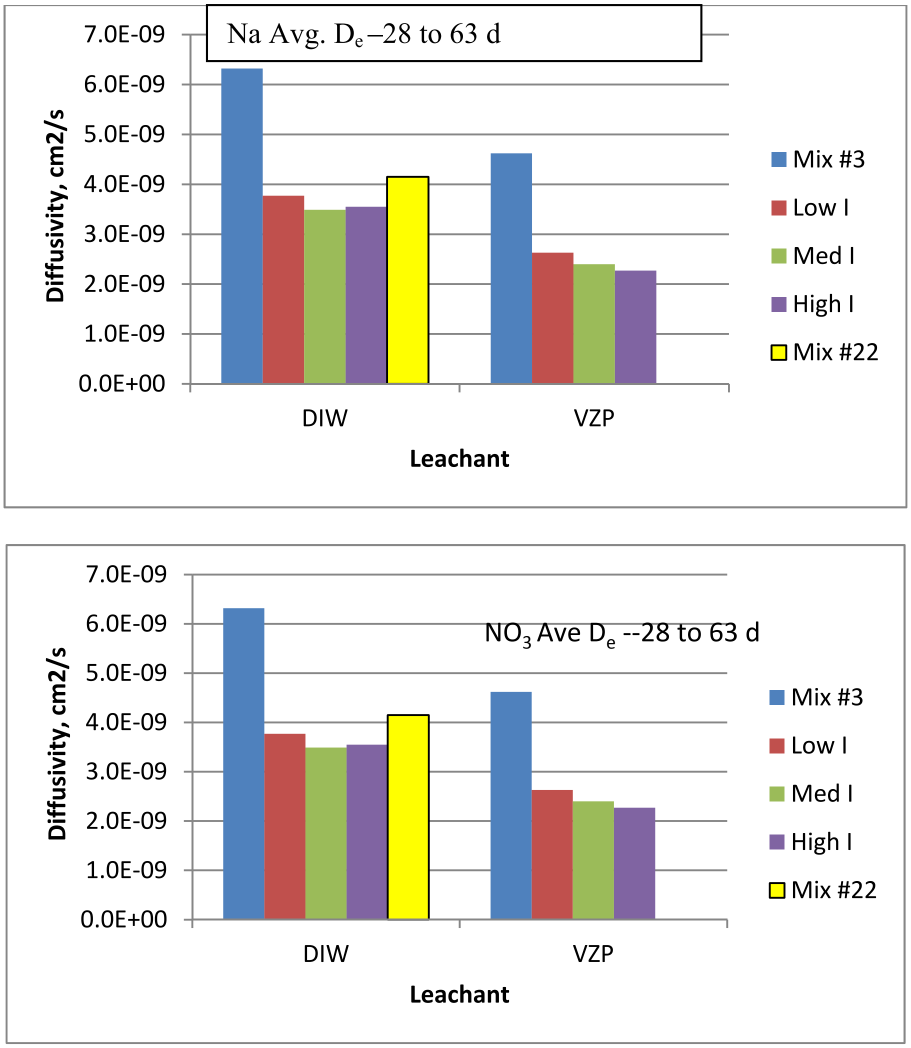


Figure 3.9. Averaged Interval D_e Values for Na and NO_3 from Iodide Suite and Mix 3and Mix 22 Monoliths

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).

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

Figure 3.10. Photographs of Iodide Suite Monoliths After 569 Days of Leaching

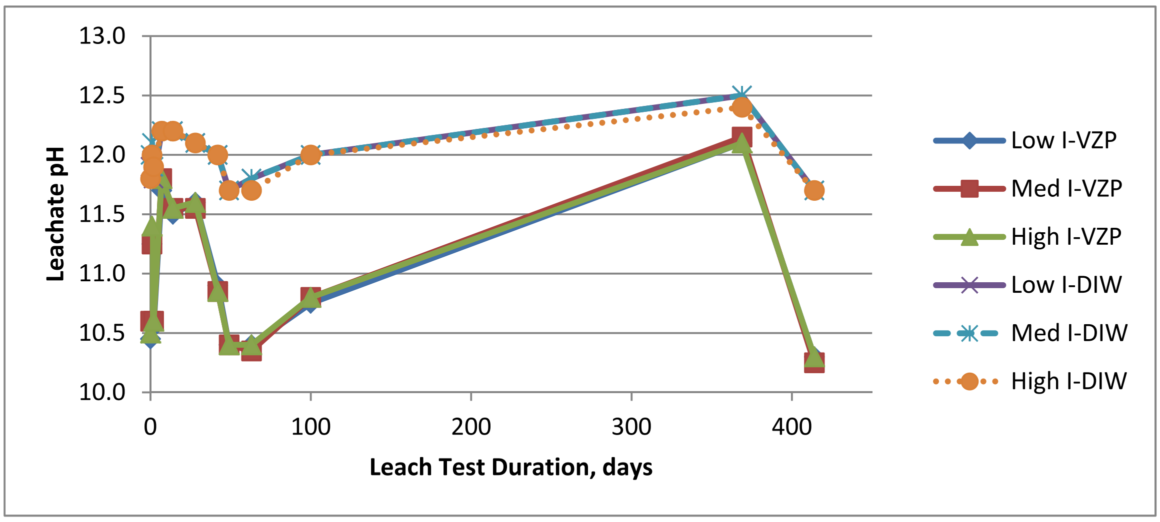


Figure 3.11. pH in Iodide Suite Leachates

3.4 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, 2014 and Serne and Rapko 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 leachants. 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.

Table 3.7 lists the Tc(I)-Gluconate Suite monolith IDs (and other relevant monoliths with Tc), details on Tc loading, and which leachant was used in the EPA Method 1315 leach tests. Table 3.8 shows the averaged interval D_e 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 leachants used and speciation of the Tc-spike. The averaged interval D_e values in each cell of Table 3.8 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. Figure 3.12 shows a bar chart for the averaged 28- to 63-day interval D_e values for ^{99}Tc for all the monoliths. Recall that only the Tc4-Gluc monoliths contain the non-pertechnetate spike, and all the other monoliths contain pertechnetate. Figure 3.12 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 leachant (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 3.13 shows the comparable averaged 28- to 63-day interval D_e values for nitrate and sodium from these Tc-bearing monoliths. Based on the fact that all the monoliths should have identical concentrations of nitrate, one might expect the averaged interval D_e values for nitrate to be similar. 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 leachants, 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 leachates 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 leachates 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 leachates 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 leachant 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_e value for pertechnetate. If this hypothesis is accurate the fact that the D_e 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 non-sorbing soluble species. We proffer that the Tc(I) species are not inert/stable and that they are being 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 3.7. 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%	Leachant	Duration of Leach Test (d)
Tc-Gluconate Suite					
CS-T4-VZ-1	Tc-gluconate	3.29	3.3×10^{-4}	VZP	408
CS-T4-VZ-2	Tc-gluconate	3.29	3.3×10^{-4}	VZP	408
CS-T4-DI-3	Tc-gluconate	3.29	3.3×10^{-4}	DIW	408
CS-T4-DI-4	Tc-gluconate	3.29	3.3×10^{-4}	DIW	408
CS-T5-VZ-1	Pertechnetate	7.915	7.9×10^{-4}	VZP	408
CS-T5-VZ-2	Pertechnetate	7.915	7.9×10^{-4}	VZP	408
CS-T5-DI-3	Pertechnetate	7.915	7.9×10^{-4}	DIW	408
CS-T5-DI-4	Pertechnetate	7.915	7.9×10^{-4}	DIW	408
Archive Suite					
T3HCS2-7.8AVG-3	Pertechnetate	9.32	9.3×10^{-4}	VZP	427
T3HCS2-7.8AVG-4	Pertechnetate	9.32	9.3×10^{-4}	VZP	427
T3HCS2-7.8AVG-5	Pertechnetate	9.32	9.3×10^{-4}	DIW	427
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

Table 3.8. Averaged Interval D_e Values (cm^2/s) for Tc-Gluconate Suite and Related Archive and Screening Test Monoliths

	Leachant = VZP							Leachant = DIW					
	D_e 7d to 427d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	σ		D_e 7d to 427d Avg.	σ	D_e 28d to 63d Avg.	σ	D_e 28d to 427d Avg.	σ
	Tc							**Tc**					
T4 (Tc-Gluconate)	2.39E-11	9.28E-12	2.77E-11	9.55E-12	2.64E-11	8.82E-12	T4 (Tc-Gluconate)	2.01E-10	6.11E-11	2.56E-10	2.73E-11	2.12E-10	6.48E-11
T5 (Pertech)	1.33E-12	3.74E-13	1.30E-12	2.16E-13	1.30E-12	3.50E-13	T5 (Pertech)	2.28E-11	7.67E-12	2.66E-11	5.20E-12	2.27E-11	7.43E-12
Archive T3HCS2-7.8AVG	2.57E-12	6.19E-13	2.70E-12	5.94E-13	2.61E-12	6.67E-13	Archive T3HCS2-7.8AVG	7.93E-11	7.27E-11	6.31E-11	3.10E-11	9.47E-11	7.61E-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)	3.67E-09	2.43E-09	3.08E-09	5.21E-10	2.58E-09	8.10E-10	T4 (Tc-Gluconate)	4.37E-09	2.15E-09	3.72E-09	3.91E-10	3.38E-09	8.01E-10
T5 (Pertech)	2.93E-09	2.38E-09	2.25E-09	5.19E-10	1.81E-09	6.92E-10	T5 (Pertech)	3.81E-09	2.29E-09	3.06E-09	3.84E-10	2.80E-09	8.20E-10
Archive T3HCS2-7.8AVG	4.38E-09	1.82E-09	4.37E-09	4.96E-10	3.60E-09	1.04E-09	Archive T3HCS2-7.8AVG	5.84E-09	2.05E-09	5.52E-09	5.89E-10	5.18E-09	1.76E-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)	1.79E-09	1.08E-09	1.60E-09	1.47E-10	1.34E-09	4.29E-10	T4 (Tc-Gluconate)	2.37E-09	8.84E-10	2.25E-09	9.79E-11	1.98E-09	3.90E-10
T5 (Pertech)	1.44E-09	9.90E-10	1.24E-09	1.53E-10	1.01E-09	3.38E-10	T5 (Pertech)	2.16E-09	7.72E-10	2.00E-09	2.28E-10	1.82E-09	3.12E-10
Archive T3HCS2-7.8AVG	4.82E-09	3.53E-09	4.03E-09	3.44E-10	4.34E-09	3.87E-09	Archive T3HCS2-7.8AVG	5.31E-09	1.40E-09	5.39E-09	3.63E-10	4.81E-09	1.03E-09

	Leachant = VZP						Leachant = DIW						
	D_e 7d to 427d Avg.	**σ**	**D_e 28d to 63d Avg.**	**σ**	**D_e 28d to 427d Avg.**	**σ**		**D_e 7d to 427d Avg.**	**σ**	**D_e 28d to 63d Avg.**	**σ**	**D_e 28d to 427d 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)	3.28E-09	1.99E-09	2.56E-09	2.31E-10	2.39E-09	4.10E-10	T4 (Tc-Gluconate)	4.27E-09	1.81E-09	3.57E-09	2.15E-10	3.43E-09	3.76E-10
T5 (Pertech)	2.49E-09	1.72E-09	1.86E-09	3.19E-10	1.70E-09	3.31E-10	T5 (Pertech)	3.72E-09	1.40E-09	3.07E-09	4.15E-10	3.11E-09	3.97E-10
Archive T3HCS2-7.8AVG	4.90E-09	1.97E-09	4.62E-09	7.49E-10	4.06E-09	9.39E-10	Archive T3HCS2-7.8AVG	6.70E-09	1.51E-09	6.32E-09	1.29E-09	6.14E-09	1.02E-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)	*1.59E-15*	2.38E-15	*2.89E-15*	3.10E-15	*1.75E-15*	2.66E-15	T4 (Tc-Gluconate)	1.13E-13	6.63E-14	9.57E-14	2.59E-14	8.57E-14	4.67E-14
T5 (Pertech)	*3.57E-15*	7.20E-15	*3.01E-15*	3.22E-15	*1.86E-15*	2.74E-15	T5 (Pertech)	8.41E-14	5.11E-14	6.73E-14	2.07E-14	6.47E-14	3.97E-14
Archive T3HCS2-7.8AVG	*1.66E-15*	2.40E-15	*2.95E-15*	3.16E-15	*1.80E-15*	2.71E-15	Archive T3HCS2-7.8AVG	3.25E-13	7.52E-13	7.98E-14	2.62E-14	3.93E-13	8.54E-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

Italicized numbers for Cr interval D_e values included leachates where Cr concentrations were less than detection values.

Note that none of the interval averaged D_e values have been corrected for inventory depletion.

Figure 3.14 shows photographs of the T4 (containing Tc-gluconate) and T5 monoliths (containing pertechnetate) after 361 days of leaching in either VZP or DIW. 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 3.15 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 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.

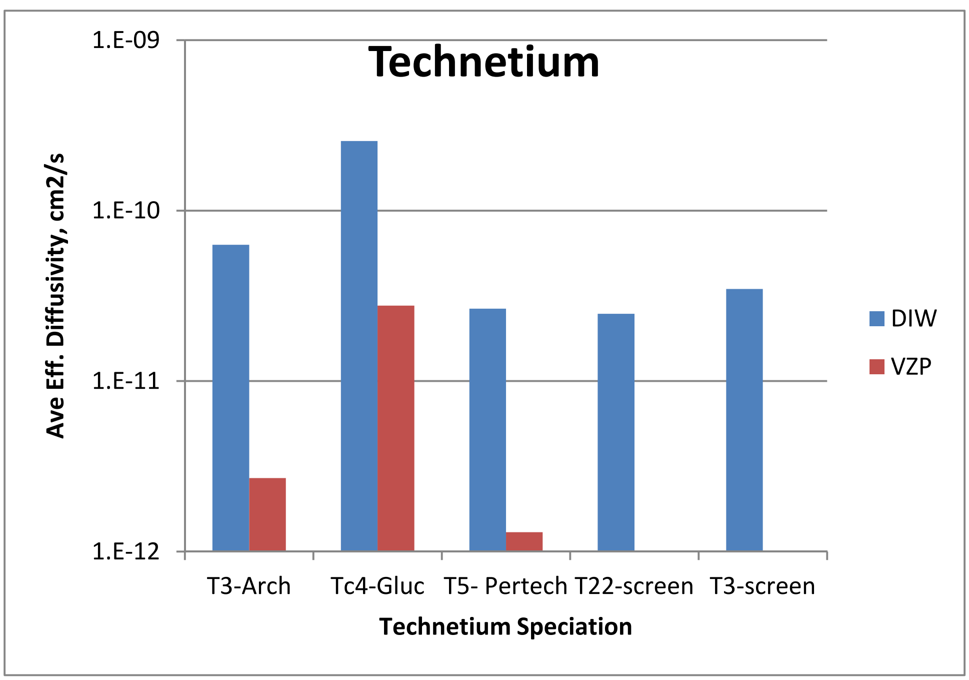


Figure 3.12. Averaged Interval 28-d to 63-d D_e Values for Tc from Monoliths Leached in DIW vs. VZP

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 effective diffusion coefficient calculations based on Equation 1 are no longer accurate. Thus, we have been recommending the use of the averaged interval D_e 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 408 days of leaching. Table 3.9 shows the cumulative percentage leached for these two constituents after 408 days of testing. Of note for Tc is the difference in % released between VZP and DIW leachants and the increased release % between the T4 (Tc-gluconate) and T5 (pertechnetate) monoliths. The latter again suggests that there may be a demonstrable difference in Tc leach tendencies dependent on Tc speciation.

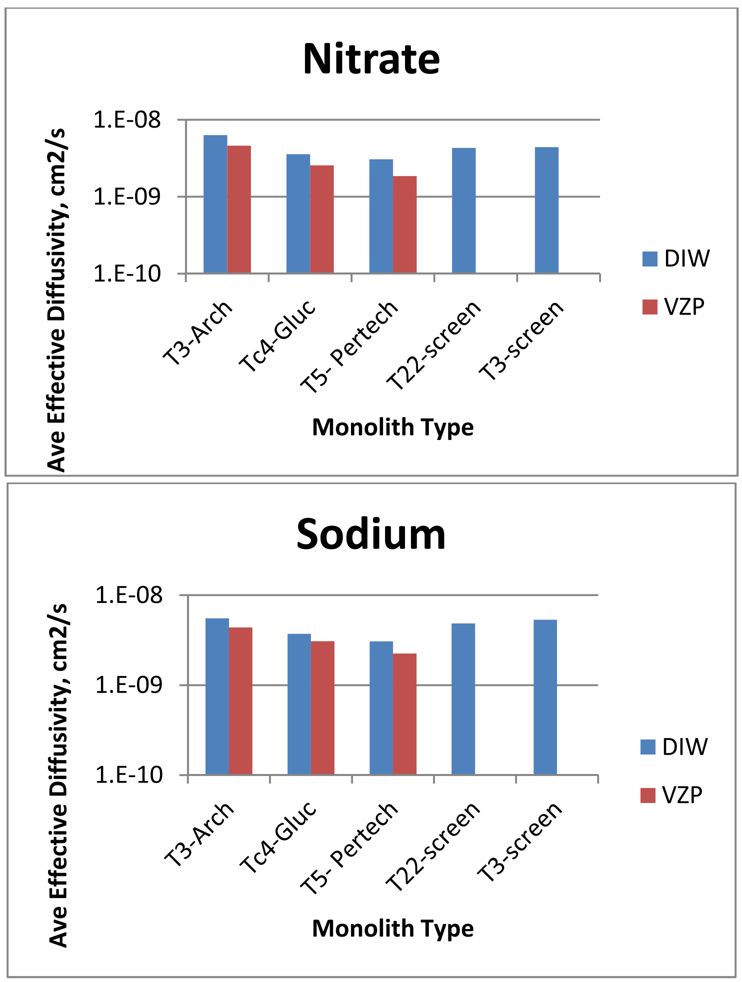


Figure 3.13. Averaged Interval 28-d to 63-d D_e Values for Nitrate and Sodium from Tc-Monoliths Leached in DIW vs. VZP

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

Figure 3.14. Photographs of Tc-Gluconate and Pertechnetate Monoliths Leached in Two Leachants for 361 Days

Table 3.9. Tc and Cr Cumulative Fraction (%) Leached After 408 Days Leaching for T4 and T5 Monoliths

Leachant	T4 (Tc)	T5 (Tc)	T4(Cr)	T5 (Cr)
VZP	<4	<1	<0.2	<0.06
DIW	~9	3	~0.2	0.15

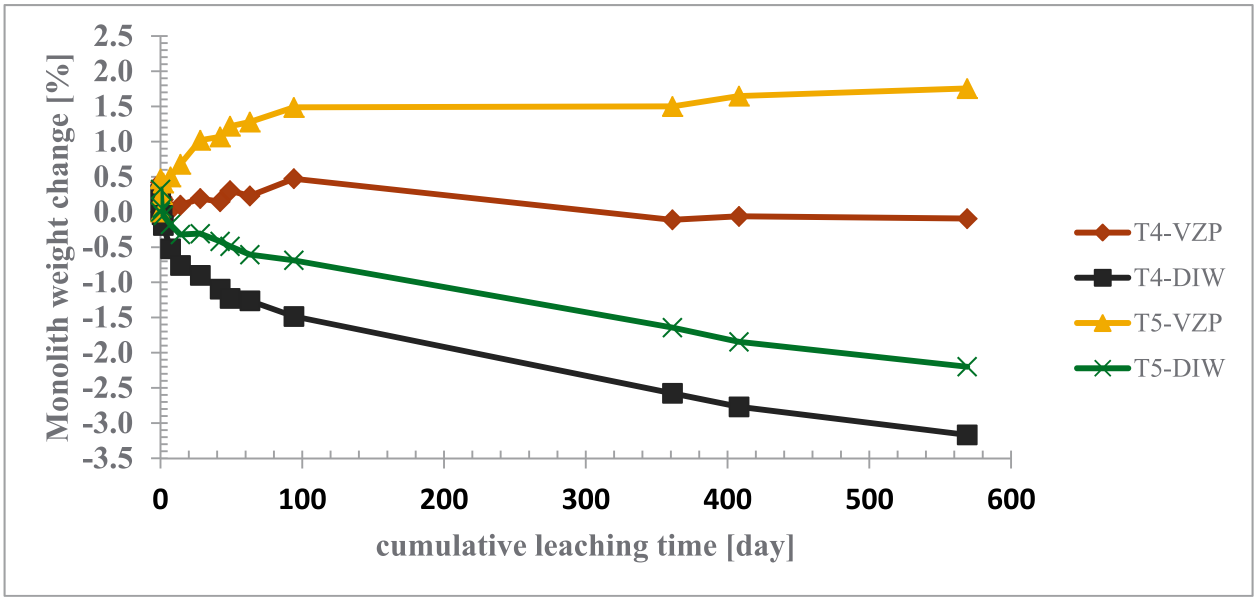


Figure 3.15. Change in Monolith Weights as a Function of Time and Leachant Used
(Data points are average weight change for two monoliths leached under designated conditions.)

4.0 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.

4.1 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 effective diffusivities and almost an order of magnitude range in effective 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 two years follow. One universal observation from these extended leach studies on LAW Cast Stone monoliths, when leached in VZP, 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 leachant'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 20 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. However, the most recent DIW leach results for a few monoliths are showing signs of an increased rate of release that might be caused by the formation of new surface micro-cracks visually observed after 427 to 790 days of leaching (see Figure 3.6 for example). These cracks are not covered by white precipitate, suggesting that they are newly formed. It is possible this observation is the first signs of some internal degradation of Cast Stone monoliths after about 427 to 790 days of leaching in DIW. For monoliths leached in VZP no "late stage" increases in leach rates have been observed; however, because of the significant coating of secondary precipitates on the VZP-contacted monoliths their surfaces cannot be readily observed visually to evaluate whether there are new micro-cracks developing.

For all the monoliths studied herein, tables of interval averaged D_e 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 leachant is used. The same range of interval averaged D_e 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_e values for VZP leachants are generally not quantifiable because leachate 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 leachates as well as DIW leachates. The average 28 to 63 day interval D_e 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_e) 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 leachant, 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 790 d in DIW only, Cast Stone Mixes 14 and 18 are the best performing mix for restricting the release of ^{99}Tc, and Mixes 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 monoliths from these same mixes were leached for up to 427 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.

Regardless of the Cast Stone mix composition, the D_e values for the anions iodide, nitrate, and nitrite and the cation sodium are very similar (as mentioned the range in averaged interval D_e values for these four constituents varies by only a factor of three to six). Cr leaches the slowest especially when VZP leachant 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 leachant generally from 49 to 63 days of leaching, we are recommending that the VZP leached averaged 28 to 63 day interval D_e 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. Thus, for future IDF PA predictive modeling, the highlighted values for the 28- to 63-day averaged interval D_e values for the VZP leachant are recommended as the most probable values, if an empirical diffusivity conceptual model is chosen for the release from LAW Cast Stone. The D_e values for the COCs from earlier leaching periods (0.08 to 14 days) and the D_e values for tests that used DIW as the leachant could be used to create a range of D_e 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_e values; often an order of magnitude and in a few cases up to two orders of magnitude higher than interval D_e 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_e 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. Regardless, if there is a need to select maximum (worst case) D_e values to obtain a range to perform sensitivity analyses, the early leaching data and D_e values obtained in DIW found in Appendix B could be used to select the range of D_e values. Recall that 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 ongoing work that has solidified liquid secondary waste simulants in a different dry blend mix. It is the solidified secondary waste simulants that will be addressed in the 2017 IDF PA.

When the cumulative mass of a constituent released from the monoliths starts to exceed 20% of their starting inventories, Equation 1 used to calculate the effective diffusion coefficient is not accurate because the leaching conditions no longer satisfy the assumed semi-infinite boundary condition. Based on Equation 1, 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 4.1 shows an example plot for monolith T5HCS-7.8 AVG-2 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. 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 Equation 1. 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 reoxidation 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. 2012; 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 $\sim 10^{-3}$ cm^2/s while the diffusion coefficients of dissolved species in water are $\sim 10^{-5}$ cm^2/s.

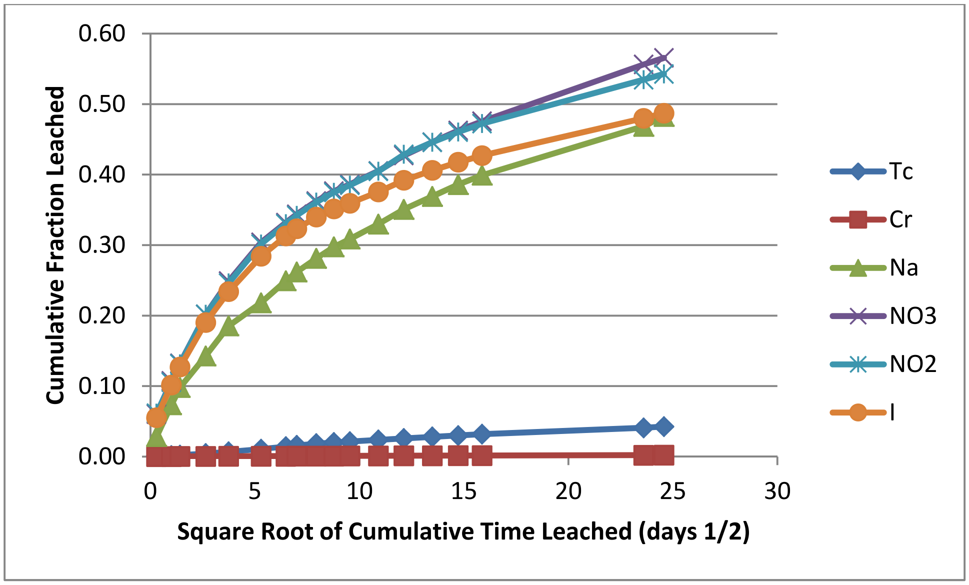


Figure 4.1. T5HCS-7.8 AVG-2 Leached in DIW for 609 d

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 4.1. There are other mathematical relationships available in the literature (Thomas 1987; Pescatore 2010, 2011) that could be used to calculate effective diffusion coefficients (D_e values) for experiments where inventory depletion is occurring (such as ours). If more accurate D_e 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 Cast Stone and ETF-processed secondary waste forms destined for burial at IDF.

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.

[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.

The iodide leach results from 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. 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 leachates 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. 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 monoliths from the Archive and Extended Leach Suites in both VZP and DIW leachants. Whether the Tc in leachates from the T4 monoliths remains in a non-pertechnetate form has not been determined because the concentration of total Tc in the T4 monolith leachates 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 3.8 visually shown in Figure 3.12). 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

[1] Using the initial pertechnetate loading in Cast Stone monoliths (dry weight basis) shown in Table 3.7 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 ~ 1.0×10^{-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 diffuses out of the porous matrix.

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. However, the D_e 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_e 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 leachates 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 100% relative humidity) show remarkably good precision in leach tendencies for each constituent that we have been tracking. Figure 4.2 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_e 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 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.

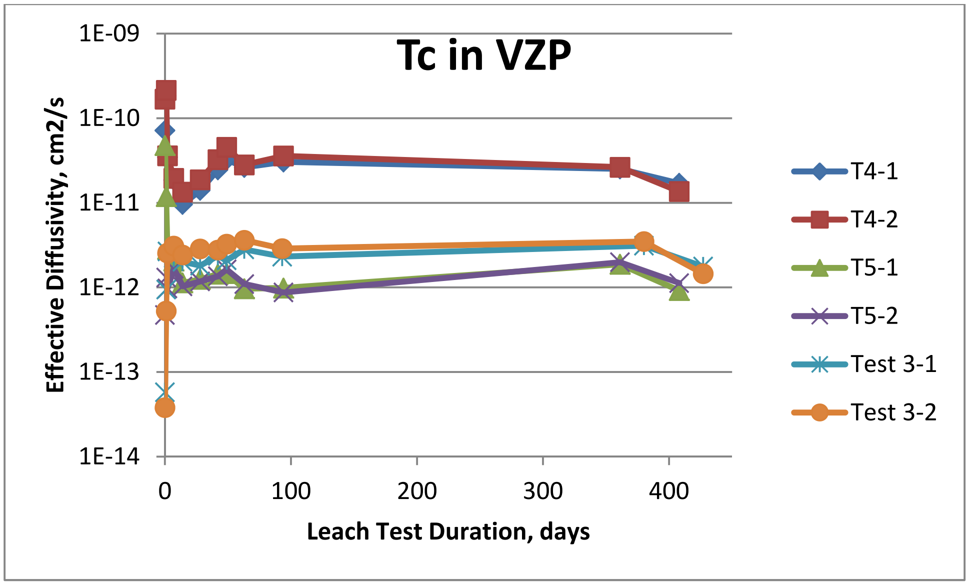


Figure 4.2. Shows Excellent Precision in Leach Tendencies for Duplicate Monoliths from the Same Batch

4.2 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 effective 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 and Wilmarth 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 4.3) 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, effective 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.

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.

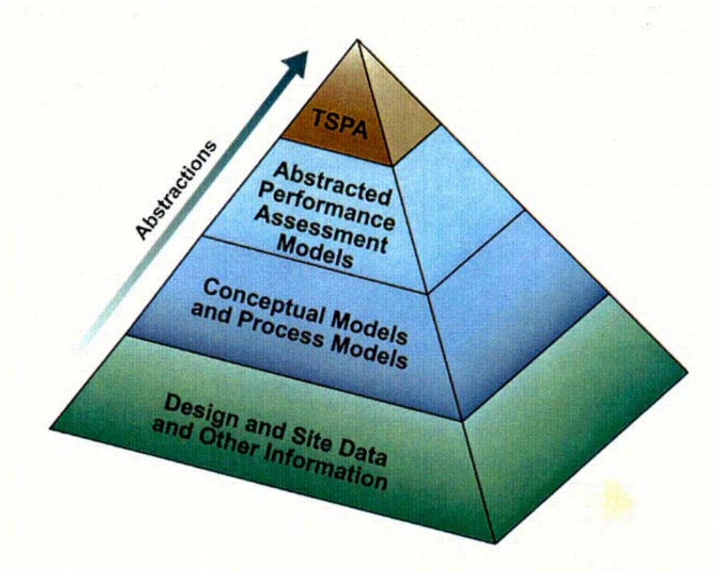


Figure 4.3. Total System Performance Assessment Information Pyramid (from TRW 2000)

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. (2013) 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.

4.3 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. Reoxidation 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 leachates 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 leachates that have contacted the solid waste form (e.g., all the EPA Method 1315 leachates 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, N2 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,

- 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 2012; Langton 2013) 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. 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 reoxidation 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.

4.4 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 4.4 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 codes/simulators available from the Cementitious Barriers Partnership (CBP). Other codes/simulators can be substituted for those shown without changing the logic. For example, 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 as are the CBP codes listed in Figure 4.4. However, after adding cement-specific algorithms, input parameters for the algorithms, and appropriate boundary conditions, eSTOMP has recently been applied to predicting 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 (Perko et al. 2015). More detailed descriptions of eSTOMP attributes are found in Yabusaki et al. (2015) and references therein.

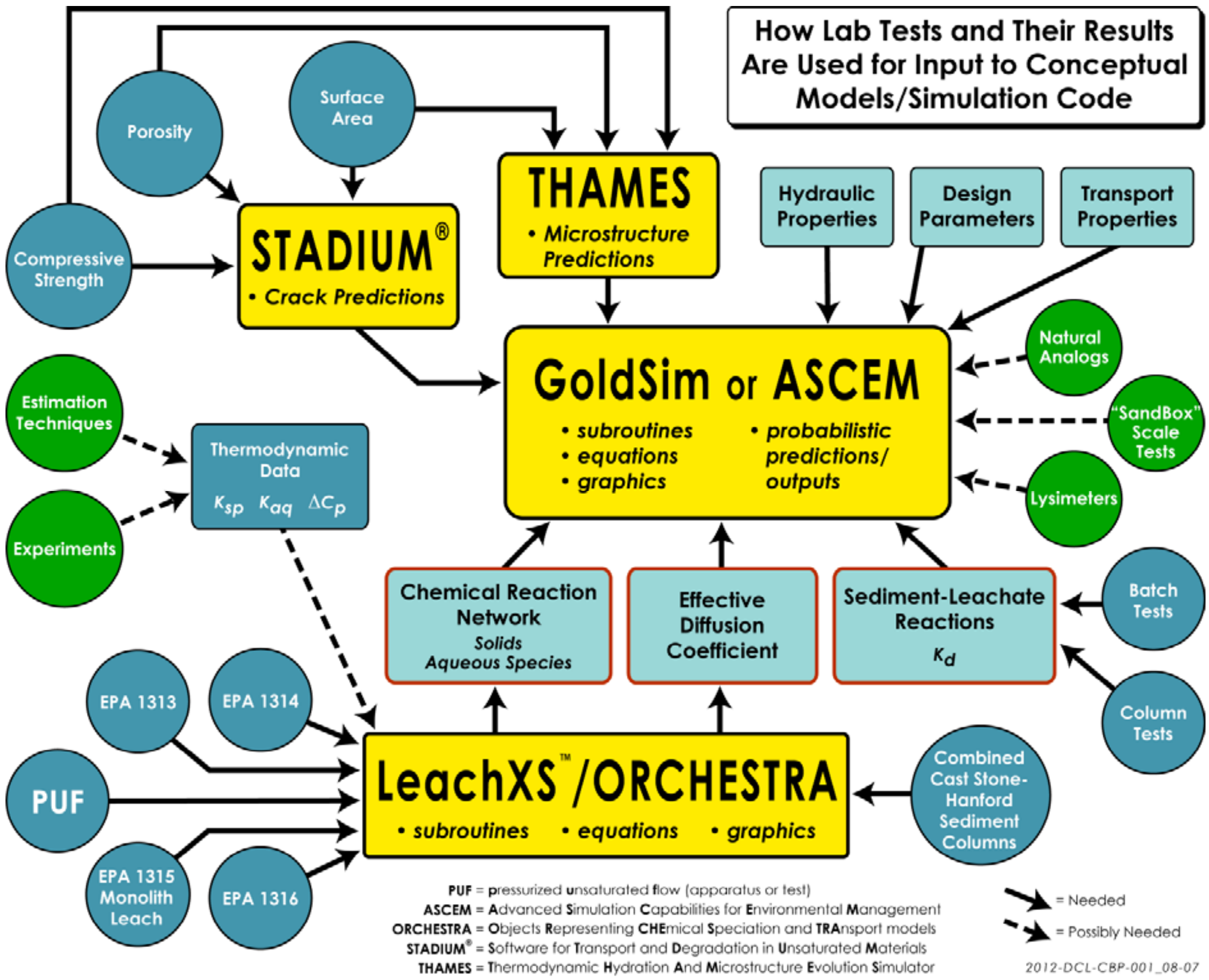


Figure 4.4. Flow/Logic Diagram Showing How Data from Test Methods/Experiments Feed Computer Codes

In this subsection, we will focus our discussion on the blue and green circles in Figure 4.4 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 effective 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 leachant 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 leachant to maintain constant atmospheric concentrations of oxygen and carbon dioxide in the leachates. As long as this air flow-through approach does not cause leachate 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 leachant 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 leachants will soon be 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 leachate 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 reoxidation 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 reoxidation 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 effective diffusion coefficient (D_e) for contaminants such as Tc(VII) and iodide leaching from spiked concrete into unsaturated Hanford sediments are several orders of magnitude lower than D_e 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 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 effective diffusion coefficients, D_e. 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 leachates. Interactions include adsorption of trace constituents in the leachates 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 4.4 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 4.3) 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

[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).

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 leachants, 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 4.5 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.

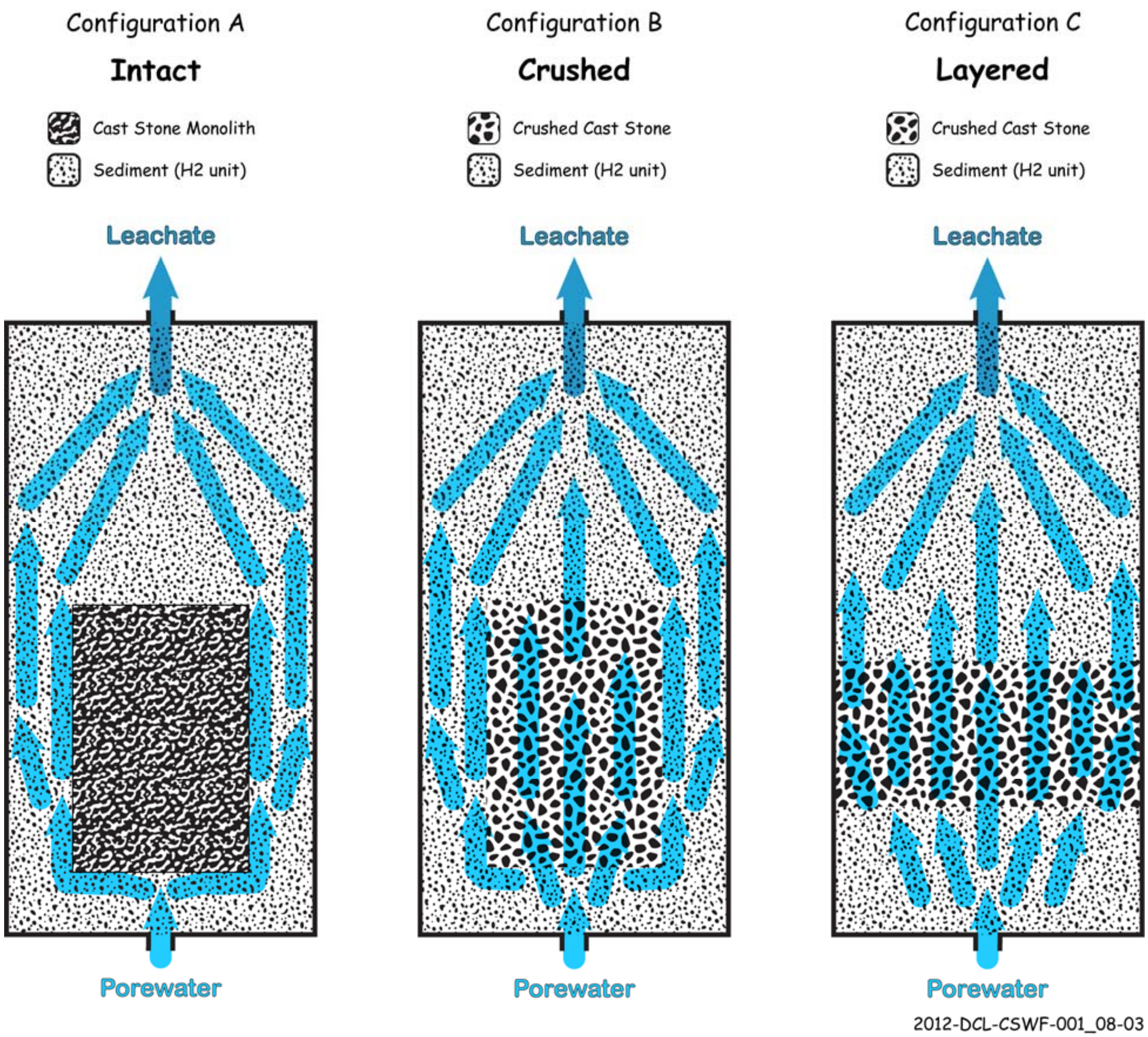


Figure 4.5. Schematic of Multi-Component Flow-Through Column Tests

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 4.4 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, such as the LeachXS/ORCHESTRA speciation code in this example. 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), and Almond et al. (2012a). 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 4.4 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)) currently being performed at PNNL under funding from WRPS.

Finally on the right-hand side of Figure 4.4 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.

5.0 References

Almond PM, DI Kaplan, CA Langton, DB Stefanko, WA Spencer, A Hatfield and Y Arai. 2012a. *Method Evaluation and Field Sample Measurements for the Rate of Movement of the Oxidation Front in Saltstone*. SRNL-STI-2012-00468 Rev 0, Savannah River National Laboratory, Aiken, SC.

Almond PM, DI Kaplan, and EP Shine. 2012b. *Variability of Kd Values in Cementitious Materials and Sediments*. SRNL-STI-2011-00672, Savannah River National Laboratory, Aiken, SC.

ANSI/ANS—American National Standards Institute/American Nuclear Society. 2003. *Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure.* ANSI/ANS-16.1, La Grange Park, Illinois.

ASTM. 2008b. *Standard Test Methods for Determination of the Soil Water Characteristic Curve for Desorption Using Hanging Column, Pressure Extractor, Chilled Mirror Hygrometer, or Centrifuge.* ASTM D 6836-02 (Reapproved 2008), ASTM International, West Conshohocken, Pennsylvania.

ASTM. 2008a. *Standard Test Method for Accelerated Leach Test for Diffusive Releases from Solidified Waste and a Computer Program to Model Diffusive, Fractional Leaching from Cylindrical Waste Forms.* ASTM C 1308-08, ASTM International, West Conshohocken, PA.

Atkinson A, K Nelson, and TM Valentine. 1986. "Leach Test Characterization of Cement-Based Nuclear Waste Forms." *Nucl. & Chem. Waste Manage.*6:241-253.

Atkinson A and AK Nickerson. 1988. "Diffusion and Sorption of Cesium, Strontium, and Iodine.

in Water-Saturated Cement." *Nuc. Tech.* 81(1):100-113.

Brown CF, WR Wilmarth, JH Westsik, Jr, RJ Serne, DJ McCabe, CA Nash, BM Rapko, and AD Cozzi. 2014. *Preliminary Assessment of the Low Temperature Waste Form Technology Coupled with Technetium Removal.* PNNL-22103 Rev. 1 and SRNL-STI-2013-0002 Rev. 1, Pacific Northwest National Laboratory, Richland, WA and Savannah River National Laboratory, Aiken, SC.

Cantrell KJ and BD Williams. 2012. *Equilibrium Solubility Model for Technetium Release from Saltstone Based on Anoxic Single-Pass Flow through Experiments*. PNNL-21723, Pacific Northwest National Laboratory, Richland, WA.

Cantrell KJ and BD Williams. 2013. "Solubility Control of Technetium Release from Saltstone by $Tc0_2 \bullet xH_2O$." *Journal of Nuclear Materials* 437(1-3):424-431. doi:10.1016/j.jnucmat.2013.02.049.

Chang H-S, W Um, K Rod, RJ Serne, A Thompson, N Perdrial, CI Steefel, and J Chorover. 2011. "Strontium and Cesium Release Mechanisms during Unsaturated Flow through Waste Weathered Hanford Sediments." *Environmental Science and Technology,* 45(19), 8313-8320.

Crank J. 1986. Mathematics of Diffusion. Oxford University Press, London.

DOE—U.S. Department of Energy. 2012. *Final Tank Closure and Waste Management Environmental Impact Statement for the Hanford Site, Richland, Washington*. DOE/EIS-0391, U.S. Department of Energy, Washington, D.C.

EPA—U.S. Environmental Protection Agency. 2012. *Method 1313 – Liquid-Solid Partitioning as a Function of Extract pH using a Parallel Batch Extraction Procedure*, Rev. 0.

EPA—U.S. Environmental Protection Agency. 2013. *Method 1315 – Mass Transfer Rates of Constituents in Monolithic or Compacted Granular Materials Using a Semi-dynamic Tank Leaching Procedure,* Rev. 0.

Estes SL, DI Kaplan, and BA Powell. 2012. *Technetium Sorption by Cementitious Materials Under Reducing Conditions.* SRNL-STI-2012-00596, Savannah River National Laboratory, Aiken, SC.

Estes SL, Y Arai, DI Kaplan, and BA Powell. 2013. *Technetium Sorption by Cementitious Materials Under Reducing Conditions.* SRNL-STI-2011-00716, Revision 1, Savannah River National Laboratory, Aiken, SC.

Golovich EC, SV Mattigod, MMV Snyder, L Powers, GA Whyatt, and DM Wellman. 2014. *Radionuclide Migration through Sediment and Concrete: 16 Years of Investigations*. PNNL-23841, Pacific Northwest National Laboratory, Richland, WA.
Full Publication (pdf).

Grathwohl P. Diffusion in Natural Porous Media: Contaminant Transport, Sorption/desorption and Dissolution Kinetics. Kluwer Academic Publishers, 1998, ISBN 0-7923-8102-5.

Horton R, PJ Wierenga, and DR Nielsen. 1982. "A Rapid Technique for Obtaining Uniform Water Content Distributions in Unsaturated Soil Columns." *Soil Sci.*, 133:397-399.

Kaplan DI, MS Lilley, PM Almond, and BA Powell. 2011. *Long-term Technetium Interactions with Reducing Cementitious Materials.* SRNL-STL-2010-00668, Savannah River National Laboratory, Aiken, SC.

Kaplan DI and L Dien. 2013. *Solubility of Technetium Dioxides (TcO2-c,TcO2●1.6H2O and TcO2 ●2H2O) in Reducing Cementitious Material Leachates: A Thermodynamic Calculation*. SRNL-STI-2012-00769, Rev. 1, Savannah River Site, Aiken, SC.

Langton CA. 2014. *Technetium Oxidation in Slag-based Sodium Salt Waste Forms Exposed to Water and Moist Hanford Soil*. SRNL-STI-2014-00399 Rev 0. Savannah River National Laboratory, Aiken, SC.

Langton CA and PM Almond. 2013. *Cast Stone Oxidation Front Evaluation: Preliminary Results for Samples Exposed to Moist Air*. SRNL-STI-2013-00541, Revision 0, Savannah River National Laboratory, Aiken, South Carolina.

Levitskaia TG, BM Rapko, A Anderson, JM Peterson, SD Chatterjee, ED Walter, HM Cho, and NM Washton. 2014. *Speciation and Oxidative Stability of Alkaline Soluble, Non-Pertechnetate Technetium.* PNNL-23654, Pacific Northwest National Laboratory, Richland, WA.

Li D and DI Kaplan. 2013. *Solubility of Technetium Dioxides (TcO2-c, TcO2·a.6 H2O and TcO2·2H2O) in Reducing Cementitious Material Leachates: A Thermodynamic Calculation.* SRNL-STI-2012-00769, Rev. 1, Savannah River National Laboratory, Aiken, SC.

Lockrem LL. 2005. *Cast Stone Technology for Treatment and Disposal of Iodine-Rich Caustic Waste Demonstration - Final Report*. RPP-RPT-26725, CH2M HILL Hanford Group, Inc., Richland, WA.

Mann FM, RJ Puigh, R Khaleel, S Finfrock, BP McGrail, DH Bacon, and RJ Serne. 2003. *Risk Assessment Supporting the Decision on the Initial Selection of Supplemental ILAW Technologies*. RPP-17675, Revision 0, CH2M Hill Hanford Group Inc., Richland, WA.

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

Mattigod SV, GA Whyatt, RJ Serne, PF Martin, KE Schwab, and MI Wood. 2001. *Diffusion and Leaching of Selected Radionuclides (Iodine-129, Technetium-99, and Uranium) Through Category 3 Waste Encasement Concrete and Soil Fill Material*. PNNL-13639, Pacific Northwest National Laboratory, Richland, WA.

Neeway JJ, N Qafoku, RJ Serne, AR Lawter, JR Stephenson, WW Lukens, and JH Westsik, Jr. 2014. *Evaluation of Technetium Getters to Improve the Performance of Cast Stone*. PNNL-23667, Pacific Northwest National Laboratory, Richland, WA.

Ojovan MI, GA Varlackova, ZI Golubeva, and ON Burlaka. 2011. "Long-term field and laboratory leaching tests of cemented radioactive wastes." *Journal of Hazardous Materials*, 187 (1-3), pp. 296-302.

Perko, J, KU Mayer, G Kosakowski, L De Windt, J Govaerts, D Jacques, D Su, and JCL Meeussen. 2015. "Decalcification of cracked cement structures." *Computational Geosciences* 19:673–693. DOI 10.1007/s10596-014-9467-2.

Pescatore C. 1990. "Improved Expressions of Modeling Diffusive, Fractional Cumulative Leaching from Finite Size Waste Forms." *Waste Management* 10:155–159.

Pescatore C. 1991. "Leach Rate Expressions for Performance Assessment of Solidified, Low-Level Radioactive Waste." *Waste Management* 11:223–229.

Pierce EM, BP McGrail, EA Rodriguez, HT Schaef, KP Saripalli, RJ Serne, KM Krupka, PF Martin, SR Baum, KN Geiszler, LR Reed, and WJ Shaw. 2004. *Waste Form Release Data Package for the 2005 Integrated Disposal Facility Performance Assessment*. PNNL-14805, Pacific Northwest National Laboratory, Richland, WA.

Qafoku N, JJ Neeway, AR Lawter, TG Levitskaia, RJ Serne, JH Westsik, Jr, and MMV Snyder. 2015. *Technetium and Iodine Getters to Improve Cast Stone Performance.* PNNL-23282 Rev. 1, Pacific Northwest National Laboratory, Richland, WA.

Rapko BM. 2014. *Protocol for Identifying the Presence of and Understanding the Nature of Soluble, Non-pertechnetate Technetium in Hanford Tank Supernatants.* PNNL-23180; EMSP-RPT-020, Pacific Northwest National Laboratory, Richland, WA.

Rapko BM, SA Bryan, JL Bryant, S Chatterjee, MK Edwards, JY Houchin, T Janik, TG Levitskaia, JM Peterson, RA Peterson, SI Sinkov, FN Smith, and RS Wittman. 2013a. *Development of a Chemistry-Based, Predictive Method for Determining the Amount of Non-Pertechnetate Technetium in the Hanford Tanks: FY 2012 Progress Report*. PNNL-22173, Pacific Northwest National Laboratory, Richland, WA.

Rapko BM, SA Bryan, S Chatterjee, MK Edwards, TG Levitskaia, JM Peterson, RA Peterson, and SI Sinkov. 2013b. *Investigations into the Nature of Alkaline Soluble, Non-Pertechnetate Technetium*. PNNL-22957, Pacific Northwest National Laboratory, Richland, WA.

Raymond RE, RW Powell, DW Hamilton, WA Kitchen, BM Mauss, and TM Brouns. 2004. "Initial Selection of Supplemental Treatment Technologies for Hanford's Low-Activity Tank Waste." in *WM'04 Conference*, Phoenix, AZ.

Rod KA, W Um, and M Flury. 2010. "Transport of strontium and cesium in simulated Hanford tank waste leachate through quartz sand under saturated and unsaturated flow." *Environmental Science and Technology* 44(21), 8089-8094.

Russell RL, JH Westsik, Jr, DJ Swanberg, RE Eibling, AD Cozzi, MJ Lindberg, GB Josephson, and DE Rinehart. 2013. L*etter Report: LAW Simulant Development for Cast Stone Screening Tests.* PNNL-22352, Pacific Northwest National Laboratory, Richland, WA.

Sarott F-A, MH Bradbury, P Pandolfo and P Spieler. 1992. "Diffusion and adsorption studies on hardened cement paste and the effect of carbonation on diffusion rates." *Cement and Concrete Research* 22: 439-444.

Serne RJ, BM Rapko and IL Pegg. 2014. *Technetium Inventory, Distribution, and Speciation in Hanford Tanks.* PNNL-23319, Rev. 1, Pacific Northwest National Laboratory, Richland, WA.

Serne RJ, WJ Martin, SB McLaurine, SP Airhart, VL LeGore, and RL Treat. 1987. *Laboratory Leach Tests of Phosphate/Sulfate Waste Grout and Leachate Adsorption Tests Using Hanford Sediment.* PNL-6019, Pacific Northwest Laboratory, Richland, WA.

Serne RJ and JH Westsik, Jr. 2011. Data Package for Secondary Waste Form Down-Selection—Cast Stone. PNNL-20706, Pacific Northwest National Laboratory, Richland, WA.

SRR Closure & Waste Disposal Authority (SRR CWDA). 2014. *FY2014 Special Analysis for the Saltstone Disposal Facility at the Savannah River Site*. SRR-CWDA-2014-00006, Revision 2, Savannah River Remediation, Aiken, SC.

Sun W, S Jayaraman, W Chen, KA Persson, and G Ce. 2015. "Nucleation of Metastable Aragoite $CaCO_3$ in Seawater." *Proc. Of National Academy of Sciences* 112(11):3199-3204.

Sundaram SK, J Chun, W Um, KE Parker, C-W Chung, JH Westsik Jr, ME 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, WA.

Thomas GF. 1987. "Diffusional Release of a Single Component Material from a Finite Cylindrical Waste Form." *Annals Nucl. Energy* 14(6):283-294.

TRW. 2000. *Total System Performance Assessment for the Site Recommendation*. TDR-WIS-PA-000001 REV 00 ICN 01, Las Vegas, NV.

Um W, H-S Chang, JP Icenhower, WW Lukens, RJ Serne, NP Qafoku, JH Westsik, Jr, EC Buck, and SC Smith. 2011b. "Immobilization of 99-Technetium (VII) by Fe(II)-Goethite and Limited Reoxidation." *Environ. Sci. Technol*. 45: 4904–4913.

Um W, HB Jung, G Wang, JH Westsik, Jr, and RA Peterson. 2013. *Characterization of Technetium Speciation in Cast Stone*. PNNL-22977, Pacific Northwest National Laboratory, Richland, WA.

Um W, RJ Serne, MM Valenta, KE Parker, C-W Chung, G Wang, JS Yang, KJ Cantrell, MH Engelhard, and JH Westsik, Jr. 2011a. *Radionuclide Retention Mechanisms in Secondary Waste-Form Testing: Phase II*. PNNL-20753, Pacific Northwest National Laboratory, Richland, WA.

van Brakel J and PM Heertjes. 1974. "Analysis of Diffusion in Macroporous Media in Terms of a Porosity, a Tortuosity, and a Constrictivity Factor." *Int. J. Heat and Mass Transfer* 17:1093-1103.

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. 2013a. *Supplemental Immobilization of Hanford Low-Activity Waste: Cast Stone Screening Tests*. PNNL-22747, Pacific Northwest National Laboratory, Richland, WA.

Westsik JH, Jr, RJ Serne, EM Pierce, AD Cozzi, C Chung, and DJ Swanberg. 2013b. *Supplemental Immobilization Cast Stone Technology Development and Waste Form Qualification Testing Plan*. PNNL-21823 Rev. 1, Pacific Northwest National Laboratory, Richland, WA. Full Publication (pdf).

Westsik JH Jr. and RJ Serne. 2012. *Secondary Waste Cast Stone Waste Form Qualification Testing Plan*. PNNL-21656 Rev. 1, Pacific Northwest National Laboratory, Richland, WA. Full Publication (pdf)

Wierenga PJ, MH Young, GW Gee, RG Hills, CT Kincaid, TJ Nicholson, and RE Cady. 1993. *Soil Characterization Methods for Unsaturated Low-Level Waste Sites*. PNL-8480, Pacific Northwest Laboratory, Richland, Washington.

Yabusaki SB, RJ Serne, ML Rockhold, G Wang, and JH Westsik, Jr. 2015. *Technical Approach for Determining Key Parameters Needed for Modeling the Performance of Cast Stone for the Integrated Disposal Facility Performance Assessment.* PNNL-24022. Pacific Northwest National Laboratory, Richland, WA.

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 leachates 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 leachant with its theoretical composition based on chemical reagents added.

A.1

	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 (blue) **Mix # -Replicates** (green)

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

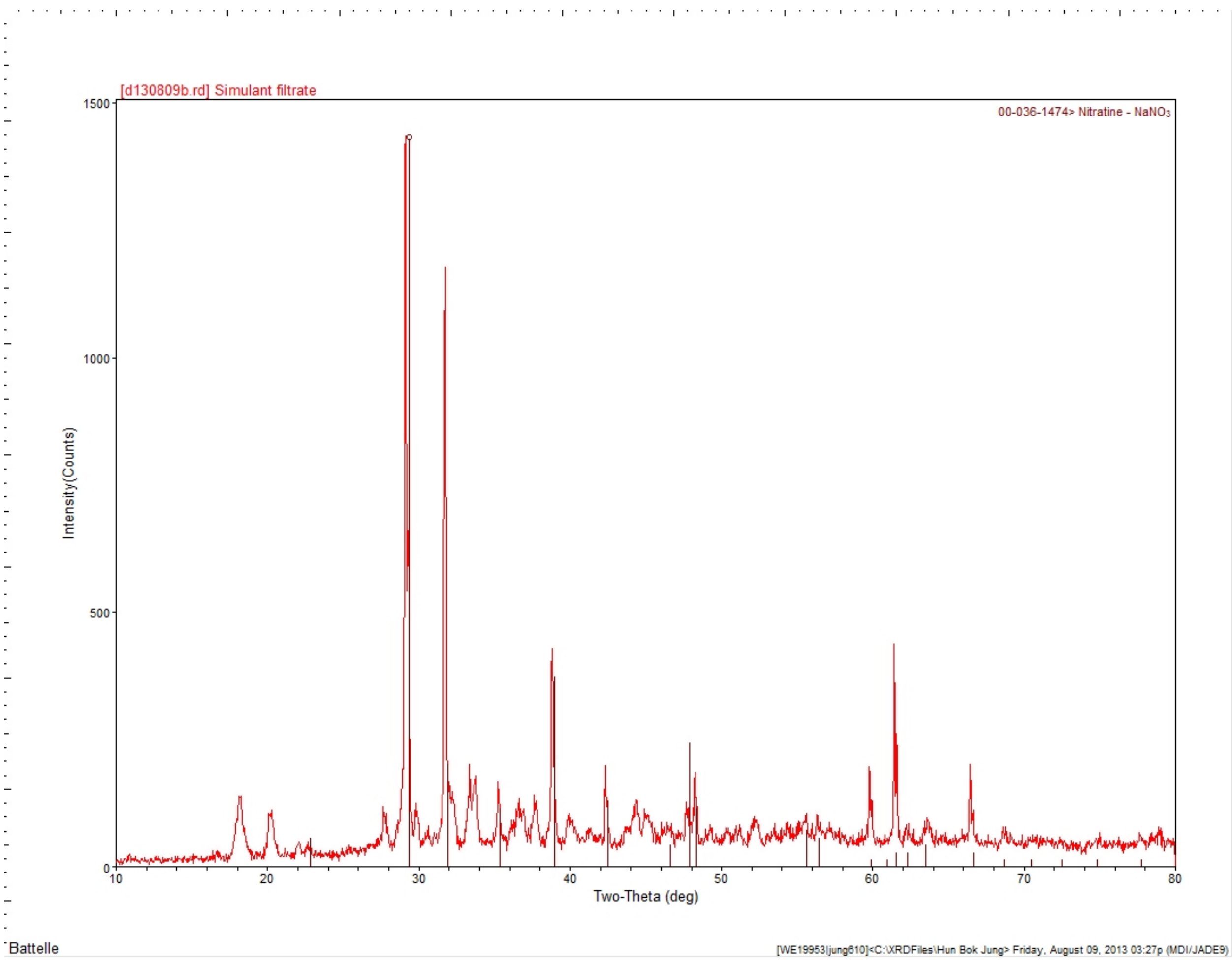


Figure A.2. XRD Spectra for Suspended Solids in 7.8 M Na Average Simulant-New Batch June 2013

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. 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
Effective Diffusion Coefficient, Fraction Leached, and Leachate pH and EC Worksheets

Appendix B

Effective Diffusion Coefficient, Fraction Leached, and Leachate pH and EC Worksheets

This appendix contains tables with the calculated effective diffusion coefficients, the fraction of each constituent's starting mass that has leached, the leachate pH and leachate 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 effective 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 100% relative humidity. The Archived Suite monoliths were cured/stored at room temperature and 100% 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 100% 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 2.5 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 leachants after the prolonged curing period (see Table 3.2 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 3.5 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 3.7 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 leachates be corrected for the leachant 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 leachant contains measureable concentrations of two of the COPCs for which effective 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 leachates 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 leachant 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 leachates. 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 leachant corrections were also made on the Cr leachate data for tests using VZP leachant.

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 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_e values. This is a "conservative" approach that leads to a slightly larger D_e calculated value, which is the protocol for use in performance assessment predictive modeling.

One oddity occurred in the batch of VZP leachant 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 leachant 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 leachate 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: http://www.epa.gov/epawaste/hazard/testmethods/sw846/new_meth.htm#1315.

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

Appendix B.1 includes the same D_e data for the first 91 days of leaching the selected monoliths for the Extended Suite as are found in Appendix C. We note in comparing the tables in Appendix B.1 and Appendix C, that there are small differences in the third digit for the reported effective diffusivities. We have checked the calculations and determined that the differences are due to the number of significant figures used for the monoliths' surface area, volume, and density in the calculations. The values reported in Appendix B.1 are based on calculations using six significant digits and the D_e values reported in Appendix C are based on calculations using three significant digits for the monoliths' surface area, volume, and density. Eleven monoliths from the original Screening Test were subjected to extended leaching in DIW leachant. The specific monoliths are listed in Table 2.5 in the main text.

Table B.1.1. Sodium Extended Effective Diffusion Coefficient (cm^2/s)

Sodium Effective 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

Table B.1.2. Nitrate Extended Effective Diffusion Coefficient (cm^2/s)

Nitrate Effective 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

Table B.1.3. Nitrite Extended Effective Diffusion Coefficient (cm^2/s)

Nitrite Effective 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

Table B.1.4. Iodine Extended Effective Diffusion Coefficient (cm^2/s)

Iodine Effective 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

Table B.1.5. Technetium Extended Effective Diffusion Coefficient (cm^2/s)

Technetium Effective 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

Table B.1.6. Chromium Extended Effective Diffusion Coefficient (cm^2/s)

Chromium Effective 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

Table B.1.7. 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

Table B.1.8. 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

Table B.1.9. 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

Table B.1.10. 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

Table B.1.11. 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

Table B.1.12. 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

Table B.1.13. pH of Leachates 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

Table B.1.14. Electrical Conductivity of Leachates 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

B.2 Archive Leach Tests

Table B.2.1. Sodium Archive Test Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Sodium Effective 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
Leachant		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

Interval (days)	Total Duration (days)	Sodium Effective 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
Leachant		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

Table B.2.1. Sodium Archive Test Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Sodium Effective 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
Leachant		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

Interval (days)	Total Duration (days)	Sodium Effective 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
Leachant		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

Table B.2.2. Nitrate Archive Test Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Nitrate Effective 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
Leachant		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

Interval (days)	Total Duration (days)	Nitrate Effective 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
Leachant		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

Table B.2.2. Nitrate Archive Test Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrate Effective 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
Leachant		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

Interval (days)	Total Duration (days)	Nitrate Effective 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
Leachant		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

Table B.2.3. Nitrite Archive Test Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Nitrite Effective 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
Leachant		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

Interval (days)	Total Duration (days)	Nitrite Effective 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
Leachant		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

Table B.2.3. Nitrite Archive Test Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrite Effective 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**
Leachant		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

Interval (days)	Total Duration (days)	Nitrite Effective 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**
Leachant		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

Table B.2.4. Iodine Archive Test Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Iodine Effective 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
Leachant		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

Interval (days)	Total Duration (days)	Iodine Effective 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
Leachant		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

Table B.2.4. Iodine Archive Test Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Iodine Effective 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**
Leachant		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

Interval (days)	Total Duration (days)	Iodine Effective 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**
Leachant		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

Table B.2.5. Technetium Archive Test Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Technetium Effective 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
Leachant		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

Interval (days)	Total Duration (days)	Technetium Effective 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
Leachant		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

Table B.2.5. Technetium Archive Test Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Technetium Effective 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
Leachant		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

Interval (days)	Total Duration (days)	Technetium Effective 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
Leachant		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

Table B.2.6. Chromium Archive Test Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Chromium Effective 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**
Leachant		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

Interval (days)	Total Duration (days)	Chromium Effective 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**
Leachant		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

Table B.2.6. Chromium Archive Test Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm²/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**
Leachant		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

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm²/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**
Leachant		VZP	VZP	DIW	VZP	VZP	DIW	VZP	VZP	DIW
8.11E-15	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

Table B.2.7. 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
Leachant		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

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
Leachant		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

Table B.2.7. Fraction of Sodium Leached versus Total Leach Time Archive Monoliths (contd)

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
Leachant		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

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
Leachant		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

Table B.2.8. 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
Leachant		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

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
Leachant		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

Table B.2.8. Fraction of Nitrate Leached versus Total Leach Time Archive Monoliths (contd)

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
Leachant		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

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
Leachant		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

Table B.2.9. 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
Leachant		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

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
Leachant		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

Table B.2.9. Fraction of Nitrite Leached versus Total Leach Time Archive Monoliths (contd)

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
Leachant		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

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
Leachant		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

Table B.2.10. 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
Leachant		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

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
Leachant		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

Table B.2.10. Fraction of Iodide Leached versus Total Leach Time Archive Monoliths (contd)

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
Leachant		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

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
Leachant		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

Table B.2.11. 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
Leachant		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

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
Leachant		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

Table B.2.11. Fraction of Technetium Leached versus Total Leach Time Archive Monoliths (contd)

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
Leachant		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

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
Leachant		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

Table B.2.12. 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
Leachant		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

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
Leachant		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

Table B.2.12. Fraction of Chromium Leached versus Total Leach Time Archive Monoliths (contd)

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**
Leachant		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

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**
Leachant		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

Table B.2.13. pH of Archive Leachates

Interval (days)	Total Duration (days)	pH of Archive Leachates (unitless) Mix Number and VZP Duplicates (a,b)								
		3Arch-a	3Arch-b	3Arch-c	5Arch-a	---	5Arch-c	8Arch-a	8Arch-b	8Arch-c
Leachant		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

Interval (days)	Total Duration (days)	pH of Archive Leachates (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
Leachant		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

Table B.2.13. pH of Archive Leachates (contd)

Interval (days)	Total Duration (days)	pH of Archive Leachates (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
Leachant		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

Interval (days)	Total Duration (days)	pH of Archive Leachates (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
Leachant		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

Table B.2.14. Electrical Conductivity of Archive Leachates

Interval (days)	Total Duration (days)	EC of Archive Leachates (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
Leachant		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

Interval (days)	Total Duration (days)	EC of Archive Leachates (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
Leachant		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

Table B.2.14. Electrical Conductivity of Archive Leachates (contd)

Interval (days)	Total Duration (days)	EC of Archive Leachates (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**
Leachant		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

Interval (days)	Total Duration (days)	EC of Archive Leachates (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**
Leachant		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

B.3 I-Spike Leach Tests

Table B.3.1. Na Effective Diffusion Coefficient (cm^2/s) I-spike Leach Tests

		Na in I-spiked Monoliths Effective Diffusion Coefficient (cm^2/s)										
I Conc	Low	Low	Med	Med	High	High	Low	Low	Med	Med	High	High
Leachant	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

Table B.3.2. Nitrate Effective Diffusion Coefficient (cm^2/s) I-spike Leach Tests

		Nitrate in I-spiked Monoliths Effective Diffusion Coefficient (cm^2/s)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Leachant	**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

Table B.3.3. Nitrite Effective Diffusion Coefficient (cm²/s) I-spike Leach Tests

		Nitrite in I-spiked Monoliths Effective Diffusion Coefficient (cm²/s)											
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Leachant	**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

Table B.3.4. Iodide in Iodide-spiked Monoliths Effective Diffusion Coefficient (cm^2/s)

		Iodide in I-spiked Monoliths Effective Diffusion Coefficient (cm^2/s)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Leachant	**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

Table B.3.5. 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
Leachant	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

Table B.3.6. 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**
Leachant	**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

Table B.3.7. 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
Leachant	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

Table B.3.8. 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
Leachant	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

Table B.3.9. pH of Leachates from I-spike Leach Tests

		pH in Leachates from I-spiked Monoliths (unitless)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Leachant	**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

Table B.3.10. Electrical Conductivity in Leachates from Iodide in Iodide-spiked Monoliths

		EC in I-spiked Monoliths Leachates (mS/cm)										
I Conc	**Low**	**Low**	**Med**	**Med**	**High**	**High**	**Low**	**Low**	**Med**	**Med**	**High**	**High**
Leachant	**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

B.4 Tc-Gluconate Leach Tests

Table B.4.1. Sodium Effective Diffusion Coefficient (cm^2/s) in Tc-Gluconate Leach Tests

Sodium Effective Diffusion Coefficient (cm^2/s)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Leachant	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

Table B.4.2. Nitrate Effective Diffusion Coefficient (cm^2/s) in Tc-Gluconate Leach Tests

Nitrate Effective Diffusion Coefficient (cm^2/s)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Leachant	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

Table B.4.3. Nitrite Technetium Effective Diffusion Coefficient (cm^2/s) in Tc-Gluconate Leach Tests

Nitrite Effective Diffusion Coefficient (cm^2/s)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Leachant	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

Table B.4.4. Technetium Effective Diffusion Coefficient (cm^2/s) Tc-Gluconate Leach Tests

Technetium Effective Diffusion Coefficient (cm^2/s)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Leachant	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

Table B.4.5. Chromium Effective Diffusion Coefficient (cm^2/s) in Tc-Gluconate Leach Tests

Chromium Effective Diffusion Coefficient (cm^2/s)										
Test Duration		**Tc Form**	**Tc-gluconate**				**Pertechnetate**			
Interval (days)	**Total (days)**	**Leachant**	**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

Table B.4.6. 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)	Leachant	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

Table B.4.7. 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)**	**Leachant**	**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

Table B.4.8. 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)	Leachant	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

Table B.4.9. 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)	Leachant	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		0.0008	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

Table B.4.10. Chromium Sodium 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)	Leachant	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		7.76E-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

Table B.4.11. pH in Leachates from Tc-Gluconate Leach Tests

pH in Leachates (unitless)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Leachant	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		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

Table B.4.12. EC in Leachates from Tc-Gluconate Leach Tests

Electrical Conductivity in Leachates (mS/cm)										
Test Duration		Tc Form	Tc-gluconate				Pertechnetate			
Interval (days)	Total (days)	Leachant	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		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

Appendix C
Corrections to Previously Reported D_e Values in PNNL-22747

Appendix C

Corrections to Previously Reported D_e Values in PNNL-22747

In Appendix D of the screening tests report (PNNL-22747) included tables of the measured effective diffusivities (D_e) values for the EPA 1315 test method through 91 days of testing for all 26 Cast Stone mixes. After the screening test report was issued, further reviews of the inputs to the effective diffusivity calculations identified a number of small errors in a few of the leachant volumes, test durations, and starting nitrate concentrations in the monoliths for the HTWOS Average simulants. The errors were small and most were for leach intervals that were not included in the statistical analyses of the screening test report. There are no impacts to the conclusions of the screening test report, PNNL-22747.

Regarding the leachant volume and test duration errors in PNNL-22747:

- Of the four mis-transcribed volumes, three (CS-T3HCS2-7.8AVG-6-77, CS-T5HCS1-7.8AVG-4-77, and CS-T5HCS1-7.8AVG-4-91) were for leach durations at 77 or 91 days. The data for these tests were not included in the statistical analyses of the screening test report, and so the only impact is in the diffusivity data tables in the screening test report Appendix D. For volume CS-T1LCS1-5HIS-4-28, the calculated technetium diffusivity is 5.25E-11 versus the PNNL-22747 reported value of 5.19E-11, or a 1% difference. This is less than the variability among replicate samples (5.03E-11 for the second sample). The difference between the correct average of the 28, 42, 49, and 63 day samples (4.75E-11) and the reported value (4.76E-11) is insignificant.

- Of the leach intervals, only the results for CS-T4HCS2-5HIA-4-28 were used in the screening test report. The difference in the time interval is 40 minutes out of 14 days or a difference of <0.2%. The calculated effective diffusivity is the same at 1.92E-11 for both incorrect and correct time intervals.

- For the other leach times (CS-T10HCS1-5HIS-1-7, CS-T11LCS1-7.8HIA-1-14, CS-T11LCS1-7.8HIA-6-14, CS-T12HCS2-5RAS-1-14, CS-T6LCS1-7.8AVG-1-7, CS-T15HCS1-7.8HIS-1-2, CS-T23LCS1-7.8RAS-1-14, and CS-T23LCS1-7.8RAS-3-14), the calculated effective diffusivities are shown in the tables in Appendix D of the screening test report, but are not used in any of the studies as part of the screening tests. Thus, there is no impact.

Regarding the starting nitrate concentration errors, in CALC-62745-008, Rev. 0, calculations of total nitrate concentrations in the "Overall Average" (or "Average") simulants neglected the contribution from the potassium nitrate (KNO_3) constituent for both the 5M Na and 7.8M Na preparations. This had the effect of making it appear that there was no potassium nitrate in the "Average" simulants.

- The input error in the KNO_3 constituent masses for the two "Average" simulants results in CALC-62745-008, Rev. 0 showing total NO_3 concentrations that are ~2% lower than the true NO_3 concentrations for Cast Stone monoliths containing the "Average" simulant. The following Cast Stone mixes shown in Table A-1 used the “Average” simulants: 2, 3, 5, 6, 13, 20, and 22.

- The error was propagated when CALC-62745-008, Rev. 0, was used as input in follow-on calculations in CALC-62745-007, Rev. 0 for the nitrate effective diffusivities for tests 2, 3, 5, 6, 13, 20, and 22. Impact to CALC-62745-007, Rev. 0 for tests 2, 3, 5, 6, 13, 20, and 22 is that the nitrate effective diffusivities shown in PNNL-22747 Appendix D are ~4% higher than with the corrected nitrate C_0 for the 5 M and 7.8 M Na Average simulants.

- The CALC-62745-007, Rev. 0 was used as input to CALC-62745-010, Rev. 0, which provided figures of the effective diffusivities. Impact to plots in CALC-62745-010, Rev. 0 is negligible because the figures use logarithmic axes spanning several orders of magnitude for the diffusivity values.

- The nitrate concentration error in the Average simulant has no impact on the statistical analyses in the screening tests report. The statistical analyses of the effective diffusivities were conducted on the leachability index, which is a logarithmic transformation of the effective diffusivity measurements. Also, the multiplicative approximate 4% error in effective diffusivities translates (through the logarithmic transformation) to a 0.017 additive error in leachability indices, which is considerably smaller than the pooled uncertainty estimate of 0.107 based on the replicate pairs in the testing as described in Section 8.7.2.2 of PNNL-22747.

- The nitrate effective diffusivities for tests 2, 3, 4, 6, 13, 20, and 22 in Tables D-8 through D-14 of the screening test report (PNNL-22747) are high by 4%. The leachability indices in Table D-15 in PNNL-22747 are low by 0.02. This has no impact on the conclusions of the screening test report.

Table C.1 through Table C.7, which were Tables D.7 through D.13 in PNNL-22747, provide corrected values for the measured effective diffusivities of Na, nitrate (NO_3), nitrite (NO_2), I, Tc, Cr, and U based on performing EPA Draft Method 1315 on duplicate (i.e., replicate) samples from each of the 26 Cast Stone mixes prepared at PNNL (Mixes 1–26) as reported in the screening test report. The leach tests were conducted for a total of 91 days and leachate exchanges occurred at 0.08, 1, 2, 7, 14, 28, 42, 49, 63, 77, and 91 cumulative days. The 77- and 91-day intervals provide longer-term data beyond the 63 days in EPA Method 1315, but they were not included in the statistical analyses for the screening tests. The results are provided for both monoliths of each of the 26 mixes. In the following Appendix C tables changes from the screening tests report Appendix D are italicized.

Appendix B.1 in this report includes the same D_e data for the first 91 days of leaching the selected monoliths for the Extended Suite as are found in Appendix C. We note in comparing the tables in Appendix B.1 and Appendix C, that there are small differences in the third digit for the reported effective diffusivities. We have checked the calculations and determined that the differences are due to the number of significant figures used for the monoliths’ surface area, volume, and density in the calculations. The values reported in Appendix B.1 are based on calculations using six significant digits and the D_e values reported in Appendix C are based on calculations using three significant digits for the monoliths’ surface area, volume, and density.

Table C.1. Sodium Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm^2/s) Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	1.53E-09	1.83E-07	3.84E-07	3.81E-07	3.45E-07	1.19E-07	7.57E-07	6.66E-08	8.69E-08	1.82E-07
0.92	1	2.26E-08	1.58E-08	7.63E-08	8.97E-08	9.86E-08	9.11E-08	1.69E-08	3.04E-08	3.60E-08	3.54E-08
1	2	4.32E-09	4.16E-09	3.22E-08	3.09E-08	2.85E-08	2.91E-08	1.86E-08	1.66E-08	3.16E-08	2.96E-08
5	7	1.97E-09	2.17E-09	2.01E-08	1.99E-08	1.37E-08	1.74E-08	5.17E-08	2.80E-08	1.17E-08	2.18E-08
7	14	1.97E-09	1.61E-09	1.36E-08	1.27E-08	9.96E-09	1.01E-08	1.52E-08	1.23E-08	1.37E-08	1.30E-08
14	28	***1.43E-09***	1.51E-09	8.60E-09	8.43E-09	7.01E-09	6.63E-09	***2.30E-08***	9.71E-09	3.99E-09	9.95E-09
14	42	1.22E-09	1.21E-09	5.72E-09	1.39E-08	2.40E-09	5.46E-09	***7.67E-09***	7.09E-09	6.16E-09	6.49E-09
7	49	1.16E-09	1.11E-09	6.01E-09	5.46E-09	5.33E-09	5.22E-09	***5.73E-09***	7.95E-09	5.16E-09	4.87E-09
14	63	1.00E-09	1.06E-09	3.67E-09	3.55E-09	3.96E-09	3.95E-09	***6.20E-09***	5.92E-09	3.79E-09	4.11E-09
14	77	1.09E-09	1.02E-09	2.15E-09	3.09E-09	3.81E-09	***2.75E-09***	4.34E-09	4.51E-09	3.40E-09	***2.08E-09***
14	91	9.22E-10	9.61E-10	2.51E-09	1.67E-09	3.50E-09	3.04E-09	4.28E-09	4.69E-09	1.86E-09	***2.40E-09***

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm^2/s) Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	3.04E-07	2.84E-07	2.05E-08	6.16E-08	1.21E-07	6.20E-08	5.23E-08	3.51E-07	4.01E-07	2.52E-07
0.92	1	5.58E-08	5.43E-08	2.22E-08	2.05E-08	1.17E-08	1.44E-08	3.55E-08	3.54E-08	3.69E-08	4.18E-08
1	2	4.13E-08	4.08E-08	1.87E-08	1.96E-08	6.03E-09	6.30E-09	2.66E-08	2.39E-08	3.00E-08	3.64E-08
5	7	***1.71E-08***	2.60E-08	1.00E-08	1.07E-08	4.22E-09	3.85E-09	1.33E-08	1.11E-08	***1.70E-08***	2.18E-08
7	14	***1.40E-08***	1.43E-08	8.66E-09	8.32E-09	3.91E-09	3.55E-09	6.04E-09	7.81E-09	***2.54E-08***	1.69E-08
14	28	***1.01E-08***	1.07E-08	4.93E-09	6.07E-09	2.80E-09	2.79E-09	5.85E-09	5.73E-09	1.02E-08	1.07E-08
14	42	6.81E-09	6.62E-09	3.38E-09	3.74E-09	2.53E-09	2.26E-09	4.55E-09	4.59E-09	***7.57E-09***	7.56E-09
7	49	4.93E-09	5.70E-09	2.77E-09	3.13E-09	2.35E-09	2.33E-09	4.44E-09	4.43E-09	6.44E-09	6.98E-09
14	63	3.50E-09	3.63E-09	1.97E-09	2.14E-09	2.11E-09	2.05E-09	3.76E-09	3.71E-09	5.35E-09	5.42E-09
14	77	1.42E-09	1.99E-09	9.43E-10	1.23E-09	1.65E-09	1.95E-09	2.52E-09	1.84E-09	***4.08E-09***	3.70E-09
14	91	1.15E-09	1.70E-09	1.08E-09	1.44E-09	1.06E-09	1.38E-09	2.50E-09	2.81E-09	3.32E-09	3.37E-09

Table C.1. Sodium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	7.50E-07	1.73E-07	5.50E-08	2.55E-08	7.17E-08	2.57E-07	1.10E-07	3.12E-07	3.97E-08	3.81E-08
0.92	1	4.91E-08	4.87E-08	7.87E-09	8.15E-09	4.02E-08	3.62E-08	2.80E-08	2.92E-08	3.38E-08	3.01E-08
1	2	3.36E-08	3.49E-08	6.30E-09	6.81E-09	3.28E-08	3.14E-08	2.38E-08	2.65E-08	***2.82E-08***	2.70E-08
5	7	1.88E-08	1.84E-08	5.46E-09	4.56E-09	2.04E-08	1.89E-08	1.34E-08	1.49E-08	1.40E-08	1.54E-08
7	14	***1.09E-08***	***1.25E-08***	***4.36E-09***	4.86E-09	1.37E-08	1.27E-08	8.89E-09	1.00E-08	1.24E-08	1.33E-08
14	28	***8.06E-09***	***7.21E-09***	4.08E-09	3.73E-09	8.86E-09	8.81E-09	5.65E-09	6.00E-09	9.50E-09	9.10E-09
14	42	***6.36E-09***	6.53E-09	4.21E-09	4.15E-09	7.04E-09	6.25E-09	4.82E-09	4.64E-09	8.22E-09	7.20E-09
7	49	5.55E-09	5.92E-09	***4.03E-09***	4.04E-09	6.18E-09	6.10E-09	4.06E-09	4.31E-09	7.45E-09	6.14E-09
14	63	4.43E-09	5.29E-09	4.09E-09	3.77E-09	4.16E-09	4.13E-09	3.04E-09	3.06E-09	5.88E-09	5.18E-09
14	77	2.26E-09	3.43E-09	3.31E-09	4.08E-09	3.59E-09	3.09E-09	2.42E-09	2.17E-09	3.07E-09	3.90E-09
14	91	1.64E-09	2.31E-09	3.57E-09	2.23E-09	2.90E-09	2.38E-09	1.55E-09	1.56E-09	3.03E-09	2.16E-09

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		16a	16b	17a	17b	18a	18b	19a	19b	20a	20b
0.08	0.08	4.39E-08	2.62E-08	7.81E-08	4.89E-08	2.37E-08	2.20E-08	5.85E-08	5.69E-08	3.14E-08	3.07E-08
0.92	1	2.44E-08	2.37E-08	3.81E-08	3.96E-08	9.93E-09	1.04E-08	3.43E-08	3.42E-08	1.65E-08	1.87E-08
1	2	1.98E-08	1.97E-08	3.78E-08	3.42E-08	9.23E-09	9.36E-09	2.57E-08	2.48E-08	1.54E-08	1.96E-08
5	7	1.10E-08	***1.09E-08***	1.76E-08	1.67E-08	4.65E-09	4.36E-09	1.16E-08	1.09E-08	1.93E-08	1.57E-08
7	14	1.15E-08	***1.18E-08***	1.29E-08	1.69E-08	5.18E-09	4.88E-09	9.25E-09	1.02E-08	2.51E-08	1.25E-08
14	28	8.36E-09	***8.83E-09***	9.47E-09	9.14E-09	3.04E-09	3.22E-09	6.09E-09	5.18E-09	1.18E-08	6.55E-09
14	42	7.00E-09	***7.58E-09***	6.61E-09	6.98E-09	2.80E-09	2.89E-09	3.68E-09	3.64E-09	9.35E-09	6.76E-09
7	49	6.49E-09	***6.65E-09***	5.92E-09	6.30E-09	2.76E-09	2.85E-09	2.54E-09	2.78E-09	9.38E-09	6.38E-09
14	63	5.75E-09	***5.89E-09***	4.60E-09	4.33E-09	2.70E-09	2.46E-09	1.71E-09	2.01E-09	7.50E-09	6.51E-09
14	77	2.31E-09	***3.39E-09***	3.53E-09	3.44E-09	1.75E-09	2.12E-09	1.01E-09	1.11E-09	3.72E-09	5.16E-09
14	91	3.45E-09	***3.44E-09***	2.92E-09	2.65E-09	1.82E-09	2.26E-09	8.68E-10	9.77E-10	4.03E-09	5.03E-09

Table C.1. Sodium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		21a	**21b**	**22a**	**22b**	**23a**	**23b**
0.08	0.08	1.50E-07	1.44E-07	5.69E-08	6.27E-08	9.95E-08	1.07E-07
0.92	1	9.58E-08	9.94E-08	3.55E-08	3.30E-08	1.01E-07	1.02E-07
1	2	7.57E-08	8.11E-08	2.71E-08	2.59E-08	1.02E-07	1.11E-07
5	7	3.51E-08	3.79E-08	1.14E-08	1.19E-08	5.96E-08	7.56E-08
7	14	3.10E-08	3.22E-08	9.27E-09	8.16E-09	***5.05E-08***	***5.44E-08***
14	28	1.53E-08	1.47E-08	5.49E-09	5.77E-09	3.38E-08	3.40E-08
14	42	1.13E-08	1.10E-08	5.17E-09	4.93E-09	2.42E-08	2.87E-08
7	49	9.14E-09	9.31E-09	4.61E-09	4.76E-09	2.30E-08	2.56E-08
14	63	6.46E-09	6.65E-09	3.92E-09	4.09E-09	1.72E-08	2.02E-08
14	77	2.81E-09	2.87E-09	2.91E-09	1.94E-09	8.71E-09	1.01E-08
14	91	2.70E-09	2.66E-09	3.82E-09	1.94E-09	6.16E-09	6.26E-09

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		24a	**24b**	**25a**	**25b**	**26a**	**26b**
0.08	0.08	1.01E-07	1.04E-07	5.21E-08	5.06E-08	1.39E-07	1.38E-07
0.92	1	6.05E-08	5.91E-08	3.36E-08	3.47E-08	6.28E-08	6.37E-08
1	2	4.92E-08	4.84E-08	2.85E-08	2.86E-08	4.83E-08	4.92E-08
5	7	2.48E-08	2.52E-08	1.90E-08	1.90E-08	2.42E-08	2.59E-08
7	14	1.73E-08	1.95E-08	1.15E-08	1.01E-08	1.42E-08	1.23E-08
14	28	1.42E-08	1.31E-08	1.00E-08	8.64E-09	1.10E-08	1.19E-08
14	42	9.16E-09	9.78E-09	7.32E-09	7.55E-09	8.98E-09	8.81E-09
7	49	8.58E-09	8.71E-09	5.90E-09	6.12E-09	6.84E-09	7.02E-09
14	63	7.86E-09	7.09E-09	4.75E-09	4.60E-09	5.07E-09	5.00E-09
14	77	3.69E-09	5.04E-09	3.66E-09	3.11E-09	2.71E-09	1.58E-09
14	91	3.44E-09	4.68E-09	2.93E-09	2.67E-09	2.74E-09	2.99E-09

Table C.2. Nitrate Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	4.73E-08	9.87E-08	***4.24E-07***	***7.34E-07***	***3.27E-07***	6.82E-09	1.83E-07	1.95E-07	***3.89E-07***	***3.79E-07***
0.92	1	2.33E-08	1.70E-08	***9.88E-08***	***1.11E-07***	***9.44E-08***	***1.07E-07***	1.66E-08	4.16E-08	***3.96E-08***	***4.29E-08***
1	2	3.83E-09	3.28E-09	***4.05E-08***	***3.93E-08***	***2.65E-08***	***2.70E-08***	1.83E-08	1.49E-08	***3.25E-08***	***3.41E-08***
5	7	2.62E-09	2.47E-09	***3.05E-08***	***2.73E-08***	***1.81E-08***	***1.60E-08***	3.95E-08	4.37E-08	***2.83E-08***	***2.95E-08***
7	14	1.53E-09	1.41E-09	***1.83E-08***	***1.73E-08***	***9.25E-09***	***8.45E-09***	1.47E-08	1.43E-08	***1.67E-08***	***1.65E-08***
14	28	***1.20E-09***	1.08E-09	***1.04E-08***	***1.02E-08***	***5.35E-09***	***5.41E-09***	***7.75E-09***	7.53E-09	***1.12E-08***	***1.11E-08***
14	42	6.07E-10	8.23E-10	***5.00E-09***	***5.62E-09***	***4.06E-09***	***4.45E-09***	5.59E-09	4.62E-09	***4.95E-09***	***4.92E-09***
7	49	7.97E-10	8.49E-10	***5.57E-09***	***5.27E-09***	***4.16E-09***	***3.95E-09***	4.50E-09	5.36E-09	***4.61E-09***	***5.02E-09***
14	63	7.93E-10	8.30E-10	***4.62E-09***	***4.41E-09***	***3.95E-09***	***3.95E-09***	4.51E-09	4.33E-09	***3.62E-09***	***3.98E-09***
14	77	7.79E-10	8.00E-10	***2.50E-09***	***3.37E-09***	***3.45E-09***	***2.31E-09***	2.71E-09	3.08E-09	***2.48E-09***	***1.46E-09***
14	91	7.54E-10	8.04E-10	***2.95E-09***	***1.91E-09***	***3.23E-09***	***2.78E-09***	3.19E-09	3.63E-09	***1.40E-09***	***2.05E-09***

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	***8.74E-07***	***1.10E-06***	4.18E-07	1.31E-07	5.02E-07	7.07E-07	2.06E-07	2.52E-07	4.46E-07	2.06E-07
0.92	1	***7.69E-08***	***8.08E-08***	3.46E-08	3.45E-08	1.28E-08	1.51E-08	3.55E-08	3.40E-08	4.01E-08	4.84E-08
1	2	***5.56E-08***	***5.72E-08***	2.97E-08	2.85E-08	6.23E-09	6.78E-09	2.21E-08	2.30E-08	3.25E-08	3.85E-08
5	7	***4.41E-08***	***4.31E-08***	1.94E-08	2.09E-08	5.98E-09	5.28E-09	1.31E-08	1.27E-08	***2.52E-08***	2.96E-08
7	14	***2.51E-08***	***2.53E-08***	1.18E-08	1.20E-08	4.31E-09	4.07E-09	6.95E-09	7.11E-09	***1.48E-08***	1.60E-08
14	28	***1.54E-08***	***1.65E-08***	8.08E-09	1.01E-08	3.35E-09	3.23E-09	5.42E-09	5.29E-09	***9.81E-09***	9.35E-09
14	42	***7.49E-09***	***9.04E-09***	3.22E-09	4.06E-09	2.27E-09	1.78E-09	2.73E-09	3.40E-09	5.20E-09	5.82E-09
7	49	***8.93E-09***	***7.76E-09***	3.23E-09	4.10E-09	2.85E-09	2.36E-09	4.41E-09	3.48E-09	5.60E-09	4.60E-09
14	63	***4.76E-09***	***5.26E-09***	2.37E-09	2.66E-09	2.53E-09	2.41E-09	3.94E-09	4.10E-09	4.28E-09	4.04E-09
14	77	***1.59E-09***	***2.37E-09***	9.46E-10	1.26E-09	1.87E-09	2.16E-09	2.42E-09	1.73E-09	2.66E-09	2.35E-09
14	91	***1.39E-09***	***2.07E-09***	1.14E-09	1.85E-09	1.19E-09	1.59E-09	3.41E-09	3.24E-09	2.62E-09	2.86E-09

Table C.2. Nitrate Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	1.98E-07	4.51E-07	1.45E-07	1.44E-07	***3.02E-07***	***4.26E-07***	4.71E-07	4.18E-07	5.33E-08	4.24E-08
0.92	1	5.34E-08	5.48E-08	8.70E-09	7.75E-09	***5.73E-08***	***5.36E-08***	4.43E-08	4.81E-08	4.11E-08	3.76E-08
1	2	3.56E-08	3.64E-08	6.22E-09	6.20E-09	***4.08E-08***	***4.14E-08***	3.82E-08	4.17E-08	***3.09E-08***	3.05E-08
5	7	2.15E-08	2.43E-08	7.51E-09	5.91E-09	***3.39E-08***	***3.23E-08***	3.02E-08	3.19E-08	2.27E-08	2.23E-08
7	14	1.20E-08	***1.10E-08***	***4.76E-09***	5.41E-09	***2.09E-08***	***2.12E-08***	1.64E-08	1.86E-08	1.61E-08	1.63E-08
14	28	***9.77E-09***	1.02E-08	4.16E-09	4.46E-09	***1.36E-08***	***1.31E-08***	9.71E-09	1.01E-08	1.08E-08	1.06E-08
14	42	7.98E-09	7.40E-09	***3.49E-09***	3.35E-09	***8.40E-09***	***6.39E-09***	5.05E-09	4.29E-09	4.20E-09	5.15E-09
7	49	8.06E-09	9.15E-09	4.13E-09	4.32E-09	***7.79E-09***	***6.98E-09***	6.60E-09	6.26E-09	8.27E-09	6.45E-09
14	63	***7.49E-09***	8.79E-09	4.05E-09	3.58E-09	***5.50E-09***	***5.46E-09***	3.93E-09	4.07E-09	5.83E-09	4.71E-09
14	77	3.57E-09	4.97E-09	2.92E-09	3.64E-09	***4.02E-09***	***3.18E-09***	2.77E-09	2.41E-09	3.03E-09	3.90E-09
14	91	***2.70E-09***	4.02E-09	3.91E-09	2.20E-09	***3.72E-09***	***3.08E-09***	1.94E-09	1.89E-09	2.53E-09	1.98E-09

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		16a	16b	17a	17b	18a	18b	19a	19b	20a	20b
0.08	0.08	5.60E-08	5.08E-08	7.20E-08	6.84E-08	3.33E-08	3.46E-08	8.30E-08	8.00E-08	***3.71E-08***	***3.32E-08***
0.92	1	3.16E-08	3.13E-08	5.93E-08	5.90E-08	1.50E-08	1.51E-08	5.07E-08	5.17E-08	***1.48E-08***	***1.93E-08***
1	2	2.69E-08	2.75E-08	5.22E-08	4.78E-08	1.23E-08	1.25E-08	3.71E-08	3.54E-08	***1.20E-08***	***2.07E-08***
5	7	1.67E-08	***1.76E-08***	2.95E-08	2.92E-08	7.84E-09	7.75E-09	1.94E-08	2.10E-08	***2.63E-08***	***2.36E-08***
7	14	1.38E-08	***1.49E-08***	1.84E-08	1.92E-08	5.50E-09	5.39E-09	1.20E-08	1.42E-08	***2.57E-08***	***1.14E-08***
14	28	1.03E-08	***1.19E-08***	1.15E-08	1.21E-08	3.89E-09	4.00E-09	8.90E-09	8.47E-09	***9.66E-09***	***6.33E-09***
14	42	6.49E-09	***6.34E-09***	5.57E-09	7.08E-09	2.47E-09	3.04E-09	4.37E-09	3.76E-09	***5.48E-09***	***4.40E-09***
7	49	8.14E-09	***7.91E-09***	6.86E-09	7.07E-09	3.44E-09	3.44E-09	3.60E-09	4.08E-09	***6.23E-09***	***4.66E-09***
14	63	6.62E-09	***6.67E-09***	5.72E-09	5.79E-09	3.16E-09	3.08E-09	2.34E-09	2.75E-09	***5.37E-09***	***4.53E-09***
14	77	2.78E-09	***4.20E-09***	4.82E-09	4.93E-09	2.42E-09	2.86E-09	1.38E-09	1.54E-09	***2.80E-09***	***3.86E-09***
14	91	4.02E-09	*3.76E-09*	3.83E-09	3.48E-09	2.23E-09	2.80E-09	1.00E-09	1.14E-09	***2.63E-09***	***3.53E-09***

Table C.2. Nitrate Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	2.06E-07	2.01E-07	***6.41E-08***	***6.96E-08***	1.48E-07	1.48E-07
0.92	1	1.31E-07	1.28E-07	***4.52E-08***	***4.03E-08***	1.69E-07	1.74E-07
1	2	1.06E-07	1.06E-07	***2.72E-08***	***2.67E-08***	1.67E-07	1.77E-07
5	7	6.62E-08	6.10E-08	***1.35E-08***	***1.27E-08***	1.15E-07	1.36E-07
7	14	4.41E-08	3.79E-08	***7.79E-09***	***7.68E-09***	***7.33E-08***	***8.84E-08***
14	28	2.42E-08	2.31E-08	***5.16E-09***	***5.13E-09***	4.86E-08	***5.76E-08***
14	42	1.47E-08	1.20E-08	***4.40E-09***	***3.06E-09***	4.12E-08	3.66E-08
7	49	1.37E-08	1.37E-08	***3.69E-09***	***4.21E-09***	2.86E-08	3.37E-08
14	63	9.83E-09	9.97E-09	***3.62E-09***	***3.90E-09***	2.29E-08	***2.85E-08***
14	77	4.28E-09	4.36E-09	***2.99E-09***	***1.69E-09***	1.21E-08	1.35E-08
14	91	3.63E-09	3.44E-09	***3.38E-09***	***1.67E-09***	7.59E-09	7.62E-09

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		24a	24b	25a	25b	26a	26b
0.08	0.08	1.15E-07	1.30E-07	6.84E-08	6.53E-08	1.50E-07	1.54E-07
0.92	1	6.92E-08	7.12E-08	4.36E-08	4.30E-08	7.13E-08	6.89E-08
1	2	6.03E-08	5.00E-08	3.49E-08	3.31E-08	4.73E-08	4.80E-08
5	7	3.38E-08	3.52E-08	2.03E-08	2.08E-08	2.56E-08	2.61E-08
7	14	1.78E-08	2.21E-08	1.29E-08	9.35E-09	1.09E-08	1.24E-08
14	28	1.47E-08	1.37E-08	1.12E-08	1.04E-08	1.21E-08	1.21E-08
14	42	7.25E-09	9.24E-09	7.36E-09	6.21E-09	7.87E-09	6.73E-09
7	49	8.69E-09	8.61E-09	5.56E-09	5.70E-09	7.06E-09	6.71E-09
14	63	7.68E-09	7.22E-09	4.73E-09	4.71E-09	5.07E-09	5.25E-09
14	77	3.99E-09	5.47E-09	3.25E-09	3.48E-09	1.61E-09	3.05E-09
14	91	3.04E-09	4.67E-09	2.81E-09	2.44E-09	2.48E-09	2.91E-09

Table C.3. Nitrite Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		1a	**1b**	**2a**	**2b**	**3a**	**3b**	**4a**	**4b**	**5a**	**5b**
0.08	0.08	5.31E-08	8.63E-08	2.65E-07	8.01E-07	3.70E-07	5.93E-09	9.52E-08	1.01E-07	4.06E-07	3.91E-07
0.92	1	2.09E-08	1.55E-08	7.98E-08	9.17E-08	8.18E-08	9.12E-08	1.42E-08	3.45E-08	3.42E-08	3.68E-08
1	2	4.16E-09	3.59E-09	4.26E-08	4.14E-08	3.10E-08	3.15E-08	1.97E-08	1.63E-08	3.52E-08	3.70E-08
5	7	3.17E-09	2.97E-09	3.35E-08	2.95E-08	2.05E-08	1.84E-08	4.10E-08	4.40E-08	2.89E-08	2.99E-08
7	14	1.61E-09	1.52E-09	1.86E-08	1.74E-08	1.01E-08	8.83E-09	1.41E-08	1.38E-08	1.49E-08	1.52E-08
14	28	***1.52E-09***	1.33E-09	1.44E-08	1.34E-08	7.52E-09	7.15E-09	***9.19E-09***	8.64E-09	1.13E-08	1.09E-08
14	42	7.41E-10	9.77E-10	5.55E-09	6.37E-09	4.95E-09	5.30E-09	6.71E-09	5.37E-09	5.26E-09	5.11E-09
7	49	8.75E-10	9.37E-10	5.45E-09	5.11E-09	4.54E-09	4.37E-09	***4.69E-09***	5.59E-09	4.63E-09	5.08E-09
14	63	8.67E-10	9.16E-10	4.99E-09	4.65E-09	4.93E-09	4.73E-09	***4.79E-09***	4.48E-09	3.57E-09	3.92E-09
14	77	9.02E-10	8.45E-10	2.52E-09	3.31E-09	4.27E-09	***2.82E-09***	2.90E-09	3.00E-09	2.47E-09	1.39E-09
14	91	8.95E-10	9.34E-10	3.12E-09	2.10E-09	4.69E-09	3.95E-09	3.68E-09	4.16E-09	1.57E-09	***2.27E-09***

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		6a	**6b**	**7a**	**7b**	**8a**	**8b**	**9a**	**9b**	**10a**	**10b**
0.08	0.08	8.91E-07	1.06E-06	2.11E-07	8.54E-08	2.70E-07	3.90E-07	2.45E-07	2.92E-07	2.22E-07	1.18E-07
0.92	1	6.55E-08	6.83E-08	3.22E-08	3.19E-08	1.06E-08	1.22E-08	3.31E-08	3.18E-08	3.48E-08	4.17E-08
1	2	6.34E-08	6.50E-08	3.49E-08	3.42E-08	6.29E-09	6.65E-09	2.80E-08	2.86E-08	3.51E-08	4.18E-08
5	7	***4.61E-08***	4.47E-08	2.10E-08	2.37E-08	6.37E-09	5.64E-09	1.61E-08	1.55E-08	***2.58E-08***	3.12E-08
7	14	***2.30E-08***	2.40E-08	1.18E-08	1.18E-08	3.93E-09	3.79E-09	7.67E-09	7.74E-09	***1.32E-08***	1.39E-08
14	28	***1.63E-08***	1.67E-08	7.88E-09	9.45E-09	3.02E-09	2.92E-09	6.30E-09	5.93E-09	***8.42E-09***	8.10E-09
14	42	***8.21E-09***	9.92E-09	3.36E-09	4.30E-09	2.27E-09	1.73E-09	3.69E-09	4.43E-09	5.11E-09	5.80E-09
7	49	***9.25E-09***	8.30E-09	3.30E-09	4.15E-09	2.74E-09	2.35E-09	5.88E-09	4.53E-09	*5.31E-09*	4.44E-09
14	63	4.99E-09	5.37E-09	2.34E-09	2.66E-09	2.30E-09	2.16E-09	4.86E-09	4.88E-09	3.85E-09	3.45E-09
14	77	1.59E-09	2.44E-09	9.12E-10	1.22E-09	1.73E-09	2.04E-09	3.19E-09	2.30E-09	2.54E-09	2.15E-09
14	91	1.55E-09	2.35E-09	1.20E-09	1.96E-09	1.16E-09	1.54E-09	4.63E-09	4.99E-09	2.48E-09	2.71E-09

Table C.3. Nitrite Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	2.13E-07	4.46E-07	9.15E-08	8.75E-08	1.52E-07	2.11E-07	2.26E-07	2.21E-07	6.00E-08	4.76E-08
0.92	1	4.76E-08	4.83E-08	8.12E-09	7.20E-09	4.85E-08	4.54E-08	4.17E-08	4.44E-08	4.56E-08	4.22E-08
1	2	4.36E-08	4.47E-08	6.66E-09	6.67E-09	4.36E-08	4.42E-08	4.51E-08	4.88E-08	***3.57E-08***	3.61E-08
5	7	2.44E-08	2.66E-08	7.92E-09	6.23E-09	3.45E-08	3.27E-08	3.37E-08	3.55E-08	2.06E-08	2.02E-08
7	14	***1.24E-08***	***1.14E-08***	***4.22E-09***	4.76E-09	1.85E-08	1.83E-08	1.55E-08	1.66E-08	2.02E-08	1.88E-08
14	28	***9.94E-09***	1.02E-08	3.48E-09	3.71E-09	1.13E-08	1.08E-08	8.60E-09	9.01E-09	1.22E-08	1.03E-08
14	42	9.33E-09	***8.82E-09***	3.41E-09	3.26E-09	7.92E-09	6.19E-09	5.21E-09	4.43E-09	4.79E-09	5.73E-09
7	49	1.01E-08	1.16E-08	4.07E-09	4.23E-09	7.57E-09	6.74E-09	6.66E-09	6.36E-09	7.69E-09	6.04E-09
14	63	8.17E-09	9.58E-09	3.57E-09	3.15E-09	4.72E-09	4.48E-09	3.45E-09	3.69E-09	5.96E-09	4.66E-09
14	77	4.31E-09	6.05E-09	2.54E-09	3.36E-09	3.52E-09	2.78E-09	2.61E-09	2.23E-09	3.29E-09	4.18E-09
14	91	3.77E-09	5.41E-09	***3.61E-09***	1.95E-09	3.31E-09	2.84E-09	1.81E-09	1.80E-09	3.00E-09	2.31E-09

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		16a	16b	17a	17b	18a	18b	19a	19b	20a	20b
0.08	0.08	6.15E-08	5.66E-08	7.60E-08	7.00E-08	3.69E-08	3.78E-08	1.06E-07	1.02E-07	3.82E-08	3.32E-08
0.92	1	3.45E-08	3.46E-08	6.27E-08	6.24E-08	1.63E-08	1.64E-08	6.11E-08	6.31E-08	1.72E-08	2.25E-08
1	2	3.10E-08	3.05E-08	5.80E-08	5.23E-08	1.32E-08	1.37E-08	4.91E-08	4.74E-08	1.44E-08	2.50E-08
5	7	1.53E-08	1.46E-08	2.54E-08	2.54E-08	6.97E-09	6.92E-09	1.98E-08	2.12E-08	2.52E-08	2.28E-08
7	14	1.49E-08	***1.61E-08***	2.19E-08	2.10E-08	5.26E-09	5.01E-09	1.49E-08	1.64E-08	2.61E-08	1.19E-08
14	28	9.47E-09	***1.07E-08***	1.10E-08	1.07E-08	3.07E-09	3.23E-09	8.89E-09	8.27E-09	8.50E-09	5.64E-09
14	42	6.27E-09	***6.60E-09***	6.09E-09	7.54E-09	2.34E-09	2.93E-09	5.63E-09	4.63E-09	5.71E-09	4.86E-09
7	49	7.28E-09	***6.98E-09***	6.46E-09	6.44E-09	3.16E-09	3.15E-09	4.10E-09	4.63E-09	6.45E-09	4.91E-09
14	63	6.01E-09	***5.91E-09***	5.53E-09	5.22E-09	2.80E-09	2.80E-09	2.73E-09	3.25E-09	5.30E-09	4.55E-09
14	77	2.74E-09	***4.05E-09***	5.05E-09	4.91E-09	2.34E-09	2.85E-09	1.73E-09	1.95E-09	2.85E-09	3.88E-09
14	91	3.90E-09	***3.96E-09***	4.45E-09	3.78E-09	2.21E-09	2.71E-09	1.42E-09	1.57E-09	2.51E-09	3.62E-09

Table C.3. Nitrite Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		21a	**21b**	**22a**	**22b**	**23a**	**23b**
0.08	0.08	2.18E-07	2.16E-07	7.53E-08	8.24E-08	1.55E-07	1.56E-07
0.92	1	1.41E-07	1.40E-07	5.14E-08	4.65E-08	1.53E-07	1.66E-07
1	2	1.27E-07	1.19E-07	3.38E-08	3.30E-08	1.74E-07	1.87E-07
5	7	5.64E-08	5.10E-08	1.35E-08	1.29E-08	8.58E-08	9.79E-08
7	14	4.35E-08	3.73E-08	8.03E-09	8.01E-09	***6.08E-08***	***7.47E-08***
14	28	2.01E-08	1.90E-08	4.49E-09	4.57E-09	3.23E-08	3.65E-08
14	42	1.55E-08	1.25E-08	4.76E-09	3.40E-09	2.66E-08	2.88E-08
7	49	1.23E-08	1.28E-08	3.86E-09	4.35E-09	2.27E-08	2.62E-08
14	63	8.80E-09	9.35E-09	***3.70E-09***	***3.99E-09***	1.73E-08	2.06E-08
14	77	4.40E-09	4.42E-09	***3.32E-09***	***1.88E-09***	1.03E-08	1.05E-08
14	91	3.79E-09	3.58E-09	***3.76E-09***	***1.87E-09***	6.57E-09	6.15E-09

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		24a	**24b**	**25a**	**25b**	**26a**	**26b**
0.08	0.08	1.17E-07	1.30E-07	7.25E-08	7.10E-08	1.58E-07	1.66E-07
0.92	1	6.68E-08	6.94E-08	4.46E-08	4.51E-08	7.69E-08	7.64E-08
1	2	6.56E-08	5.53E-08	3.81E-08	3.70E-08	5.60E-08	5.63E-08
5	7	2.75E-08	2.90E-08	1.74E-08	1.79E-08	2.38E-08	2.43E-08
7	14	1.58E-08	1.90E-08	1.12E-08	8.39E-09	1.10E-08	1.26E-08
14	28	1.07E-08	1.01E-08	8.10E-09	7.60E-09	9.72E-09	9.65E-09
14	42	6.77E-09	8.33E-09	6.58E-09	5.67E-09	7.59E-09	6.66E-09
7	49	7.76E-09	7.40E-09	5.04E-09	5.24E-09	6.81E-09	6.50E-09
14	63	6.55E-09	6.26E-09	4.04E-09	4.12E-09	4.71E-09	5.06E-09
14	77	3.63E-09	5.08E-09	3.00E-09	3.21E-09	1.66E-09	3.16E-09
14	91	2.97E-09	4.56E-09	2.47E-09	2.20E-09	2.47E-09	2.75E-09

Table C.4. Iodine Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	1.19E-09	5.41E-08	1.83E-07	5.25E-07	3.53E-07	6.48E-07	8.16E-08	1.43E-07	3.28E-07	4.25E-07
0.92	1	1.71E-08	1.71E-08	1.05E-07	1.25E-07	1.33E-07	1.36E-07	1.27E-08	2.27E-08	3.77E-08	3.91E-08
1	2	3.15E-09	2.82E-09	3.99E-08	3.91E-08	3.39E-08	3.32E-08	1.31E-08	1.14E-08	3.45E-08	3.43E-08
5	7	1.67E-09	1.63E-09	2.50E-08	2.42E-08	1.76E-08	1.71E-08	2.41E-08	2.45E-08	2.33E-08	2.32E-08
7	14	1.05E-09	1.03E-09	1.64E-08	1.57E-08	9.73E-09	8.68E-09	9.18E-09	8.85E-09	1.44E-08	1.45E-08
14	28	***8.63E-10***	8.51E-10	1.09E-08	1.05E-08	6.32E-09	6.06E-09	***5.36E-09***	5.37E-09	9.32E-09	9.48E-09
14	42	7.01E-10	7.04E-10	7.17E-09	6.97E-09	4.72E-09	5.51E-09	4.01E-09	3.70E-09	5.23E-09	5.31E-09
7	49	6.18E-10	6.04E-10	5.79E-09	5.40E-09	4.54E-09	4.46E-09	***3.03E-09***	3.44E-09	3.55E-09	3.70E-09
14	63	6.00E-10	6.09E-10	4.76E-09	4.72E-09	4.18E-09	4.08E-09	3.19E-09	3.29E-09	2.77E-09	2.94E-09
14	77	5.88E-10	6.01E-10	2.61E-09	3.54E-09	3.59E-09	***2.36E-09***	2.06E-09	2.32E-09	1.74E-09	1.07E-09
14	91	5.70E-10	6.02E-10	2.97E-09	1.85E-09	3.17E-09	2.69E-09	2.34E-09	2.70E-09	9.51E-10	***1.34E-09***

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	8.06E-07	5.82E-07	6.90E-09	1.60E-07	1.07E-07	6.37E-08	5.66E-07	4.46E-07	3.04E-07	1.60E-07
0.92	1	9.52E-08	9.67E-08	3.77E-08	3.68E-08	1.17E-08	1.29E-08	4.78E-08	4.77E-08	2.83E-08	3.32E-08
1	2	7.19E-08	7.27E-08	3.21E-08	2.99E-08	6.18E-09	6.14E-09	3.03E-08	2.84E-08	2.29E-08	2.80E-08
5	7	***4.54E-08***	4.55E-08	1.90E-08	1.91E-08	4.74E-09	4.34E-09	1.45E-08	1.40E-08	***1.64E-08***	1.94E-08
7	14	***2.69E-08***	2.82E-08	1.12E-08	1.12E-08	3.79E-09	3.54E-09	8.01E-09	8.20E-09	1.06E-08	1.18E-08
14	28	***1.76E-08***	1.78E-08	7.09E-09	8.36E-09	3.19E-09	3.03E-09	6.41E-09	6.05E-09	6.66E-09	6.93E-09
14	42	1.05E-08	1.07E-08	3.93E-09	4.53E-09	2.68E-09	2.44E-09	5.08E-09	5.01E-09	4.47E-09	4.49E-09
7	49	***7.00E-09***	7.52E-09	2.68E-09	2.97E-09	2.42E-09	2.27E-09	4.70E-09	4.91E-09	3.58E-09	3.42E-09
14	63	4.74E-09	5.29E-09	2.02E-09	2.37E-09	2.22E-09	2.13E-09	4.62E-09	4.75E-09	3.08E-09	2.78E-09
14	77	***1.47E-09***	2.29E-09	8.01E-10	1.05E-09	1.68E-09	1.82E-09	2.77E-09	2.00E-09	1.93E-09	1.60E-09
14	91	1.14E-09	1.74E-09	9.78E-10	1.47E-09	1.07E-09	1.40E-09	3.22E-09	3.70E-09	1.91E-09	1.92E-09

Table C.4. Iodine Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm^2/s) Mix Number and Duplicate (Repeat)									
		11a	**11b**	**12a**	**12b**	**13a**	**13b**	**14a**	**14b**	**15a**	**15b**
0.08	0.08	2.77E-07	4.74E-07	1.17E-08	3.67E-08	1.79E-07	3.77E-07	3.40E-07	2.80E-07	3.15E-08	2.89E-08
0.92	1	7.79E-08	7.97E-08	6.32E-09	6.32E-09	5.82E-08	5.44E-08	5.45E-08	5.85E-08	2.83E-08	2.58E-08
1	2	5.25E-08	5.17E-08	4.66E-09	4.79E-09	4.12E-08	4.01E-08	4.29E-08	4.50E-08	***2.43E-08***	2.33E-08
5	7	2.93E-08	2.94E-08	4.90E-09	4.04E-09	2.91E-08	2.74E-08	2.77E-08	2.98E-08	1.87E-08	1.87E-08
7	14	***1.62E-08***	***1.59E-08***	***3.48E-09***	3.84E-09	2.01E-08	1.95E-08	1.64E-08	1.84E-08	1.35E-08	1.30E-08
14	28	1.33E-08	1.32E-08	2.77E-09	3.12E-09	1.30E-08	1.24E-08	8.85E-09	9.05E-09	8.83E-09	8.26E-09
14	42	1.24E-08	1.29E-08	2.77E-09	2.74E-09	9.18E-09	8.54E-09	6.13E-09	5.94E-09	6.13E-09	5.52E-09
7	49	1.11E-08	1.24E-08	2.53E-09	2.56E-09	6.94E-09	6.54E-09	4.43E-09	4.56E-09	5.22E-09	4.16E-09
14	63	***9.98E-09***	1.19E-08	2.72E-09	2.54E-09	5.75E-09	5.50E-09	3.24E-09	3.40E-09	4.03E-09	3.20E-09
14	77	4.74E-09	6.64E-09	2.03E-09	2.64E-09	4.04E-09	3.30E-09	2.25E-09	2.01E-09	2.04E-09	2.52E-09
14	91	3.74E-09	5.43E-09	2.68E-09	1.62E-09	3.86E-09	3.12E-09	1.66E-09	1.63E-09	1.85E-09	1.37E-09

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm^2/s) Mix Number and Duplicate (Repeat)									
		16a	**16b**	**17a**	**17b**	**18a**	**18b**	**19a**	**19b**	**20a**	**20b**
0.08	0.08	4.53E-08	4.17E-08	5.89E-08	5.74E-08	2.94E-08	2.97E-08	9.25E-08	8.69E-08	2.59E-08	2.19E-08
0.92	1	2.65E-08	2.56E-08	5.10E-08	5.10E-08	1.37E-08	1.41E-08	6.22E-08	6.32E-08	1.03E-08	1.29E-08
1	2	2.23E-08	2.25E-08	4.77E-08	4.52E-08	1.14E-08	1.08E-08	4.73E-08	4.74E-08	8.14E-09	1.25E-08
5	7	1.66E-08	***1.50E-08***	2.74E-08	2.88E-08	7.24E-09	7.27E-09	2.62E-08	2.81E-08	1.73E-08	1.40E-08
7	14	1.35E-08	***1.50E-08***	1.79E-08	1.87E-08	5.37E-09	5.35E-09	1.64E-08	1.79E-08	1.60E-08	6.93E-09
14	28	9.98E-09	***1.09E-08***	1.11E-08	1.16E-08	3.73E-09	3.85E-09	1.04E-08	9.85E-09	6.37E-09	3.93E-09
14	42	7.93E-09	***8.70E-09***	7.72E-09	7.76E-09	3.04E-09	3.28E-09	5.92E-09	5.62E-09	4.38E-09	3.15E-09
7	49	6.64E-09	***6.88E-09***	5.83E-09	6.14E-09	2.72E-09	2.87E-09	3.28E-09	3.81E-09	3.71E-09	2.67E-09
14	63	5.98E-09	***6.13E-09***	5.20E-09	5.32E-09	2.79E-09	2.85E-09	2.19E-09	2.67E-09	3.54E-09	2.71E-09
14	77	2.41E-09	***3.67E-09***	4.44E-09	4.52E-09	2.20E-09	2.60E-09	1.21E-09	1.35E-09	1.92E-09	2.58E-09
14	91	3.64E-09	***3.33E-09***	3.63E-09	3.35E-09	2.08E-09	2.64E-09	8.84E-10	9.86E-10	1.88E-09	2.41E-09

Table C.4. Iodine Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm²/s)					
		Mix Number and Duplicate (Repeat)					
		21a	**21b**	**22a**	**22b**	**23a**	**23b**
0.08	0.08	1.69E-07	1.80E-07	5.79E-08	6.40E-08	1.42E-07	1.45E-07
0.92	1	1.55E-07	1.51E-07	5.16E-08	4.42E-08	1.72E-07	1.79E-07
1	2	1.18E-07	1.19E-07	2.88E-08	2.69E-08	1.72E-07	1.91E-07
5	7	7.55E-08	7.27E-08	1.44E-08	1.39E-08	1.16E-07	1.41E-07
7	14	5.16E-08	4.92E-08	8.49E-09	8.38E-09	***7.88E-08***	***9.77E-08***
14	28	2.82E-08	2.79E-08	5.78E-09	5.53E-09	4.87E-08	5.94E-08
14	42	1.73E-08	1.75E-08	4.56E-09	4.40E-09	3.20E-08	3.84E-08
7	49	1.25E-08	1.28E-08	3.79E-09	3.99E-09	2.65E-08	***2.98E-08***
14	63	9.08E-09	9.11E-09	3.61E-09	3.71E-09	2.00E-08	2.41E-08
14	77	4.09E-09	3.97E-09	3.07E-09	1.68E-09	1.17E-08	1.18E-08
14	91	3.40E-09	3.23E-09	3.28E-09	1.69E-09	***6.99E-09***	5.81E-09

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm²/s)					
		Mix Number and Duplicate (Repeat)					
		24a	**24b**	**25a**	**25b**	**26a**	**26b**
0.08	0.08	8.45E-08	8.69E-08	4.33E-08	4.16E-08	1.22E-07	1.21E-07
0.92	1	5.67E-08	5.70E-08	3.16E-08	3.24E-08	6.60E-08	6.69E-08
1	2	4.72E-08	4.52E-08	2.59E-08	2.55E-08	4.64E-08	4.72E-08
5	7	2.94E-08	3.03E-08	1.81E-08	1.72E-08	2.62E-08	2.72E-08
7	14	1.63E-08	1.99E-08	9.22E-09	9.11E-09	1.14E-08	1.14E-08
14	28	1.27E-08	1.25E-08	8.52E-09	7.66E-09	1.17E-08	1.21E-08
14	42	8.37E-09	8.17E-09	4.96E-09	4.94E-09	7.94E-09	7.65E-09
7	49	6.22E-09	6.22E-09	3.42E-09	3.59E-09	5.46E-09	5.43E-09
14	63	5.71E-09	5.41E-09	2.93E-09	2.93E-09	3.98E-09	4.14E-09
14	77	3.26E-09	4.21E-09	2.04E-09	2.16E-09	1.31E-09	2.47E-09
14	91	2.47E-09	3.50E-09	1.73E-09	1.52E-09	1.92E-09	2.29E-09

Table C.5. Technetium Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm^2/s) Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	3.67E-11	6.45E-09	2.60E-09	2.76E-09	2.08E-09	6.86E-10	3.75E-13	2.68E-12	5.24E-11	5.69E-11
0.92	1	5.25E-10	4.28E-10	4.06E-10	5.75E-10	4.04E-10	4.04E-10	2.91E-12	5.77E-12	1.08E-11	4.58E-11
1	2	1.39E-10	1.26E-10	9.22E-11	1.18E-10	8.18E-12	7.54E-12	9.30E-12	9.99E-12	2.37E-11	7.44E-11
5	7	1.12E-10	1.05E-10	4.79E-11	5.02E-11	1.26E-11	1.33E-11	1.47E-11	1.62E-11	2.76E-11	5.51E-11
7	14	7.50E-11	7.22E-11	5.37E-11	4.67E-11	2.51E-11	2.88E-11	2.05E-11	1.73E-11	5.34E-11	6.58E-11
14	28	***5.25E-11***	5.03E-11	7.06E-11	5.56E-11	3.13E-11	3.02E-11	1.92E-11	2.51E-11	5.00E-11	5.57E-11
14	42	4.22E-11	3.97E-11	7.23E-11	5.89E-11	3.41E-11	3.49E-11	2.28E-11	2.57E-11	8.41E-11	7.96E-11
7	49	4.43E-11	4.15E-11	6.59E-11	5.13E-11	3.61E-11	3.82E-11	***2.91E-11***	3.65E-11	8.58E-11	8.77E-11
14	63	5.14E-11	5.07E-11	5.86E-11	4.56E-11	3.71E-11	3.56E-11	3.05E-11	3.72E-11	6.44E-11	6.44E-11
14	77	4.89E-11	4.87E-11	3.32E-11	3.60E-11	3.29E-11	***2.14E-11***	2.28E-11	2.93E-11	3.73E-11	***2.19E-11***
14	91	4.39E-11	4.56E-11	3.89E-11	1.90E-11	3.01E-11	2.46E-11	2.66E-11	3.92E-11	2.16E-11	***2.78E-11***

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm^2/s) Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	2.44E-09	3.30E-09	1.09E-12	8.96E-12	2.19E-09	1.25E-09	8.99E-12	1.96E-10	7.07E-09	6.91E-09
0.92	1	5.49E-11	3.22E-11	1.03E-11	1.06E-11	2.28E-10	3.95E-10	2.40E-11	3.76E-11	1.11E-09	2.01E-09
1	2	7.61E-12	5.60E-12	9.74E-12	1.25E-11	2.51E-10	5.10E-10	5.74E-11	5.58E-11	5.99E-10	1.03E-09
5	7	***7.00E-12***	6.36E-12	7.97E-12	1.22E-11	2.89E-10	4.43E-10	3.74E-11	3.73E-11	***3.02E-10***	4.57E-10
7	14	***1.81E-11***	1.80E-11	7.91E-12	1.09E-11	2.75E-10	3.15E-10	4.73E-11	4.21E-11	***2.00E-10***	2.42E-10
14	28	***4.23E-11***	4.08E-11	8.74E-12	9.66E-12	2.22E-10	2.23E-10	2.98E-11	3.03E-11	1.40E-10	1.49E-10
14	42	1.26E-10	1.24E-10	9.22E-12	8.75E-12	2.21E-10	2.03E-10	3.92E-11	3.79E-11	***1.14E-10***	1.19E-10
7	49	2.52E-10	2.35E-10	9.51E-12	1.01E-11	2.20E-10	2.08E-10	4.16E-11	3.84E-11	1.16E-10	1.16E-10
14	63	1.96E-10	2.01E-10	8.40E-12	8.17E-12	2.26E-10	2.03E-10	3.20E-11	2.86E-11	1.15E-10	1.11E-10
14	77	8.05E-11	1.07E-10	3.58E-12	4.90E-12	1.62E-10	1.75E-10	1.88E-11	1.29E-11	8.20E-11	7.17E-11
14	91	***8.12E-11***	1.20E-10	5.85E-12	7.67E-12	8.25E-11	1.18E-10	2.14E-11	2.40E-11	8.35E-11	8.80E-11

Table C.5. Technetium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm²/s) Mix Number and Duplicate (Repeat)									
		11a	**11b**	**12a**	**12b**	**13a**	**13b**	**14a**	**14b**	**15a**	**15b**
0.08	0.08	5.09E-10	6.94E-11	1.16E-10	6.57E-11	7.59E-11	3.00E-10	4.64E-11	1.07E-10	2.30E-09	1.45E-09
0.92	1	3.75E-11	2.59E-11	4.13E-11	6.35E-11	7.16E-11	5.37E-11	1.87E-11	1.65E-11	9.35E-10	5.62E-10
1	2	4.43E-11	3.62E-11	4.47E-11	6.11E-11	2.20E-11	2.62E-11	1.94E-11	1.80E-11	***3.01E-10***	2.20E-10
5	7	1.90E-11	1.75E-11	4.56E-11	5.31E-11	2.26E-11	2.89E-11	1.32E-11	1.47E-11	1.78E-10	1.86E-10
7	14	***2.95E-11***	***4.00E-11***	***3.77E-11***	4.53E-11	5.13E-11	5.34E-11	9.07E-12	1.24E-11	1.25E-10	1.33E-10
14	28	***5.02E-11***	***4.42E-11***	3.91E-11	4.19E-11	7.94E-11	6.46E-11	7.03E-12	1.12E-11	5.98E-11	6.16E-11
14	42	8.84E-11	1.08E-10	4.10E-11	4.05E-11	9.71E-11	7.90E-11	8.44E-12	1.13E-11	4.27E-11	4.80E-11
7	49	1.84E-10	1.81E-10	4.13E-11	4.44E-11	9.73E-11	8.59E-11	9.40E-12	1.17E-11	4.37E-11	4.55E-11
14	63	1.97E-10	***1.89E-10***	4.46E-11	4.33E-11	8.46E-11	7.42E-11	7.54E-12	1.02E-11	3.43E-11	3.36E-11
14	77	8.95E-11	1.11E-10	3.36E-11	4.51E-11	6.10E-11	5.11E-11	7.28E-12	7.33E-12	1.73E-11	2.86E-11
14	91	7.67E-11	1.11E-10	3.81E-11	2.24E-11	5.73E-11	5.01E-11	5.35E-12	5.78E-12	1.81E-11	1.34E-11

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm²/s) Mix Number and Duplicate (Repeat)									
		16a	**16b**	**17a**	**17b**	**18a**	**18b**	**19a**	**19b**	**20a**	**20b**
0.08	0.08	2.03E-10	3.03E-10	3.52E-10	3.48E-10	5.87E-11	5.19E-11	1.80E-11	1.25E-11	1.31E-11	7.21E-13
0.92	1	1.76E-10	2.95E-10	6.97E-11	6.94E-11	3.06E-11	1.44E-11	2.48E-12	2.06E-12	2.93E-11	3.27E-12
1	2	1.44E-10	2.18E-10	1.32E-10	1.09E-10	3.32E-11	1.57E-11	1.57E-12	1.67E-12	3.19E-11	8.34E-12
5	7	1.47E-10	***1.68E-10***	1.47E-10	1.74E-10	2.42E-11	1.35E-11	1.72E-12	2.11E-12	4.17E-11	2.46E-11
7	14	1.67E-10	***1.78E-10***	1.89E-10	1.72E-10	7.64E-12	6.15E-12	3.83E-12	4.54E-12	4.73E-11	3.43E-11
14	28	1.37E-10	***1.37E-10***	1.53E-10	1.36E-10	4.42E-12	4.44E-12	1.08E-11	1.18E-11	3.68E-11	3.03E-11
14	42	1.50E-10	***1.62E-10***	1.89E-10	1.88E-10	4.80E-12	5.33E-12	3.82E-11	3.30E-11	4.42E-11	3.73E-11
7	49	1.74E-10	***1.84E-10***	2.20E-10	2.02E-10	6.19E-12	6.56E-12	6.64E-11	6.44E-11	4.80E-11	3.74E-11
14	63	1.54E-10	***1.60E-10***	1.61E-10	1.59E-10	6.88E-12	6.75E-12	7.26E-11	6.81E-11	3.60E-11	3.22E-11
14	77	6.39E-11	1.08E-10	1.52E-10	1.45E-10	6.18E-12	7.15E-12	4.88E-11	4.94E-11	2.43E-11	2.71E-11
14	91	1.07E-10	1.02E-10	1.29E-10	1.13E-10	5.42E-12	8.14E-12	3.87E-11	4.10E-11	1.96E-11	2.60E-11

Table C.5. Technetium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		21a	**21b**	**22a**	**22b**	**23a**	**23b**
0.08	0.08	1.02E-08	3.05E-09	2.90E-09	2.37E-09	1.72E-09	4.67E-09
0.92	1	8.06E-09	9.79E-10	1.30E-09	4.32E-10	1.30E-09	3.88E-09
1	2	5.32E-10	1.19E-10	1.28E-10	1.49E-11	4.84E-10	1.17E-09
5	7	1.73E-11	1.44E-11	2.91E-11	9.59E-12	3.78E-10	4.58E-10
7	14	2.18E-11	1.41E-11	1.43E-11	1.29E-11	***7.17E-10***	***4.03E-10***
14	28	2.69E-11	1.97E-11	1.37E-11	1.68E-11	6.63E-10	***4.30E-10***
14	42	5.02E-11	4.61E-11	2.01E-11	2.62E-11	1.11E-09	6.90E-10
7	49	1.02E-10	1.61E-10	2.60E-11	3.49E-11	1.91E-09	1.19E-09
14	63	1.20E-10	2.27E-10	2.72E-11	3.36E-11	1.24E-09	***8.69E-10***
14	77	9.19E-11	2.26E-10	2.47E-11	1.64E-11	7.59E-10	6.01E-10
14	91	1.73E-10	3.52E-10	2.90E-11	1.43E-11	4.12E-10	3.98E-10

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		24a	**24b**	**25a**	**25b**	**26a**	**26b**
0.08	0.08	1.80E-09	1.21E-09	3.77E-09	4.57E-09	1.37E-09	1.48E-09
0.92	1	6.65E-10	3.64E-10	1.32E-09	1.75E-09	9.27E-11	6.80E-11
1	2	1.84E-10	6.91E-11	2.94E-10	5.17E-10	7.72E-12	5.86E-12
5	7	1.06E-10	3.99E-11	9.28E-11	1.78E-10	8.60E-12	8.44E-12
7	14	5.71E-11	4.25E-11	3.76E-11	3.44E-11	2.01E-11	1.32E-11
14	28	5.80E-11	5.37E-11	3.39E-11	2.51E-11	4.20E-11	3.41E-11
14	42	6.43E-11	6.44E-11	3.56E-11	2.48E-11	1.45E-10	1.51E-10
7	49	7.19E-11	6.96E-11	3.64E-11	2.56E-11	2.96E-10	2.51E-10
14	63	7.28E-11	6.88E-11	2.88E-11	2.05E-11	2.20E-10	1.89E-10
14	77	4.81E-11	6.29E-11	1.69E-10	2.10E-11	1.26E-11	6.09E-11
14	91	4.45E-11	6.57E-11	1.65E-11	1.16E-11	1.02E-10	1.49E-10

Table C.6. Chromium Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	6.59E-12	1.47E-09	1.14E-11	1.16E-11	2.30E-12	6.63E-13	1.10E-13	4.58E-13	1.79E-12	2.92E-12
0.92	1	3.76E-11	2.74E-11	2.25E-12	2.57E-12	3.10E-12	2.91E-12	8.12E-14	1.66E-13	1.90E-13	2.02E-12
1	2	9.19E-12	7.82E-12	1.05E-12	1.09E-12	8.56E-13	9.59E-13	1.42E-13	1.42E-13	2.17E-13	2.62E-12
5	7	3.01E-12	2.60E-12	4.43E-13	4.42E-13	2.74E-13	3.12E-13	9.58E-14	9.96E-14	5.77E-14	5.59E-13
7	14	6.76E-13	5.99E-13	1.96E-13	1.92E-13	2.20E-13	2.44E-13	7.27E-14	6.20E-14	4.93E-14	1.14E-13
14	28	***2.83E-13***	2.42E-13	1.15E-13	1.14E-13	1.68E-13	1.66E-13	***6.10E-14***	6.52E-14	4.37E-14	6.32E-14
14	42	1.53E-13	1.37E-13	7.04E-14	7.12E-14	1.57E-13	1.50E-13	4.33E-14	4.86E-14	5.77E-14	6.11E-14
7	49	1.91E-13	1.65E-13	8.38E-14	7.31E-14	2.31E-13	2.15E-13	***6.65E-14***	7.51E-14	1.16E-13	1.19E-13
14	63	1.64E-13	1.64E-13	6.32E-14	6.73E-14	1.09E-13	1.16E-13	***6.88E-14***	7.44E-14	9.14E-14	8.97E-14
14	77	1.66E-13	1.74E-13	4.59E-14	5.62E-14	1.16E-13	***8.63E-14***	5.26E-14	5.68E-14	8.16E-14	***4.37E-14***
14	91	1.79E-13	1.68E-13	5.02E-14	3.05E-14	1.06E-13	9.14E-14	5.85E-14	6.27E-14	6.16E-14	***8.09E-14***

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	5.72E-13	2.44E-13	<3.82E-14	<4.00E-14	7.57E-10	2.36E-10	2.14E-13	1.42E-11	4.31E-11	4.56E-11
0.92	1	9.85E-14	6.92E-14	2.81E-14	3.19E-14	2.83E-11	4.44E-11	7.08E-13	1.78E-12	6.77E-12	1.23E-11
1	2	8.63E-14	7.43E-14	2.29E-14	3.13E-14	5.22E-12	1.23E-11	1.06E-12	1.87E-12	5.69E-12	1.26E-11
5	7	***4.81E-14***	4.78E-14	1.11E-14	1.61E-14	1.17E-12	1.33E-12	4.57E-13	7.04E-13	***1.94E-12***	4.71E-12
7	14	***6.42E-14***	6.03E-14	7.17E-15	6.74E-15	7.84E-13	9.84E-13	3.27E-13	4.27E-13	***8.36E-13***	2.19E-12
14	28	***8.41E-14***	7.83E-14	6.36E-15	6.49E-15	6.52E-13	8.23E-13	1.67E-13	2.16E-13	***5.09E-13***	1.42E-12
14	42	1.15E-13	1.01E-13	5.32E-15	5.54E-15	5.55E-13	6.25E-13	1.30E-13	1.72E-13	***2.98E-13***	9.18E-13
7	49	2.73E-13	1.83E-13	1.00E-14	1.14E-14	7.39E-13	8.51E-13	1.72E-13	2.00E-13	3.38E-13	9.56E-13
14	63	***1.93E-13***	1.96E-13	1.01E-14	1.11E-14	6.90E-13	7.82E-13	9.97E-14	1.20E-13	3.21E-13	8.31E-13
14	77	8.48E-14	1.28E-13	6.13E-15	8.29E-15	5.73E-13	6.43E-13	5.58E-14	4.62E-14	2.30E-13	5.08E-13
14	91	1.11E-13	1.56E-13	7.93E-15	1.13E-14	2.96E-13	5.14E-13	6.05E-14	8.33E-14	2.18E-13	5.69E-13

Table C.6. Chromium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	1.42E-11	1.84E-13	1.70E-12	8.95E-13	8.13E-13	1.38E-12	1.12E-13	2.37E-13	4.49E-12	3.60E-12
0.92	1	6.89E-13	9.16E-14	1.05E-13	1.13E-13	5.99E-13	3.11E-13	6.68E-14	4.76E-14	3.70E-12	3.04E-12
1	2	4.87E-13	1.34E-13	1.90E-13	1.67E-13	5.14E-13	2.48E-13	4.62E-14	3.30E-14	***5.22E-12***	4.17E-12
5	7	1.56E-13	9.75E-14	1.44E-13	1.75E-13	2.04E-13	1.20E-13	1.87E-14	9.79E-15	***5.02E-12***	4.15E-12
7	14	***8.80E-14***	***1.00E-13***	1.15E-13	2.63E-13	1.00E-13	7.64E-14	9.73E-15	9.00E-15	3.03E-12	2.48E-12
14	28	1.08E-13	1.02E-13	1.09E-13	1.18E-13	6.77E-14	5.51E-14	8.49E-15	8.72E-15	9.09E-13	6.84E-13
14	42	1.07E-13	1.08E-13	1.10E-13	1.13E-13	4.82E-14	3.91E-14	6.88E-15	7.55E-15	2.24E-13	1.59E-13
7	49	1.89E-13	1.80E-13	2.14E-13	5.54E-13	5.38E-14	6.54E-14	1.20E-14	1.11E-14	1.93E-13	1.53E-13
14	63	1.39E-13	1.41E-13	2.21E-13	1.77E-13	5.61E-14	5.54E-14	1.27E-14	1.18E-14	1.10E-13	8.98E-14
14	77	6.03E-14	7.88E-14	1.28E-13	1.70E-13	5.24E-14	4.53E-14	1.57E-14	1.23E-14	1.17E-13	1.04E-13
14	91	5.29E-14	7.67E-14	1.58E-13	8.14E-14	5.16E-14	3.99E-14	1.02E-14	1.05E-14	7.13E-14	4.62E-14

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		16a	16b	17a	17b	18a	18b	19a	19b	20a	20b
0.08	0.08	1.06E-10	7.71E-11	1.06E-13	3.03E-13	<1.20E-13	<1.17E-13	3.79E-14	<3.37E-14	3.76E-12	4.34E-13
0.92	1	1.81E-11	1.48E-11	9.96E-14	1.16E-13	<1.98E-14	<1.93E-14	4.22E-14	2.97E-14	1.54E-12	3.66E-13
1	2	2.72E-12	2.89E-12	1.33E-13	1.29E-13	<5.83E-14	<5.68E-14	3.79E-14	2.83E-14	9.00E-13	2.72E-13
5	7	8.91E-13	***1.03E-12***	1.53E-13	1.71E-13	2.21E-14	2.08E-14	2.83E-14	2.93E-14	3.35E-13	1.52E-13
7	14	4.24E-13	***5.80E-13***	1.87E-13	1.76E-13	1.28E-14	1.28E-14	3.55E-14	3.65E-14	1.69E-13	1.12E-13
14	28	2.73E-13	***3.27E-13***	1.85E-13	1.74E-13	1.56E-14	1.87E-14	4.99E-14	5.07E-14	1.07E-13	8.59E-14
14	42	1.94E-13	***2.12E-13***	1.59E-13	1.47E-13	8.25E-15	8.43E-15	4.82E-14	5.01E-14	7.18E-14	6.15E-14
7	49	2.80E-13	***2.95E-13***	2.07E-13	1.75E-13	<3.72E-14	<3.62E-14	6.95E-14	8.20E-14	8.99E-14	8.25E-14
14	63	1.88E-13	***1.93E-13***	1.52E-13	1.25E-13	<1.14E-14	<1.11E-14	5.55E-14	5.99E-14	5.11E-14	5.56E-14
14	77	1.40E-13	***2.08E-13***	2.00E-13	1.83E-13	2.85E-14	3.18E-14	7.70E-14	8.27E-14	7.54E-14	9.02E-14
14	91	1.89E-13	1.77E-13	1.87E-13	1.47E-13	2.14E-14	2.58E-14	6.92E-14	7.86E-14	4.54E-14	7.53E-14

Table C.6. Chromium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		21a	**21b**	**22a**	**22b**	**23a**	**23b**
0.08	0.08	3.60E-13	6.47E-13	8.69E-12	2.47E-12	5.78E-13	1.48E-13
0.92	1	2.87E-13	3.38E-13	5.03E-12	1.68E-12	2.51E-13	1.99E-13
1	2	1.17E-13	1.35E-13	3.38E-12	1.30E-12	6.04E-13	2.71E-13
5	7	4.89E-14	4.60E-14	1.15E-12	5.89E-13	6.17E-13	3.84E-13
7	14	5.52E-14	5.67E-14	4.37E-13	3.42E-13	***8.98E-13***	***5.83E-13***
14	28	6.98E-14	8.15E-14	2.21E-13	1.94E-13	***9.16E-13***	6.96E-13
14	42	1.03E-13	1.09E-13	1.93E-13	1.62E-13	1.01E-12	***8.71E-13***
7	49	1.99E-13	2.14E-13	2.51E-13	2.16E-13	***1.80E-12***	1.55E-12
14	63	2.09E-13	2.23E-13	1.08E-13	8.87E-14	1.24E-12	1.19E-12
14	77	1.72E-13	1.91E-13	1.41E-13	6.91E-14	9.21E-13	9.47E-13
14	91	2.39E-13	2.35E-13	1.26E-13	4.48E-14	5.10E-13	5.82E-13

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		24a	**24b**	**25a**	**25b**	**26a**	**26b**
0.08	0.08	3.19E-12	1.35E-12	1.00E-11	1.34E-11	2.24E-12	2.55E-12
0.92	1	2.46E-12	9.52E-13	5.50E-12	8.39E-12	6.27E-13	5.91E-13
1	2	1.95E-12	6.93E-13	6.43E-12	9.43E-12	2.91E-13	2.73E-13
5	7	7.03E-13	2.48E-13	4.86E-12	8.27E-12	6.85E-14	6.40E-14
7	14	2.67E-13	1.24E-13	1.99E-12	3.08E-12	5.61E-14	4.21E-14
14	28	1.73E-13	8.27E-14	7.98E-13	1.30E-12	6.22E-14	5.66E-14
14	42	8.95E-14	5.65E-14	2.14E-13	2.70E-13	7.22E-14	7.28E-14
7	49	1.19E-13	8.98E-14	1.87E-13	1.95E-13	1.80E-13	1.72E-13
14	63	8.37E-14	6.13E-14	1.28E-13	1.20E-13	9.97E-14	9.58E-14
14	77	9.01E-14	1.08E-13	1.45E-13	1.21E-13	8.93E-14	5.80E-14
14	91	7.32E-14	9.99E-14	9.63E-14	7.87E-14	9.64E-14	1.37E-13

Table C.7. Uranium Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		1a	**1b**	**2a**	**2b**	**3a**	**3b**	**4a**	**4b**	**5a**	**5b**
0.08	0.08	3.02E-15	6.93E-13	5.96E-15	6.62E-15	2.76E-15	9.99E-16	4.66E-16	5.20E-15	1.05E-14	2.46E-15
0.92	1	4.98E-16	4.99E-16	2.81E-16	2.92E-16	6.22E-17	6.58E-17	1.92E-16	1.15E-16	4.68E-17	9.08E-17
1	2	2.33E-14	2.68E-16	3.47E-16	1.96E-16	<5.79E-17	<5.96E-17	1.93E-16	<1.95E-16	1.06E-16	7.25E-17
5	7	8.67E-17	8.16E-17	1.03E-16	9.04E-17	5.62E-16	<1.51E-17	2.21E-16	2.23E-15	7.72E-17	4.94E-17
7	14	9.84E-17	1.48E-16	1.11E-16	9.98E-17	<1.88E-17	<1.94E-17	1.05E-16	<6.32E-17	<2.35E-17	<2.35E-17
14	28	***9.83E-17***	1.12E-16	7.64E-17	6.51E-17	<9.30E-18	<9.57E-18	***<3.09E-17***	<3.13E-17	<1.16E-17	<1.16E-17
14	42	1.86E-16	1.97E-16	6.04E-17	5.58E-17	<1.58E-17	<1.62E-17	***<5.25E-17***	<5.30E-17	<1.97E-17	<1.97E-17
7	49	5.59E-16	6.66E-16	<2.16E-16	<2.11E-16	<8.26E-17	<8.50E-17	***<2.75E-16***	<2.78E-16	<1.03E-16	<1.03E-16
14	63	3.15E-16	3.37E-16	<6.65E-17	<6.52E-17	<2.55E-17	<2.62E-17	<8.47E-17	<8.56E-17	<3.19E-17	<3.19E-17
14	77	3.84E-16	4.39E-16	<8.31E-17	<8.14E-17	<3.18E-17	***<3.28E-17***	<1.06E-16	<1.07E-16	<3.98E-17	***<3.98E-17***
14	91	5.22E-16	5.06E-16	<9.89E-17	<9.69E-17	<3.79E-17	<3.90E-17	<1.26E-16	<1.27E-16	<4.74E-17	***<4.74E-17***

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		6a	**6b**	**7a**	**7b**	**8a**	**8b**	**9a**	**9b**	**10a**	**10b**
0.08	0.08	6.72E-15	2.93E-15	<1.52E-16	5.67E-16	6.00E-13	6.20E-14	<1.15E-16	5.33E-16	5.82E-14	3.49E-13
0.92	1	1.40E-16	1.04E-16	4.43E-17	3.88E-17	1.63E-15	2.05E-15	<1.89E-17	<1.87E-17	4.16E-16	4.16E-16
1	2	<7.81E-17	<7.94E-17	<7.38E-17	4.71E-14	1.25E-15	1.32E-15	<5.58E-17	<5.50E-17	2.25E-16	1.38E-13
5	7	3.04E-17	6.43E-14	7.02E-16	2.73E-17	2.04E-16	2.26E-16	<1.41E-17	1.72E-17	***7.96E-17***	8.95E-17
7	14	***3.94E-17***	2.94E-17	<2.40E-17	<2.52E-17	2.80E-16	2.73E-16	<1.82E-17	<1.79E-17	***6.05E-17***	8.30E-17
14	28	1.52E-17	1.55E-17	1.42E-17	1.28E-17	1.83E-16	1.89E-16	<8.96E-18	<8.83E-18	4.68E-17	4.79E-17
14	42	<2.13E-17	<2.17E-17	<2.01E-17	<2.11E-17	2.32E-16	2.14E-16	<1.52E-17	<1.50E-17	<3.70E-17	5.53E-17
7	49	2.17E-16	<1.13E-16	<1.05E-16	<1.10E-16	1.03E-15	9.41E-16	<7.96E-17	<7.85E-17	<1.94E-16	<1.96E-16
14	63	<3.44E-17	<3.49E-17	<3.25E-17	<3.40E-17	6.00E-16	4.68E-16	<2.46E-17	<2.42E-17	<5.97E-17	<6.06E-17
14	77	<4.29E-17	<4.36E-17	<4.05E-17	<4.25E-17	5.84E-16	7.19E-16	<3.06E-17	<3.02E-17	***<7.46E-17***	<7.56E-17
14	91	<5.12E-17	<5.20E-17	<4.84E-17	<5.07E-17	3.37E-16	8.98E-16	<3.66E-17	<3.61E-17	***<8.90E-17***	<9.02E-17

Table C.7. Uranium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	6.40E-15	1.56E-15	5.76E-15	3.63E-15	1.07E-15	4.61E-15	8.72E-16	3.47E-15	5.89E-16	5.87E-16
0.92	1	2.46E-17	2.72E-17	<4.57E-17	<4.63E-17	2.34E-16	2.91E-16	2.82E-17	3.52E-17	5.56E-17	5.29E-17
1	2	<6.05E-17	<5.96E-17	<1.35E-16	<1.37E-16	2.08E-16	2.37E-16	<7.37E-17	<7.58E-17	***<7.45E-17***	<7.42E-17
5	7	<1.53E-17	<1.51E-17	<3.41E-17	<3.45E-17	1.36E-16	1.28E-16	<1.87E-17	<1.92E-17	<1.90E-17	<1.90E-17
7	14	***<1.97E-17***	***<1.94E-17***	***<4.38E-17***	8.94E-17	1.09E-16	1.10E-16	2.57E-17	<2.47E-17	<2.40E-17	<2.39E-17
14	28	***<9.71E-18***	***<9.56E-18***	<2.16E-17	<2.19E-17	7.14E-17	6.65E-17	1.43E-17	<1.22E-17	<1.20E-17	<1.20E-17
14	42	<1.65E-17	***<1.63E-17***	***<3.68E-17***	4.46E-17	4.93E-17	5.02E-17	2.33E-17	<2.07E-17	<2.04E-17	<2.03E-17
7	49	<8.63E-17	<8.50E-17	<1.92E-16	1.09E-15	<2.22E-16	<2.23E-16	<1.05E-16	<1.08E-16	1.08E-16	<1.07E-16
14	63	<2.66E-17	<2.62E-17	***<5.93E-17***	6.27E-17	<6.83E-17	<6.87E-17	<3.24E-17	<3.33E-17	8.09E-17	<3.27E-17
14	77	<3.32E-17	<3.27E-17	<7.39E-17	9.60E-17	<8.52E-17	<8.56E-17	<4.04E-17	<4.16E-17	7.06E-17	<4.09E-17
14	91	<3.97E-17	<3.91E-17	<8.84E-17	<8.96E-17	<1.02E-16	<1.02E-16	<4.84E-17	<4.97E-17	6.73E-17	<4.90E-17

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm^2/s)									
		Mix Number and Duplicate (Repeat)									
		16a	16b	17a	17b	18a	18b	19a	19b	20a	20b
0.08	0.08	7.80E-15	4.92E-15	7.06E-16	6.02E-16	6.54E-16	5.27E-16	3.66E-16	3.26E-16	<3.45E-16	<3.37E-16
0.92	1	5.23E-16	5.12E-16	1.44E-16	1.33E-16	9.37E-17	8.95E-17	5.84E-17	4.62E-17	3.31E-16	7.66E-17
1	2	3.43E-16	2.37E-16	1.45E-16	<1.38E-16	1.43E-16	1.21E-16	<7.87E-17	<7.83E-17	8.07E-16	3.60E-16
5	7	6.77E-17	***6.84E-17***	7.10E-17	1.03E-16	3.88E-17	2.90E-17	<2.01E-17	<2.00E-17	1.05E-15	1.80E-16
7	14	9.93E-17	***7.97E-17***	1.33E-16	1.02E-16	8.02E-17	7.81E-17	<2.53E-17	<2.52E-17	<5.38E-17	<5.25E-17
14	28	3.91E-17	***1.21E-16***	5.00E-17	5.06E-17	7.32E-17	1.27E-16	<1.27E-17	<1.19E-17	<2.69E-17	<2.63E-17
14	42	6.08E-17	***7.49E-17***	<3.89E-17	<3.79E-17	1.28E-16	1.60E-16	<2.16E-17	<2.15E-17	<4.58E-17	<4.47E-17
7	49	1.71E-16	***2.81E-16***	<2.04E-16	<1.98E-16	2.34E-16	6.38E-16	<1.13E-16	<1.12E-16	<2.40E-16	<2.34E-16
14	63	1.10E-16	***1.36E-16***	<6.26E-17	<6.09E-17	1.75E-16	4.48E-16	<3.47E-17	<3.46E-17	<7.36E-17	<7.20E-17
14	77	9.64E-17	1.74E-16	<7.83E-17	<7.62E-17	2.05E-16	8.84E-16	<4.34E-17	<4.32E-17	<9.21E-17	<9.00E-17
14	91	1.77E-16	1.81E-16	<9.37E-17	<9.12E-17	2.25E-16	6.47E-16	<5.20E-17	<5.17E-17	<1.10E-16	6.22E-16

Table C.7. Uranium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	2.41E-15	1.32E-15	1.67E-16	1.40E-16	2.73E-15	3.57E-15
0.92	1	1.94E-16	1.72E-16	2.72E-17	<1.96E-17	2.66E-16	7.47E-16
1	2	1.01E-16	7.51E-17	<5.67E-17	<5.77E-17	3.63E-16	6.59E-16
5	7	3.70E-17	3.36E-17	<1.44E-17	1.19E-16	1.42E-16	2.31E-16
7	14	5.43E-17	4.85E-17	<1.81E-17	3.90E-17	***2.54E-16***	***3.67E-16***
14	28	3.79E-17	3.36E-17	<9.12E-18	<9.29E-18	***2.19E-16***	2.62E-16
14	42	2.39E-17	3.19E-17	<1.55E-17	<1.57E-17	2.20E-16	3.39E-16
7	49	<8.40E-17	<8.47E-17	<8.12E-17	<8.28E-17	***6.16E-16***	1.90E-15
14	63	<2.58E-17	2.99E-17	<2.49E-17	<2.54E-17	2.12E-16	4.83E-16
14	77	<3.22E-17	<3.25E-17	<3.11E-17	<3.17E-17	1.82E-16	2.64E-16
14	91	<3.86E-17	<3.89E-17	<3.73E-17	<3.80E-17	***8.99E-17***	2.91E-16

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm^2/s)					
		Mix Number and Duplicate (Repeat)					
		24a	24b	25a	25b	26a	26b
0.08	0.08	1.84E-15	2.41E-15	9.83E-16	1.04E-15	9.47E-16	1.23E-15
0.92	1	1.58E-16	1.80E-16	6.21E-17	6.94E-17	4.90E-17	4.08E-17
1	2	<1.39E-16	<1.43E-16	<7.47E-17	<7.55E-17	7.27E-17	<5.78E-17
5	7	3.79E-17	<3.65E-17	<1.90E-17	<1.93E-17	<1.48E-17	<1.47E-17
7	14	<4.45E-17	<4.58E-17	<2.39E-17	2.54E-17	<1.86E-17	<1.85E-17
14	28	<2.53E-17	<2.71E-17	<1.20E-17	1.63E-17	<9.36E-18	<9.29E-18
14	42	<3.79E-17	<3.90E-17	<2.04E-17	<2.06E-17	<1.59E-17	<1.58E-17
7	49	<1.99E-16	<2.05E-16	<1.07E-16	<1.08E-16	<8.34E-17	<8.28E-17
14	63	<6.11E-17	<6.29E-17	<3.29E-17	<3.32E-17	<2.56E-17	<2.54E-17
14	77	<7.64E-17	<7.86E-17	<4.10E-17	<4.15E-17	<3.20E-17	<3.17E-17
14	91	<9.16E-17	<9.43E-17	<4.92E-17	<4.98E-17	<3.83E-17	<3.81E-17

Distribution*

Washington River Protection Solutions
DJ Swanberg
WRPS Documents – TOCVND@rl.gov

Oak Ridge National Laboratory
EM Pierce

Savannah River National Laboratory
AD Cozzi
KM Fox
DI Kaplan
CA Langton
DJ McCabe

New Jersey City University
Department of Geoscience and Geography
2039 Kennedy Boulevard
Jersey City, NJ 07305
HB Jung

Pacific Northwest National Laboratory
M Asmussen
DH Bacon
VL Freedman
GV Last
JJ Neeway
NP Qafoku
RJ Serne
GL Smith
MMV Snyder
W Um
G Wang
JH Westsik, Jr.
BD Williams
SB Yabusaki
Project File
Information Release (pdf)

*All distribution will be made electronically.

Pacific Northwest NATIONAL LABORATORY

U.S. DEPARTMENT OF ENERGY

Proudly Operated by **Battelle** *Since 1965*

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99352
1-888-375-PNNL (7665)

www.pnnl.gov