

# Cast Stone Oxidation Front Evaluation: Preliminary Results for Samples Exposed to Moist Air

C.A. Langton and P.M. Almond

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## **PREFACE OR ACKNOWLEDGEMENTS**

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## EXECUTIVE SUMMARY

The rate of oxidation is important to the long-term performance of reducing salt waste forms because the solubility of some contaminants, e.g., technetium, is a function of oxidation state.  $\text{TcO}_4^-$  in the salt solution is reduced to Tc(IV) and has been shown to react with ingredients in the waste form to precipitate low solubility sulfide and/or oxide phases. Upon exposure to oxygen, the compounds containing Tc(IV) oxidize to the pertechnetate ion,  $\text{Tc(VII)O}_4^-$ , which is very soluble. Consequently the rate of technetium oxidation front advancement into a monolith and the technetium leaching profile as a function of depth from an exposed surface are important to waste form performance and ground water concentration predictions.

An approach for measuring contaminant oxidation rate (effective contaminant specific oxidation rate) based on leaching of select contaminants of concern is described in this report. In addition, the relationship between reduction capacity and contaminant oxidation is addressed.

Chromate (Cr(VI)) was used as a non-radioactive surrogate for pertechnetate, Tc(VII), in Cast Stone samples prepared with 5 M Simulant. Cast Stone spiked with pertechnetate was also prepared and tested. Depth discrete subsamples spiked with Cr were cut from Cast Stone exposed to Savannah River Site (SRS) outdoor ambient temperature fluctuations and moist air. Depth discrete subsamples spiked with Tc-99 were cut from Cast Stone exposed to laboratory ambient temperature fluctuations and moist air. Similar conditions are expected to be encountered in the Cast Stone curing container. The leachability of Cr and Tc-99 and the reduction capacities, measured by the Angus-Glasser method, were determined for each subsample as a function of depth from the exposed surface.

The results obtained to date were focused on continued method development and are preliminary and apply to the sample composition and curing / exposure conditions described in this report.

- The Cr oxidation front (depth to which soluble Cr was detected) for the Cast Stone sample exposed for 68 days to ambient outdoor temperatures and humid air (total age of sample was 131 days) was determined to be about 35 mm below the top sample surface exposed. The Tc oxidation front, depth at which Tc was insoluble, was not determined. Interpretation of the results indicates that the oxidation front is at least 38 mm below the exposed surface. The sample used for this measurement was exposed to ambient laboratory conditions and humid air for 50 days. The total age of the sample was 98 days.
- Technetium appears to be more easily oxidized than Cr in the Cast Stone matrix. The oxidized forms of Tc and Cr are soluble and therefore leachable. Longer exposure times are required for both the Cr and Tc spiked samples to better interpret the rate of oxidation. Tc spiked subsamples need to be taken further from the exposed surface to better define and interpret the leachable Tc profile.
- Finally Tc(VII) reduction to Tc(IV) appears to occur relatively fast. Results demonstrated that about 95 percent of the Tc(VII) was reduced to Tc(IV) during the setting and very early stage setting for a Cast Stone sample cured 10 days. Additional testing at longer curing times is required to determine whether additional time is required to reduce 100 % of the Tc(VII) in Cast Stone or whether the Tc loading exceeded the ability of the waste form to reduce 100 % of the Tc(VII). Additional testing is required for samples cured for longer times. Depth discrete subsampling in a nitrogen glove box is also required to determine whether the 5 percent Tc extracted from the subsamples was the result of the sampling process which took place in air.

Reduction capacity measurements (per the Angus-Glasser method) performed on depth discrete samples could not be correlated with the amount of chromium or technetium leached from the depth discrete subsamples or with the oxidation front inferred from soluble chromium and technetium (i.e., effective Cr

and Tc oxidation fronts). Residual reduction capacity in the oxidized region of the test samples indicates that the remaining reduction capacity is not effective in re-reducing Cr(VI) or Tc(VII) in the presence of oxygen.

Depth discrete sampling and leaching is a useful for evaluating Cast Stone and other chemically reducing waste forms containing ground granulated blast furnace slag (GGBFS) or other reduction / sequestration reagents to control redox sensitive contaminant chemistry and leachability in the near surface disposal environment.

Based on results presented in this report, reduction capacity measured by the Angus-Glasser Ce(IV) method is not an appropriate or meaningful parameter for determining or predicting Tc and Cr oxidation / retentions, speciation, or solubilities in cementitious materials such as Cast Stone. A model for predicting Tc(IV) oxidation to soluble Tc(VII) should consider the waste form porosity (pathway for oxygen ingress), oxygen source, and the contaminant specific oxidation rates and oxidation fronts.

Depth discrete sampling of materials exposed to realistic conditions in combination with short term leaching of crushed samples has potential for advancing the understanding of factors influencing performance. This information can be used to support conceptual model development.

## TABLE OF CONTENTS

EXECUTIVE SUMMARY .....	v
LIST OF TABLES .....	viii
LIST OF FIGURES .....	ix
LIST OF ABBREVIATIONS .....	x
1.0 Introduction .....	1
1.1 Background .....	1
1.2 Objectives .....	2
1.3 Approach .....	2
1.4 Quality Assurance .....	2
2.0 Experimental Procedure .....	2
2.1 Cast Stone Sample Preparation .....	2
2.1.1 Simulant .....	2
2.1.2 Cast Stone Premix Reagents .....	3
2.1.3 Cast Stone Spiked with Chromium .....	4
2.1.4 Cast Stone Spiked with Technetium .....	5
2.2 Cast Stone Depth Discrete Sub-Sampling .....	6
2.3 Cast Stone Depth Discrete Sub-Sample Leaching .....	7
2.4 Reduction Capacity Measurements .....	7
2.5 Evaluation of Tc Reduction Kinetics in Cast Stone .....	8
2.5.1 Cast Stone Sample Preparation (10 Day Old Sample) .....	8
2.5.2 10 Day Old Depth Discrete Subsample Preparation and Leaching .....	8
3.0 Results and Discussion .....	10
3.1 Leaching Result for Cast Stone Spiked with Chromium .....	10
3.2 Leaching Result for Cast Stone Spiked with Technetium-99 .....	14
3.3 Comparison of Cr, Tc, Na, and NO <sub>3</sub> <sup>-</sup> Leaching Results .....	18
3.4 Leaching Results for 10 Day Old Cast Stone Spiked with Tc-99 .....	19
3.5 Reduction Capacity Result .....	21
3.5.1 Reduction Capacity of Starting Materials and Fresh Cast Stone .....	21
3.5.2 Reduction Capacity of Depth Discrete Cast Stone Samples .....	22
4.0 Conclusions .....	25
5.0 Recommendations and Future Work .....	26
6.0 References .....	27
Appendix A . Analytical Data for 5 M Cast Stone Cr500-1 Depth Discrete Subsamples .....	1
Appendix B . Analytical Data for 5 M Cast Stone Tc2 Depth Discrete Subsamples .....	B-1

## LIST OF TABLES

Table 2-1. LAW 5 M Simulant Based on Hanford Tank Waste Operations Simulator (HTWOS) Modeling <sup>4</sup> .....	3
Table 2-2. Average 5M Simulant Composition. ....	3
Table 2-3. Ingredients in Chromium Spiked Cast Stone, w/cm = 0.60. ....	4
Table 2-4. Ingredients in Cast Stone Containing HTWOS Average 5M Na Simulant Spiked with Tc-99. ....	5
Table 2-5. Tc-99 Concentrations in Spiked 5 M Na HTWOS Average Simulant and Cast Stone Prepared with a w /cm of 0.60.....	8
Table 3-1. Cr Leaching Results for Cast Stone Mix Cr 500-1 Depth Discrete Subsamples.....	11
Table 3-2. Na Leaching Results for Cast Stone Mix Cr 500-1 Depth Discrete Subsamples. ....	12
Table 3-3. NO <sub>3</sub> <sup>-</sup> Leaching Results for Cast Stone Mix Cr 500-1 Depth Discrete Subsamples.....	13
Table 3-4. Tc-99 Leaching Results for Cast Stone Mix Tc-2 Depth Discrete Subsamples. ....	15
Table 3-5. Na Leaching Results for Cast Stone Mix Tc-2 Depth Discrete Subsamples. ....	16
Table 3-6. NO <sub>3</sub> <sup>-</sup> Leaching Results for Cast Stone Mix Tc-2 Depth Discrete Subsamples.....	17
Table 3-7. Comparison of Cr and Tc Leaching Results for Samples Exposed to Moist Air. ....	18
Table 3-8. Tc-99 Leaching Results for Depth Discrete Subsamples from Cast Stone Mix Tc 4-1 Cured for 10 days in Closed Container. ....	19
Table 3-9. Reduction Capacities of Cement, Slag, and Fly Ash Supplied by PNNL and 5 M Na HTWOS Average Simulant. ....	21
Table 3-10. Reduction Capacity Results for Cast Stone Cr 500-1 Depth Discrete Subsamples.....	23
Table 3-11. Reduction Capacity Results for Cast Stone Tc2 Depth Discrete Subsamples.....	24

## LIST OF FIGURES

Figure 2-1. Laboratory Curing Container (left); Cast Stone Sample After Curing for 30 Days Prior to Exposure Testing (center), Outdoor Exposure Container (right). .....	5
Figure 2-2. Sample Holders Utilized to Cut Depth Discrete Samples. ....	6
Figure 2-3. Depth discrete Cast Stone Sample Preparation. (a) Monolithic Cylinder in Sample Holder Prior to Sectioning, (b) Ground Depth Discrete Sample Prior to Leaching, (c) Leaching Samples Prior to Tumbling, (d) Leached Samples After Tumbling. ....	9
Figure 3-1. Cast Stone Cr 500-1 Leachate Results for $\text{NO}_3^-$ , Na, and Cr as a Function of Distance from the Top Exposed Surface. ....	14
Figure 3-2. Cast Stone Tc2 Leachate Results for Na and Tc-99 as a Function of Distance from the Top Exposed Surface. ....	18
Figure 3-3. Comparison of Tc-99 Leachability / Solubility as a Function of Distance from the Top Surface for a 10 Day Old and a 98 Day Old (exposed to air) Cast Stone Sample. ....	20
Figure 3-4. Reduction Capacity Results for Cr500-1 and Tc2 Depth Discrete Subsamples. ....	25

## LIST OF ABBREVIATIONS

AFm	Calcium ( $\text{Al}_2\text{O}_3\text{-Fe}_2\text{O}_3$ ) (mono = single formula unit of $\text{CaX}_2$ ) hydrate phases where $\text{X} = \text{OH}^-$ , $\text{SO}_4^{2-}$ , $\text{CO}_3^{2-}$ and other anions; another representation = $\text{Ca}_3(\text{Al}_2\text{O}_3, \text{Fe}_2\text{O}_3)\text{CaX}_2\text{yH}_2\text{O}$
ASTM	American Society for Testing and Materials
DOE	U.S. Department of Energy
EM	Office of Environmental Management (DOE)
EPA	U.S. Environmental Protection Agency
FAS	Ferrous Ammonium Sulfate
GC-MS	Gas Chromatography – Mass Spectrometry
GGBFS	Ground Granulated Blast Furnace Slag
HTWOS	Hanford Tank Waste Operations Simulator
IC	Ion Chromatography
IDF	Integrated Disposal Facility
ICP-OES	Inductively Coupled Plasma – Optical Emission Spectroscopy
LAW	Low Activity Waste
PNNL	Pacific Northwest National Laboratory
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
TTQAP	Task Technical and Quality Assurance Plan
WSRC	Westinghouse Savannah River Company
WTP	Hanford Tank Waste Treatment and Immobilization Plant
w/cm	Water / Total Cementitious Materials Water / (Cement + Slag + Fly Ash)

## 1.0 Introduction

### 1.1 Background

Low temperature waste forms are currently being used and considered for solidification of low-level radioactive wastes across the DOE complex. Examples include saltstone, a cementitious waste form used at the Savannah River Site (SRS) to immobilize low-activity sodium salt waste, and Cast Stone which was recently selected for solidification of secondary waste from the Hanford Tank Waste Treatment and Immobilization Plant (WTP). Cast Stone is also being considered to provide supplemental Low Activity Waste (LAW) immobilization capacity for the Hanford site.

The Cast Stone waste form and immobilization process must be tested to demonstrate compliance with the waste acceptance criteria for the Integrated Disposal Facility (IDF) and to demonstrate that the immobilization processes can be controlled to consistently provide an acceptable waste form product. Further, Cast Stone must be tested to provide the technical basis for understanding the long-term performance of the waste form in the IDF environment.

The Office of Environmental Management (EM) tasked the Savannah River National Laboratory (SRNL) and the Pacific Northwest National Laboratory (PNNL) to define and execute technology to reduce technical and programmatic risks with respect to investments in low-temperature waste forms and technetium separations for processing which leads to final disposition of Hanford waste streams.

This task addresses the technology gap, “Technetium Speciation and Impacts of Redox Changes” identified by PNNL and SRNL<sup>1</sup> and was authorized in the US DOE Inter-Entity Work Order MOSRV00091 deliverable 1(a) determine Tc oxidation behavior in Cast Stone by utilizing a depth discrete sampling and leaching method developed at SRNL.<sup>2</sup>

Both the SRS and Hanford waste streams contain soluble technetium which may require stabilization to meet disposal requirements. Technetium stabilization is a difficult problem because: 1) Tc is soluble and very mobile in the oxidized form typical of near surface environments and 2) Tc-99 is a long lived isotope with a half-life of  $2.1 \times 10^5$  years which places demanding performance requirements on the engineered barriers and environment to meet current regulatory disposal requirements.

Cast Stone and saltstone contain ground granulated blast furnace slag (GGBFS), which provides chemically reducing environment in the waste from matrix, in addition to portland cement and fly ash. All of these ingredients react with water and other constituents in the waste stream to form the waste form matrix. The GGBFS stabilizes redox sensitive contaminants such as Tc and Cr. In a high pH environment, GGBFS chemically reduces pertechnetate,  $\text{Tc(VII)O}_4^-$ , to the less soluble Tc(IV) oxidation state.<sup>a</sup> Over time, oxygen in the air, in sediment pore spaces, and dissolved in water moving through the vadose zone can oxidize the waste form and re-oxidize the Tc(IV) to the highly soluble Tc(VII) form.

An understanding of factors that affect the oxidation state of redox sensitive contaminants stabilized in cementitious waste forms is required to improve waste form designs and engineered barriers for shallow land disposal. In addition, the rate of bulk matrix oxidation and the relationship between the bulk reduction capacity and contaminant specific reduction and re-oxidation rates are parameters required for predicting long-term performance.

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<sup>a</sup> The fraction of pertechnetate chemically reduced to the less soluble Tc(IV) oxidation state depends on several factors including the: concentration, chemical form (inorganic or organic), and chemistry of the waste form and waste form pore solution.

## 1.2 Objectives

The objectives of the experimental work described in this report were to:

- Utilize depth discrete sampling and zero head space leaching to determine the contaminant specific oxidation rate for Tc in Cast Stone prepared with the Hanford Tank Waste Operations System (HTWOS) “Average” 5 M Na simulant.<sup>3, 4, 5</sup>
- Determine the relationship between the Cast Stone reduction capacity measured by the Angus-Glasser Method<sup>6</sup> and leachability of Tc from depth discrete samples.
- Provide preliminary data on the kinetics of Tc(VII) reduction in Cast Stone as a function of cure time.

## 1.3 Approach

This study is being performed in phases described in the Task Technical and Quality Assurance Plan<sup>7</sup> as funding is provided. The first set of experiments involves preparing and curing chromium and technetium spiked Cast Stone samples made with 5 M sodium Average LAW simulant and then exposing the cured samples to moist air for varying lengths of time up to about 1 year. Chromium was used in the initial testing as a non-radioactive redox sensitive surrogate for technetium. Subsamples, one to several mm thick, will be as a function of distance (depth) from the exposed surface, ground, and leached in deionized de-aerated water for 48 to 72 hours. Leachates will be analyzed for redox sensitive Cr and Tc-99 in addition to selected anions and cations. Results will be used to calculate the rate of contaminant specific oxidation front advancement.

The second set of experiments involves exposing spiked Cast Stone samples to moist Hanford soil to determine the rate of oxidation front advancement in soil representative of the Hanford IDF environment. The third part of the study involves evaluating the effect of fractures on the rate of oxidation front advancement in Cast Stone exposed to air and soil. The Task Technical and Quality Assurance Plan for this work is contained in SRNL-RP-2013-00112.<sup>7</sup>

## 1.4 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.<sup>14</sup>

# 2.0 Experimental Procedure

A method for measuring contaminant oxidation fronts for redox sensitive contaminants in cementitious waste forms containing GGBFS was recently developed at SRNL.<sup>8, 9</sup> This method is based upon leaching depth discrete sub-samples collected from a waste form surface exposed to air or oxygenated water. Leaching is performed in a zero head space container using deionized, de-aerated water to minimize oxidation during the leaching process. Details of the method are provided in another report.<sup>8, 9</sup>

## 2.1 Cast Stone Sample Preparation

### 2.1.1 Simulant

The LAW tank waste simulant used in this study was derived from an overall average of the 1046 weeks of modeled LAW feed to a supplemental immobilization facility over a 20 year mission.<sup>8</sup> This simulant is referred to as the HTWOS “Average” 5 M Na simulant and was chosen for this study because it is the baseline feed for the WTP pretreatment process. One product from this process will be a decontaminated 5 M Na aqueous waste stream which will require additional

treatment prior to disposal. Cast Stone is one of the treatment options currently being considered. The composition of the 5 M “Average” simulant is provided in Table 2-1.

**Table 2-1. LAW 5 M Simulant Based on Hanford Tank Waste Operations Simulator (HTWOS) Modeling<sup>4</sup>.**

Waste Constituent	HTWOS Overall Average Concentration		
	(moles / L)	(g / L)	(mg / g)*
Na	5.000	115.00	93.50
K	0.03	1.28	1.04
Al	0.31	8.28	6.73
Cl	0.04	1.5	1.22
F	0.030	0.6	0.49
S	0.090	2.74	2.23
P	0.050	1.52	1.24
NO <sub>2</sub>	0.57	26	21.14
NO <sub>3</sub>	1.63	101	82.11
CO <sub>3</sub>	0.27	16.5	13.41
OH	1.56	26.5	21.54

\* 1 ml 5 M Na simulant = 1.230 g 5 M Na simulant. 1 gram of Cast Stone made with 5 M Na simulate and a water to cementitious solids ratio = 0.60 contains 0.451g 5 M Na simulant.

The ingredients and proportions in the 5M Na simulated salt solution used to prepare the waste form are provided in Table 2-2. The compounds were added to the liquid in the order in which they are listed in Table 2-2.

**Table 2-2. Average 5M Simulant Composition.**

Compound	Amount (g / L)
Water	819.50
Al(NO <sub>3</sub> ) <sub>3</sub> ·9 H <sub>2</sub> O	115.165
50% by Weight NaOH	223.04
Na <sub>2</sub> SO <sub>4</sub>	12.215
Na <sub>3</sub> PO <sub>4</sub> ·12 H <sub>2</sub> O	18.5
NaCH <sub>3</sub> COO·3 H <sub>2</sub> O	5.25
Na <sub>2</sub> CO <sub>3</sub>	29.05
NaNO <sub>3</sub>	56.79
NaNO <sub>2</sub>	38.975
NaCl	2.48
NaF	1.35
KNO <sub>3</sub>	3.33
Density	1.230
Wt.% Solids	27.06

### 2.1.2 Cast Stone Premix Reagents

The cement, blast furnace slag, and fly ash used in this study were obtained from a supplier in the Hanford area via PNNL and were shipped to SRNL. The oxide compositions of the cement, fly ash,

and slag were characterized by heating at 800 °C for one hour to determine the weight percent of the refractory oxides (minus evaporable and bound water and CO<sub>2</sub>). This information and other cementitious reagent characterization data are reported elsewhere.<sup>5</sup> The three cementitious materials were pre mixed, i.e., batched, at the appropriate cement:slag:fly ash ratios of 8:47:45 by mass and were stored in sealed plastic bags. The premix was blended by manually shaking the bags.

### 2.1.3 Cast Stone Spiked with Chromium

Cast Stone was prepared in 1 kg batches using a water to cement + slag + fly ash ratio (w/cm) of 0.60. The process was as follows:

1. The amount of average 5 M Na simulant was calculated for a 1 kg batch of Cast Stone based on the water to cementitious solids ratio and the weight percent water in the simulant.
2. Cr(VI) was added to the 5 M Na simulant as Na<sub>2</sub>CrO<sub>4</sub>. The amount was based on the targeted concentration in the Cast Stone. For Cast Stone with a targeted concentration of 500 mg/kg Cr, 1.5575 g of Na<sub>2</sub>CrO<sub>4</sub> was added to 451.33 g of 5 M Na simulant. The sodium chromate was completely dissolved prior to mixing the solution with the premix reagents.
3. 548.67 g of premix was added to the spiked salt solution and mixed with an overhead stirrer for three minutes after all of the ingredients were incorporated into the slurry.

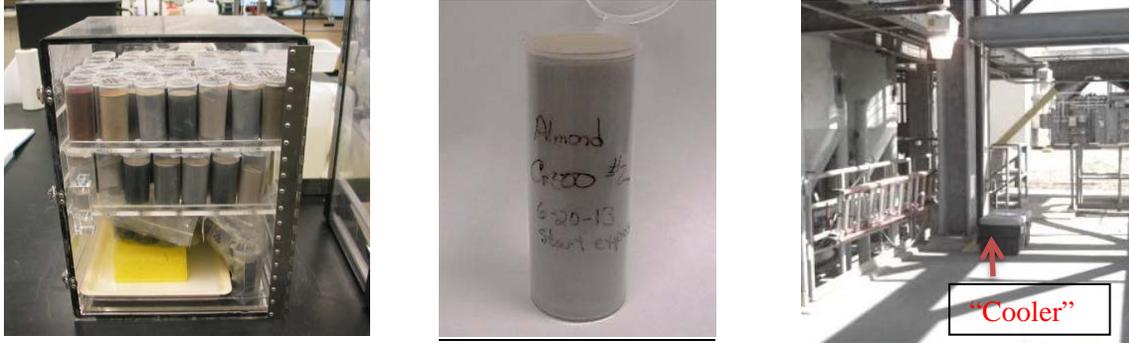
Ingredients and proportions of the Cast Stone mixes prepared for the oxidation front study are listed in Table 2-3.

**Table 2-3. Ingredients in Chromium Spiked Cast Stone, w/cm = 0.60.**

Cr Concentration in Cast stone (mg / kg)	Cast Stone Batch Size (g)	5M Na Average Simulant (g)	Na <sub>2</sub> CrO <sub>4</sub> (g)	8:47:45 Blend (g)	Blend Proportions (g)
75	1000	451.33	0.2336	548.67	Cement 43.89 Slag 257.87 Fly Ash 246.90
125			0.3894		
250			0.7788		
500			1.5575		
1000			3.1150		

After mixing, the grout slurry was poured into cylindrical molds approximately 1 inch in diameter by 4 inches high and capped.<sup>b</sup> At this time, the first exposure time for a 500 mg/kg mix has been characterized. The capped cylinders were cured at ambient temperature (about 22 °C) and 65% to 70% relative humidity for 63 days before being relocated to another container. The samples were uncapped and relocated to another container (storage cooler), which was located outdoors in Z-Area at the SRS. See Figure 2-1. The samples were subjected to ambient SRS outdoor temperature fluctuations and the surfaces of the samples were exposed to moist air. Temperature and humidity in the outdoor cooler were monitored. The cooler was opened on an irregular basis (about 2 times per month) to inspect and extract samples of other projects. The “field” location provided exposure to outdoor ambient temperature fluctuations while providing some shelter from rain and direct sunlight, in an attempt to simulate the environment in a partially or completely filled disposal unit. To date one exposure time has been analyzed for a sample containing 500 mg/kg Cr. This sample was exposed for 68 days after being cured for 63 days at ambient laboratory conditions. The total age of the sample analyzed was 131 days.

<sup>b</sup> The molds were LAVIAL hinged cap laboratory grade clarified natural polypropylene thin walled containers.



**Figure 2-1. Laboratory Curing Container (left); Cast Stone Sample After Curing for 30 Days Prior to Exposure Testing (center), Outdoor Exposure Container (right).**

#### 2.1.4 Cast Stone Spiked with Technetium

The general method for preparing Cast Stone samples spiked with Tc-99 was the same as that described for preparing samples spiked with Cr. Rather than adding the spike to achieve a pre-determined concentration in the Cast Stone, Tc-99 was added to the 5 M Na simulant to achieve a targeted concentration of about 207  $\mu\text{Ci} / \text{L}$  in the simulant, which is the current flow sheet design case for the simulant.<sup>5</sup> The Tc-99 was added to the 5 M Na simulant as Tc(VII).

The spike stock solution was an aqueous solution containing  $\text{NH}_4\text{TcO}_4$  and was purchased from Eckert and Ziegler Isotope Products, Valencia, CA. This stock solution had a specific activity of 0.629 MBq / g (0.017 Ci / g) and a concentration of 18.5 MBq / ml (0.5 mCi / ml).

The batch size for the Tc spiked samples was 1800 g. One batch was prepared with a water to cementitious materials ratio (w/cm) of 0.60 and another with a w/cm of 0.50. (At this time, only one sample with a w/cm ratio of 0.60 has been analyzed.) Ingredients and Tc concentrations are listed in Table 2-4.

**Table 2-4. Ingredients in Cast Stone Containing HTWOS Average 5M Na Simulant Spiked with Tc-99.**

Cast Stone w/cm	Average 5 M Na Simulant Tc-99 Concentration* ( $\mu\text{Ci} / \text{L}$ )	Cast Stone Batch Size (g)	5M Na Average Simulant (g)	5M Na Average Simulant (ml)	$\text{NH}_4\text{TcO}_4$ 0.5 mCi/ml Stock Solution (ml)	8:47:45 Cement: slag: Fly Ash Blend (g)	5M Cast Stone Tc-99 Concentration $\mu\text{Ci} / \text{g}$
0.50	243	1559	634	515.45	0.25 (250 $\mu\text{L}$ )	925	0.0802
0.60	189	1800	812.5	660.57	0.25 (250 $\mu\text{L}$ )	987.5	0.0694

\* The HTWOS estimated maximum concentration for Tc-99 is 4.13 E-05 Ci / moles of Na. For 5 M Na simulant, the maximum Tc-99 value is 20.58 E-05 Ci per liter of simulant (205.8  $\mu\text{Ci} / \text{L}$ ).

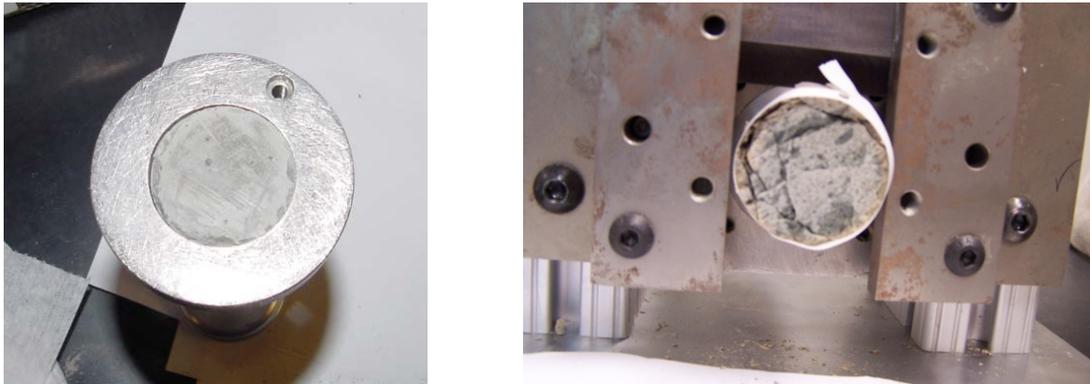
After mixing, the grout slurry was poured into cylindrical molds approximately 1 inch in diameter by 4 inches high and capped. The capped cylinders were placed in 2-liter plastic containers which contained wet towels to provide a moist environment around the sealed samples. The containers were stored in the rad laboratory. (Longer age samples are still being cured.) The sample analyzed in this study, Tc2, had a w/cm of 0.60 and was cured for 48 days at ambient temperature ( $\sim 22^\circ \text{C}$ )

after which the cap was removed and the sample was relocated to another container in the rad lab where it was exposed to moist ambient air (~ 21 wt. % oxygen). The top surface of this sample was exposed to moist air at ambient laboratory temperature for 50 days. At the time the depth discrete subsamples were collected, the total age of the sample was 98 days.<sup>c</sup>

## 2.2 Cast Stone Depth Discrete Sub-Sampling

A simple holder and blade fabricated for non rad saltstone subsampling<sup>8,9</sup> was used for removing layers from the Cast Stone spiked with 500 mg/kg Cr. This holder is shown in Figure 2-2 (left) and has an indexed screw to advance the sample. Initially, this tool could not be used in the rad hood because the blade was determined to be unacceptable. Several iterations of the holder and blade were designed and tested. A guillotine type holder and knife shown in Figure 2-2 (right) produced a cracked sample monolith with many shards and was too slow to operate. The original screw type holder and blade arrangement was eventually modified by substituting a rasp and modified blade for work in the rad hood. None of the Cast Stone monoliths had visible cracking when they were de-molded and sectioned using the holder shown in Figure 2-2 (left).

Dark spots were observed in the Cast Stone monoliths during the sectioning process. These dark spots are illustrated in the Cr spiked sample shown in the apparatus in Figure 2-2 (left). These spots were typical of both the Cr and Tc spiked samples prepared in this study.<sup>d</sup> The cylindrical monoliths were measured relative to the bottom of the cylinder after each section was removed. The thickness of the subsamples ranged from less than 1 mm to several mm depending on the location along the cylinder and the desired resolution of the sample location versus the fraction leached.



**Figure 2-2. Sample Holders Utilized to Cut Depth Discrete Samples.**

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<sup>c</sup> Although only the top of the cylindrical sample was directly exposed to air, the sides of the sample may also have been exposed to air if a gap formed between the container and sample and / or if oxygen diffused through the thin walled polypropylene vial. No gap was observed when the Cast Stone samples were de-molded and only the surfaces of the cylindrical samples were slightly lighter in color than the bulk material immediately after de-molding. Consequently, the extent of oxidation on the unexposed surfaces and through the container side walls was assumed to be minimal for the time periods evaluated.

The discoloration observed on the sides of the sample in Figure 2-2 (right) was the result of leaving a de-molded cylindrical sample on the bench top for several weeks during which time the knife and holder apparatus shown in the figure were being designed and fabricated.

<sup>d</sup> Samples were prepared according to SRNL R&D Directions – Cast Stone (CS) Grout Mixing prepared by E.K. Hansen, 2012. The light and dark regions are not noticeable on monolithic samples and are only visible when samples are cut.

### 2.3 Cast Stone Depth Discrete Sub-Sample Leaching

Each layer removed from the Cr and Tc spiked monoliths was size reduced in air (crushed to a powder) with an agate mortar and pestle to produce a sample for a short term leach test (Modified EPA 1311)<sup>10</sup>. Particle size of the damp powder was not measured. The size reduced sample was divided: one portion was leached; the other portion was used for the reduction capacity measurements. The time required for cutting, crushing, weighing, and covering the sample with leachate was typically about 5 minutes.

Deionized, de-aerated ASTM Type I/II water was used as the leachate. Zero head space leaching containers were used to leach the samples in an oxygen free or low oxygen environment, thereby preventing / minimizing additional oxidation caused by the leaching method. Filled leach vessels were loaded into a large mouth plastic bottle which was tumbled end-over-end at 30 rpm for  $48 \pm 2$  hours.

After tumbling, leachates were filtered using a 0.45 micrometer membrane attached to 20 mL syringes. Ca, K, Al, Cr, and Na concentrations were measured using an Inductively Coupled Plasma- Optical Emission Spectrometer (ICP-OES), Varian 730-ES.  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ , and  $\text{SO}_4^{2-}$  were analyzed using Ion Chromatography (IC), Dionex ICS-5000 EG (See Attachments 1 and 2). The pH of the leachates was also determined using an IQ Scientific pH meter. Leachate analyses were run in duplicate. Values were averaged and used to calculate the fraction leached (percent leached). Tc concentrations were analyzed by liquid scintillation.

The mass fraction of selected anions and cations leached or percent leached (mass fraction leached X 100) from each crushed subsample was selected as the parameter for indicating: 1) the effect of exposure to air, i.e., oxidation, on the redox sensitive contaminants and 2) the depth of penetration of oxygen into the waste form, i.e., location of the oxidation front. The percent leached was calculated using Equation 1.

$$\text{Equation 1. } \% \text{ Leached} = 100 * \left( \frac{m_{i\text{-leachate}}}{m_{i\text{-total in solid}}} \right)$$

Where:

$m_{i\text{-leached}}$  = mass of species i leached (mg). The leachate was filtered prior to analysis using a 0.45  $\mu\text{m}$  filter.

$M_{i\text{-total}}$  = mass of species i in the subsample leached (mg). The total mass in the waste form used in this calculation reflects the contribution to the waste form from the waste simulant, i.e. aqueous component. This method of calculating mass fraction or weight percent leached can result in greater than 100 wt. % leached for those ions in the premix which are also soluble in the waste form.

### 2.4 Reduction Capacity Measurements

Reduction capacity measurements were carried out following a redox titration method based on the Ce(IV)-Fe(II) chemical system.<sup>6, 10</sup> The crushed cementitious material was dissolved in an acidic 0.0608 M Ce(IV) solution for one hour after which the resulting solution was titrated with ferrous ammonium sulfate (FAS) in the presence of the indicator 1-10 phenanthroline (ferroin). The reduction capacity of the cementitious material was calculated based on the amount of FAS needed to reach the endpoint.

The Hanford fly ash and consequently Cast Stone made with the fly ash contained magnetic particles, which did not dissolve in one hour the Ce(IV) sulfuric acid-based solution used in the reduction capacity measurements. The particles were assumed to be magnetite,  $\text{Fe}_3\text{O}_4$ . If an attempt had been

made to completely dissolve these the magnetite particles, the measured reduction capacities for fly ash and Cast Stone were expected to be higher than the reported values because magnetite contains ferrous iron, (Fe(II)Fe(III)<sub>2</sub>O<sub>4</sub>).

### 2.5 Evaluation of Tc Reduction Kinetics in Cast Stone

Quantifying the kinetics of Tc(VII) reduction in Cast Stone as a function of setting and curing time was identified as a need to interpret data collected from samples cured in excess of 28 days which were then exposed to oxygen. Consequently a set of samples (Tc 4-1 to 4-10) was prepared to provide a base line for Tc reduction kinetics. Subsamples were prepared for depth discrete subsampling and leaching at several early ages without exposing the top surface to moist air.

#### 2.5.1 Cast Stone Sample Preparation (10 Day Old Sample)

Sample preparation consisted of preparing 5 M Na HTWOS Average Simulant and spiking 1495.8 g of this simulant with 500  $\mu$ L of Tc-99 stock solution with a Tc-99 concentration of 0.5 mCi / mL (18.5 MBq / mL). The Tc-99 concentration in the spiked 5 M Na HTWOS Average Simulant was determined by liquid scintillation. (The clear spiked simulant was filtered prior to submitting it for analysis.) This value was used to calculate the Tc-99 concentration in the resulting Cast Stone samples. See Table 3-6.

**Table 2-5. Tc-99 Concentrations in Spiked 5 M Na HTWOS Average Simulant and Cast Stone Prepared with a w /cm of 0.60.**

Tc-99 Concentration in Stock Solution (mCi/mL)	5 M Na HTWOS Average Simulant (g)	Volume Tc-99 Stock Solution ( $\mu$ L)	Measured Tc-99 concentration for filtered 5 M Na HTWOS Average Simulant			Mass Fraction 5 M Na HTWOS Average Simulant in Cast Stone w / pm = 0.60	Tc-99 Concentration in 1 g of Cast Stone w / pm = 0.60 (uCi Tc-99 / g)
			Sample No. 300307438 (dpm / ml)	(dpm Tc-99 / g simulant)	(uCi Tc-99 / g simulant)		
0.5	1495.8	500	4.71E+05	3.83E+05	1.72E-01	0.4514	7.79E-02

The 90 x 35 mm containers were filled to the top to the extent possible. Usually a small air space was formed as the lid was closed. The samples were stored at ambient laboratory conditions in the closed containers, which were placed in an over pack container with moist towel.

#### 2.5.2 10 Day Old Depth Discrete Subsample Preparation and Leaching

Depth discrete subsamples were collected from the first early age sample, Tc 4-1, 10 days after preparation. These subsamples were ground and leached in de-aerated deionized water for 56 hours (over the week end). The 10 day old sample dark green and damp, which is was consistent with that of GGBFS-based cementitious materials, including concrete, cured in sealed containers and / or moist conditions, See Figure 2-3 (a) and (b). (Small dark specks can be seen in the fresh surface exposed in Figure 2-3 (a). Similar dark specks were observed in all GGBFS-based samples sectioned for this study.)

Samples prepared for leaching but not yet tumbled are shown in Figure 2-3 (c) which illustrates the yellow color that is evident within 30 minutes of GGBFS waste forms in contact with de-aerated, DI water. The color has been correlated with available or “active” chemical reduction capacity with respect to reducing Tc and Cr. (No chromate was added to the sample.) Figure 2-3 (d) illustrates the yellow color of these leachates after leaching for 56 hours and filtering prior to submitting samples for analysis.



**Figure 2-3. Depth discrete Cast Stone Sample Preparation. (a) Monolithic Cylinder in Sample Holder Prior to Sectioning, (b) Ground Depth Discrete Sample Prior to Leaching, (c) Leaching Samples Prior to Tumbling, (d) Leached Samples After Tumbling.**

### 3.0 Results and Discussion

#### 3.1 Leaching Result for Cast Stone Spiked with Chromium

Cast Stone sample Cr500-1 was prepared with HTWOS average 5 M Na simulant spiked with 500 mg/kg Cr(VI) and a  $w/cm = 0.60$ . This sample was cured in the laboratory in a sealed container at ambient conditions for 63 days before being exposed to moist air for 68 days in a cooler located outdoors at the SRS. The lid on the sample container was opened during the exposure in order to expose the top surface of the Cast Stone cylinder to moist air. The exposure time under the conditions in the cooler was intended to approximate conditions in a closed Cast Stone disposal container. The age of the sample was 131 days.

After the sample was retrieved, thin layers were removed starting at the top surface. Each layer was ground and divided into two parts, one for leaching and another, about 0.5 g, for the reduction capacity measurement. Leachate results for Cr, Na, and  $\text{NO}_3^-$  are provided in Tables 3-1 to 3-3, respectively. Results are also plotted in Figures 3-1 as a function of mean sample location from the top exposed surface. The complete data set for the leachate analyses is provided in Appendix 1.

Leaching results for Na,  $\text{NO}_3^-$ , and Cr, a redox sensitive species, indicate that the top 10 to 12 mm of the 90 mm cylindrical sample are not representative of the bulk material. See Figure 3-1. This feature may be the result of inhomogeneity in the original sample due to settling or stratification in the slurry. Settling results in water accumulation on the top surface and near-surface region which ultimately results in higher porosity relative to the bulk material. Another explanation is that soluble ions were mobilized and redistributing as the result of the near-surface region being subjected to wet-dry cycling. A combination of these phenomena is also possible. Additional testing is required to characterize the top 10 to 12 mm of the sample and determine the significance of this region to the overall performance of the sample.

The mass fraction of Cr leached from the CR500-1 subsamples is much lower than those for Na,  $\text{NO}_3^-$ . The highest percent Cr leached from this sample (0.70 to 0.73 wt. %) occurred in subsamples collected from the top 1.5 mm. Between 1.5 and about 12 mm, the percent of Cr leached from subsamples ranged from about 0.19 to 0.30 wt. %. Depth discrete samples between about 12 and 35 mm below the exposed surface leached between 0.50 and 0.71 wt. % of the Cr added to the 5 M Na simulant. The amount of Cr leached below about 35 mm was less than 0.01 wt % of the concentration in the Cast Stone. The concentrations of Cr in the leachate were below the Cr detection limit of 0.01 mg / L.

Based on these results, the chromium-specific oxidation front in Cast Stone sample Cr500-1 was estimated to be about  $35 \pm 5$  mm below the exposed surface for the 131 day old sample exposed to moist air and variable outdoor temperature in a closed cooler for 68 days.

Most of the  $\text{NO}_3^-$  (75 to 97 % of the amount supplied by the 5 M Na simulant) and essentially all of the Na (90 to 112 weight % of the amount supplied by the 5 M Na simulant) leached out of the top 38 mm of Cast Stone sample Cr500-1. Explanations for many of the subsamples leaching more than 100 wt. % of the Na originating from the 5 M Na HTWOS simulant include: 1) dissolution of Na salts in the solid cementitious reagents or hydration products, which were not included in the initial Na concentration calculation 2) redistribution of the Na while the surface was exposed to moist air, and / or 3) combination of both explanations.

**Table 3-1. Cr Leaching Results for Cast Stone Mix Cr 500-1 Depth Discrete Subsamples.**

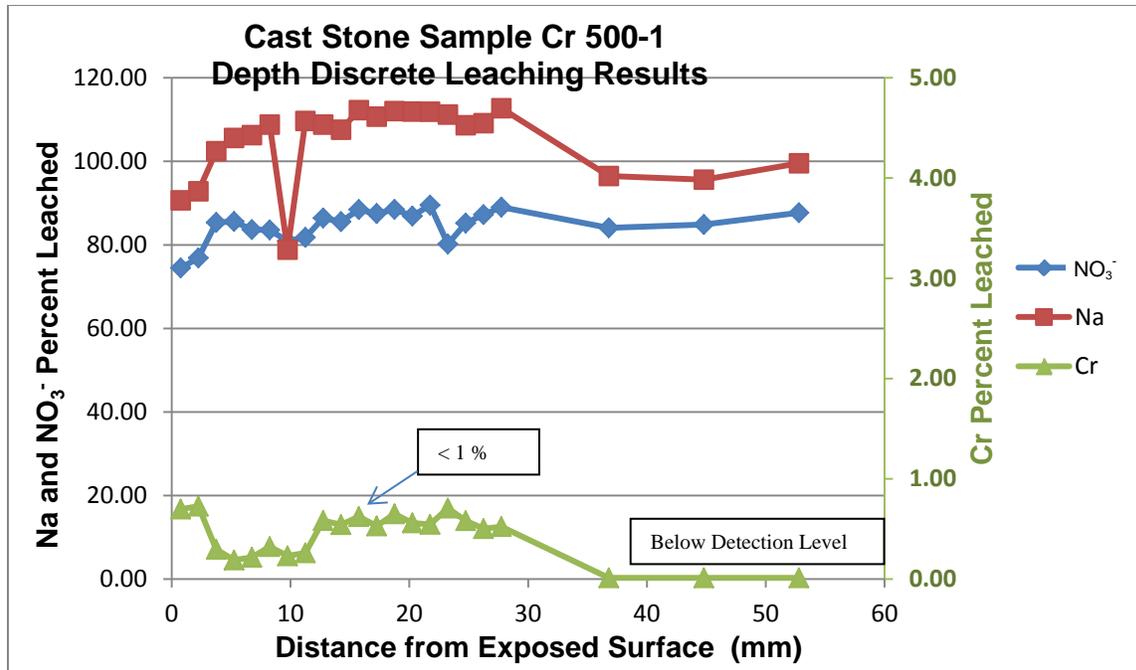
Sub-sample No.	Cylinder Length (mm)	Mean Location		Sub-sample Mass (g)	Leachate Volume (g or ml)	Cr in Leachate duplicate analyses			Cr in 1 g of Cast Stone (mg / g)	Total Cr leached (mg)	Cr in Sub-sample before leach (mg)	% Cr leached
		Bottom (mm)	From Exposed Surface (Top) (mm)			A (mg / L)	B (mg / L)	Ave. (mg / L)				
0	88.3							-				
1	86.8	87.55	0.75	0.731	42.8531	0.119	0.119	0.12	1.00	0.0051	0.73	0.70
2	85.3	86.05	2.25	1.413	42.419	0.242	0.242	0.24	1.00	0.0103	1.41	0.73
3	83.8	84.55	3.75	1.420	42.236	0.100	0.099	0.10	1.00	0.0042	1.42	0.30
4	82.3	83.05	5.25	1.213	42.907	0.052	0.055	0.05	1.00	0.0023	1.21	0.19
5	80.8	81.55	6.75	1.313	42.874	0.066	0.067	0.07	1.00	0.0029	1.31	0.22
6	79.3	80.05	8.25	1.276	42.850	0.096	0.096	0.10	1.00	0.0041	1.28	0.32
7	77.8	78.55	9.75	1.559	42.540	0.085	0.084	0.08	1.00	0.0036	1.56	0.23
8	76.3	77.05	11.25	1.362	42.714	0.080	0.086	0.08	1.00	0.0035	1.36	0.26
9	74.8	75.55	12.75	1.550	42.476	0.211	0.215	0.21	1.00	0.0090	1.55	0.58
10	73.3	74.05	14.25	1.482	42.780	0.189	0.186	0.19	1.00	0.0080	1.48	0.54
11	71.8	72.55	15.75	1.357	42.899	0.195	0.199	0.20	1.00	0.0085	1.36	0.62
12	70.3	71.05	17.25	1.607	42.364	0.198	0.203	0.20	1.00	0.0085	1.61	0.53
13	68.8	69.55	18.75	1.431	42.723	0.218	0.217	0.22	1.00	0.0093	1.43	0.65
14	67.3	68.05	20.25	1.561	42.691	0.204	0.205	0.20	1.00	0.0087	1.56	0.56
15	65.8	66.55	21.75	1.311	43.800	0.164	0.162	0.16	1.00	0.0071	1.31	0.54
16	64.3	65.05	23.25	1.529	42.392	0.258	0.252	0.26	1.00	0.0108	1.53	0.71
17	62.8	63.55	24.75	1.376	42.250	0.188	0.191	0.19	1.00	0.0080	1.38	0.58
18	61.3	62.05	26.25	1.465	42.441	0.174	0.173	0.17	1.00	0.0074	1.47	0.50
19	59.8	60.55	27.75	1.480	42.744	0.184	0.178	0.18	1.00	0.0077	1.48	0.52
20/21	53 to 50	51.5	36.8	3.718	41.377	<0.010	<0.010	<0.01	1.00	0.0004	3.72	<0.01
22/23	45 to 42	43.5	44.8	3.536	41.655	<0.010	<0.010	<0.01	1.00	0.0004	3.54	<0.01
24/25	37 to 34	35.5	52.8	3.622	41.345	<0.010	<0.010	<0.01	1.00	0.0004	3.62	<0.01

**Table 3-2. Na Leaching Results for Cast Stone Mix Cr 500-1 Depth Discrete Subsamples.**

Sub-sample No.	Cylinder Length (mm)	Mean Location		Sub-sample Mass (g)	Leachate Volume (g or ml)	Na in Leachate duplicate analyses			Na in 1 g of Cast Stone (mg / g)	Total Na leached (mg)	Na in Sub-sample before leach (mg)	% Na leached
		From Bottom (mm)	From Exposed Surface (Top) (mm)			A (mg / L)	B (mg / L)	Ave. (mg / L)				
0	88.3	--	--	--	--	--	--	--	--	--	--	--
1	86.8	87.55	0.75	0.731	42.8531	569	576	573	37.03	24.53	27.06	90.66
2	85.3	86.05	2.25	1.413	42.419	1140	1150	1145	37.03	48.57	52.32	92.83
3	83.8	84.55	3.75	1.420	42.236	1270	1280	1275	37.03	53.85	52.57	102.44
4	82.3	83.05	5.25	1.213	42.907	1110	1100	1105	37.03	47.41	44.90	105.60
5	80.8	81.55	6.75	1.313	42.874	1200	1210	1205	37.03	51.66	48.60	106.30
6	79.3	80.05	8.25	1.276	42.850	1200	1200	1200	37.03	51.42	47.27	108.79
7	77.8	78.55	9.75	1.559	42.540	1070	1070	1070	37.03	45.52	57.73	78.85
8	76.3	77.05	11.25	1.362	42.714	1300	1290	1295	37.03	55.31	50.44	109.67
9	74.8	75.55	12.75	1.550	42.476	1470	1470	1470	37.03	62.44	57.40	108.78
10	73.3	74.05	14.25	1.482	42.780	1370	1390	1380	37.03	59.04	54.89	107.56
11	71.8	72.55	15.75	1.357	42.899	1320	1310	1315	37.03	56.41	50.25	112.25
12	70.3	71.05	17.25	1.607	42.364	1550	1560	1555	37.03	65.88	59.52	110.68
13	68.8	69.55	18.75	1.431	42.723	1390	1390	1390	37.03	59.38	53.00	112.05
14	67.3	68.05	20.25	1.561	42.691	1510	1520	1515	37.03	64.68	57.80	111.90
15	65.8	66.55	21.75	1.311	43.800	1240	1240	1240	37.03	54.31	48.54	111.89
16	64.3	65.05	23.25	1.529	42.392	1470	1500	1485	37.03	62.95	56.62	111.18
17	62.8	63.55	24.75	1.376	42.250	1320	1300	1310	37.03	55.35	50.95	108.64
18	61.3	62.05	26.25	1.465	42.441	1380	1410	1395	37.03	59.20	54.25	109.14
19	59.8	60.55	27.75	1.480	42.744	1440	1450	1445	37.03	61.77	54.80	112.72
20/21	53 to 50	51.5	36.8	3.718	41.377	3220	3200	3210	37.03	132.82	137.66	96.48
22/23	45 to 42	43.5	44.8	3.536	41.655	3030	2980	3005	37.03	125.17	130.93	95.60
24/25	37 to 34	35.5	52.8	3.622	41.345	3240	3220	3230	37.03	133.54	134.14	99.56

**Table 3-3. NO<sub>3</sub><sup>-</sup> Leaching Results for Cast Stone Mix Cr 500-1 Depth Discrete Subsamples.**

Sub-sample No.	Cylinder Length (mm)	Mean Location		Sub-sample Mass (g)	Leachate Volume (g or ml)	NO <sub>3</sub> <sup>-</sup> in Leachate duplicate analyses			NO <sub>3</sub> <sup>-</sup> in 1 g of Cast Stone (mg / g)	NO <sub>3</sub> <sup>-</sup> Total Na leached (mg)	NO <sub>3</sub> <sup>-</sup> in Sub-sample before leach (mg)	% NO <sub>3</sub> <sup>-</sup> Leached
		Bottom (mm)	From Exposed Surface (Top) (mm)			A (mg / L)	B (mg / L)	Ave. (mg / L)				
0	88.3	--	--	--	--	--	--	--	--	--	--	--
1	86.8	87.55	0.75	0.731	42.8531	536	535	536	42.17	22.95	30.82	74.46
2	85.3	86.05	2.25	1.413	42.419	1080	1080	808	42.17	34.25	59.58	76.89
3	83.8	84.55	3.75	1.420	42.236	1210	1210	1145	42.17	48.36	59.86	85.37
4	82.3	83.05	5.25	1.213	42.907	1020	1020	1115	42.17	47.84	51.13	85.59
5	80.8	81.55	6.75	1.313	42.874	1080	1080	1050	42.17	45.02	55.35	83.66
6	79.3	80.05	8.25	1.276	42.850	1050	1050	1065	42.17	45.63	53.83	83.58
7	77.8	78.55	9.75	1.200	42.540	1250	1250	1150	42.17	48.92	50.60	80.89
8	76.3	77.05	11.25	1.362	42.714	1100	1100	1175	42.17	50.19	57.44	81.80
9	74.8	75.55	12.75	1.550	42.476	1330	1330	1215	42.17	51.61	65.37	86.42
10	73.3	74.05	14.25	1.482	42.780	1250	1250	1290	42.17	55.19	62.50	85.55
11	71.8	72.55	15.75	1.357	42.899	1180	1180	1215	42.17	52.12	57.23	88.45
12	70.3	71.05	17.25	1.607	42.364	1400	1400	1290	42.17	54.65	67.78	87.51
13	68.8	69.55	18.75	1.431	42.723	1250	1250	1325	42.17	56.61	60.35	88.48
14	67.3	68.05	20.25	1.561	42.691	1340	1340	1295	42.17	55.28	65.82	86.91
15	65.8	66.55	21.75	1.311	43.800	1130	1130	1235	42.17	54.09	55.28	89.54
16	64.3	65.05	23.25	1.529	42.392	1310	1130	1220	42.17	51.72	64.48	80.21
17	62.8	63.55	24.75	1.376	42.250	1170	1170	1150	42.17	48.59	58.02	85.20
18	61.3	62.05	26.25	1.465	42.441	1270	1270	1220	42.17	51.78	61.78	87.25
19	59.8	60.55	27.75	1.480	42.744	1300	1300	1285	42.17	54.93	62.40	89.05
20/21	53 to 50	51.5	36.8	3.718	41.377	3180	3190	2240	42.17	92.68	114.60	84.06
22/23	45 to 42	43.5	44.8	3.536	41.655	3040	3040	3115	42.17	129.65	149.11	84.86
24/25	37 to 34	35.5	52.8	3.622	41.345	3240	3240	3140	42.17	129.82	152.76	87.69



**Figure 3-1. Cast Stone Cr 500-1 Leachate Results for NO<sub>3</sub><sup>-</sup>, Na, and Cr as a Function of Distance from the Top Exposed Surface.**

### 3.2 Leaching Result for Cast Stone Spiked with Technetium-99.

Cast Stone sample Tc2 was prepared with the HTWOS average 5 M Na simulant and a  $w/cm = 0.60$ . The simulant was spiked with NH<sub>4</sub>TcO<sub>4</sub> aqueous solution and mixed with cementitious reagents to produce Cast Stone with a Tc-99 concentration of 0.0694  $\mu\text{Ci Tc-99} / \text{g}$  of Cast Stone. This sample was cured in a sealed container at ambient laboratory conditions for 48 days before being exposed to moist air for 50 days. The sample was left in the container in which it was cast during curing. Before the sample was exposed to moist air, the lid was removed from the sample container. A beaker of water was placed in the exposure container to maintain a moist air environment, and the exposure container was closed and stored in a rad hood with an estimated temperature of  $23 \pm 3$  °C. The age of the sample characterized in this study was 98 days.

Immediately after the sample was retrieved from the exposure container and de-molded, depth discrete samples were collected starting at the top surface and advancing into the cylindrical monolith. Each sub-sample was ground in air and divided into two parts, one for leaching and another, about 0.5 g, for the reduction capacity measurement. The time required for cutting, grinding, weighing, and covering the samples with leachate was about 5 minutes. Leaching was performed in 40 ml glass vials that were completely filled with deionized, de-aerated water to maintain a zero head space. The leach tests were performed per a modified EPA Method 1311 for 48 hours.

Leachate results for Tc-99, Na, and NO<sub>3</sub><sup>-</sup> are provided in Tables 3-4 to 3-6, respectively. Leachate concentrations are also plotted in Figure 3-2 as a function of mean sample location from the top exposed surface. The complete data set for the leachate analyses is provided in Appendix 2. The differences between the Na and NO<sub>3</sub><sup>-</sup> fractions leached and the Tc-99 fractions leached for the depth discrete subsamples indicates that Tc is chemically stabilized in the waste form relative to Na and NO<sub>3</sub><sup>-</sup>, which are mostly or entirely soluble and leachable in Cast Stone sample Tc2.

The wt. % Tc-99 leached from the depth discrete subsample between 0 and 10 mm below the exposed surface ranged from 25.8 to 56.3 wt. % of the concentration in the as-prepared Cast Stone sample. Between 10 and 16 mm below the surface, the amount leached from the depth discrete samples ranged from 33.4 to 64.1 wt. % of the concentration in the unleached subsamples. Between about 16 and 38 mm below the surface, the wt. % Tc-99 leached fell to 11.1 to 21.3 wt. % of the amount in the unleached sample. These results support the hypothesis that partial oxidation of reduced / stabilized Tc(IV) to soluble Tc(VII) has occurred between 0 and 16 mm from the surface of this sample.

Leachate concentrations for the three subsamples collected from the top 5 mm of sample Tc2 indicated that this surface region of the sample was not representative of the bulk sample. The same patterns in Na and NO<sub>3</sub><sup>-</sup> leached as a function of distance (90 to 100 wt. % of the Na and > 80 wt. % of the NO<sub>3</sub><sup>-</sup>) were observed for samples Tc2 and Cr 500-1 below the region affected by surface phenomena.

**Table 3-4. Tc-99 Leaching Results for Cast Stone Mix Tc-2 Depth Discrete Subsamples.**

Cast Stone Mix Tc-2 Sub sample No.	Length of cylinder (mm)	Distance from Top Surface (mm)	Sub sample mass (g)	Leachate Volume (ml)	Tc-99 Leachate Concentration		Cast Stone Tc-99 Concentration (μCi Tc / g)	Tc-99 Leached (uCi)	Tc-99 in Cast Stone Subsamples Before Leaching (uCi)	% Tc-99 Leached
					(dpm/ml)	(uCi/ml)				
0	90	0	--	--	--	--	--	--	--	--
1	89	0.5	1.557	42.549	2.39E+03	1.08E-03	6.94E-02	4.58E-02	1.08E-01	42.39
2	87	2.5	1.425	42.570	1.72E+03	7.75E-04	6.94E-02	3.30E-02	9.89E-02	33.35
3	85	4.5	1.330	42.585	1.39E+03	6.26E-04	6.94E-02	2.67E-02	9.23E-02	28.89
4	84	5.5	1.068	42.121	2.20E+03	9.91E-04	6.94E-02	4.17E-02	7.41E-02	56.32
5	83	6.5	1.557	41.850	1.48E+03	6.67E-04	6.94E-02	2.79E-02	1.08E-01	25.82
6	81	8.5	1.134	41.736	1.99E+03	8.96E-04	6.94E-02	3.74E-02	7.87E-02	47.54
7	80	9.5	0.807	41.663	1.44E+03	6.49E-04	6.94E-02	2.70E-02	5.60E-02	48.25
8	79	10.5	1.097	43.043	1.33E+03	5.99E-04	6.94E-02	2.58E-02	7.61E-02	33.87
9	78	11.5	1.184	42.884	1.42E+03	6.40E-04	6.94E-02	2.74E-02	8.22E-02	33.38
10	76	13.5	1.196	42.991	1.46E+03	6.58E-04	6.94E-02	2.83E-02	8.30E-02	34.06
11	75	14.5	1.037	42.859	2.39E+03	1.08E-03	6.94E-02	4.61E-02	7.20E-02	64.11
12	74	15.5	0.990	42.766	1.72E+03	7.75E-04	6.94E-02	3.31E-02	6.87E-02	48.23
13	73	16.5	2.730	42.156	1.39E+03	6.26E-04	6.94E-02	2.64E-02	1.89E-01	13.93
14	71	18.5	2.807	41.812	2.20E+03	9.91E-04	6.94E-02	4.14E-02	1.95E-01	21.27
15	68	21.5	2.966	40.602	1.48E+03	6.67E-04	6.94E-02	2.71E-02	2.06E-01	13.15
16	65	24.5	3.018	41.722	1.99E+03	8.96E-04	6.94E-02	3.74E-02	2.09E-01	17.86
17	62	27.5	2.727	42.030	1.44E+03	6.49E-04	6.94E-02	2.73E-02	1.89E-01	14.41
18	60	29.5	3.240	41.701	1.33E+03	5.99E-04	6.94E-02	2.50E-02	2.25E-01	11.11
19	57	32.5	2.674	42.016	1.42E+03	6.40E-04	6.94E-02	2.69E-02	1.86E-01	14.48
20	52	37.5	3.064	41.842	1.46E+03	6.58E-04	6.94E-02	2.75E-02	2.13E-01	12.94

**Table 3-5. Na Leaching Results for Cast Stone Mix Tc-2 Depth Discrete Subsamples.**

Sub sample No.	Cylinder Length (mm)	Mean Distance From Exposed Surface (Top) (mm)	Sub-sample Mass (g)	Leachate Volume (ml)	Na Leachate Concentration (mg/L)	Na in 1 g Cast Stone (mg / g)	Total Na Leached from Sub-sample (mg)	Na in Cast Stone Subsample before Leaching (mg)	% Na Leached
0	90	0	--	--	--	--	--	--	--
1	89	0.5	1.557	42.549	1120	42.17	47.65	65.66	72.58
2	87	2.5	1.425	42.570	1210	42.17	51.51	60.09	85.72
3	85	4.5	1.330	42.585	1160	42.17	49.40	56.08	88.08
4	84	5.5	1.068	42.121	1040	42.17	43.81	45.04	97.27
5	83	6.5	1.557	41.850	1490	42.17	62.36	65.66	94.97
6	81	8.5	1.134	41.736	1160	42.17	48.41	47.82	101.24
7	80	9.5	0.807	41.663	845	42.17	35.21	34.03	103.45
8	79	10.5	1.097	43.043	982	42.17	42.27	46.26	91.37
9	78	11.5	1.184	42.884	1190	42.17	51.03	49.93	102.21
10	76	13.5	1.196	42.991	1210	42.17	52.02	50.43	103.14
11	75	14.5	1.037	42.859	1080	42.17	46.29	43.73	105.85
12	74	15.5	0.990	42.766	1030	42.17	44.05	41.75	105.51
13	73	16.5	2.730	42.156	2600	42.17	109.60	115.12	95.21
14	71	18.5	2.807	41.812	2700	42.17	112.89	118.37	95.37
15	68	21.5	2.966	40.602	2830	42.17	114.90	125.07	91.87
16	65	24.5	3.018	41.722	2890	42.17	120.58	127.26	94.74
17	62	27.5	2.727	42.030	2630	42.17	110.54	114.99	96.13
18	60	29.5	3.240	41.701	3040	42.17	126.77	136.63	92.79
19	57	32.5	2.674	42.016	2590	42.17	108.82	112.76	96.51
20	52	37.5	3.064	41.842	2870	42.17	120.09	129.20	92.94

**Table 3-6. NO<sub>3</sub><sup>-</sup> Leaching Results for Cast Stone Mix Tc-2 Depth Discrete Subsamples.**

Sub sample No.	Cylinder Length (mm)	Mean Distance From Exposed Surface (Top) (mm)	Sub-sample Mass (g)	Leachate Volume (ml)	NO <sub>3</sub> <sup>-</sup> Leachate Concentration (mg/L)	NO <sub>3</sub> <sup>-</sup> in 1 g Cast Stone (mg / g)	Total NO <sub>3</sub> <sup>-</sup> Leached from Sub-sample (mg)	NO <sub>3</sub> <sup>-</sup> in Cast Stone Subsample Before Leaching (mg)	% NO <sub>3</sub> <sup>-</sup> Leached
0	90	0	--	--	--	--	29.02	--	--
1	89	0.5	1.557	42.549	682	42.17	38.65	65.66	44.20
2	87	2.5	1.425	42.570	908	42.17	38.84	60.09	64.33
3	85	4.5	1.330	42.585	912	42.17	36.52	56.08	69.25
4	84	5.5	1.068	42.121	867	42.17	54.82	45.04	81.09
5	83	6.5	1.557	41.850	1310	42.17	42.99	65.66	83.50
6	81	8.5	1.134	41.736	1030	42.17	31.91	47.82	89.90
7	80	9.5	0.807	41.663	766	42.17	34.39	34.03	93.78
8	79	10.5	1.097	43.043	799	42.17	42.46	46.26	74.35
9	78	11.5	1.184	42.884	990	42.17	43.29	49.93	85.03
10	76	13.5	1.196	42.991	1007	42.17	38.10	50.43	85.84
11	75	14.5	1.037	42.859	889	42.17	36.52	43.73	87.13
12	74	15.5	0.990	42.766	854	42.17	101.76	41.75	87.48
13	73	16.5	2.730	42.156	2414	42.17	107.50	115.12	88.40
14	71	18.5	2.807	41.812	2571	42.17	110.23	118.37	90.82
15	68	21.5	2.966	40.602	2715	42.17	115.86	125.07	88.14
16	65	24.5	3.018	41.722	2777	42.17	105.20	127.26	91.04
17	62	27.5	2.727	42.030	2503	42.17	122.02	114.99	91.48
18	60	29.5	3.240	41.701	2926	42.17	101.34	136.63	89.31
19	57	32.5	2.674	42.016	2412	42.17	112.60	112.76	89.88
20	52	37.5	3.064	41.842	2691	42.17	29.02	129.20	87.15

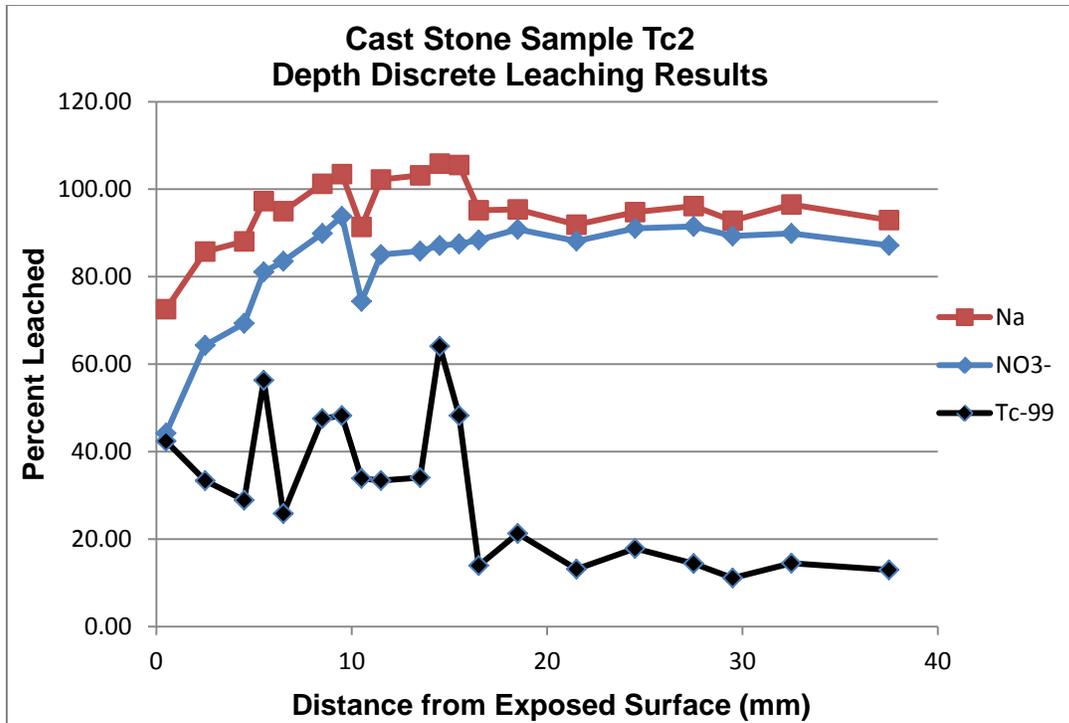


Figure 3-2. Cast Stone Tc2 Leachate Results for Na and Tc-99 as a Function of Distance from the Top Exposed Surface.

### 3.3 Comparison of Cr, Tc, Na, and NO<sub>3</sub><sup>-</sup> Leaching Results

Leaching results for non-redox sensitive species such as Na and NO<sub>3</sub><sup>-</sup> and for Cr, which is redox sensitive, suggest some inhomogeneity in the top 5 to 16 mm of the 90 to mm cylindrical samples. Results also indicate that the distance from the exposed surface impacts leaching / solubility of redox sensitive contaminants such as Cr and Tc. In the Cast Stone samples tested, Tc solubility / leachability was more affected by exposure to moist air (exposure to oxygen from the environment) than Cr solubility / leachability. Additional testing is required to quantify the rate of advancement of the effective Tc oxidation front. Results to date suggest that effective oxidation fronts are contaminant specific. A comparison of averaged leachate results for selected depth intervals is provided in Table 3-7.

Table 3-7. Comparison of Cr and Tc Leaching Results for Samples Exposed to Moist Air.

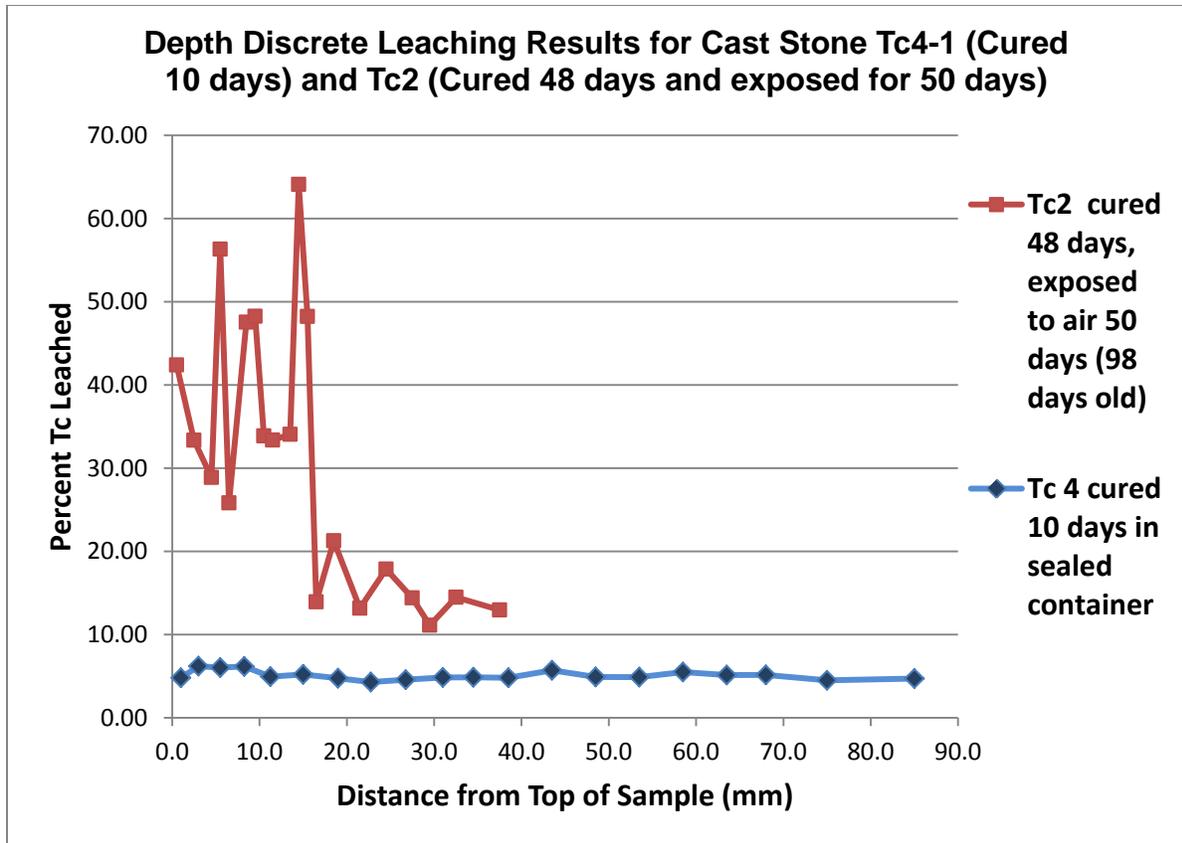
Cast Stone Sample	Distance from Surface (mm)	Wt. % Leached			
		Na	NO <sub>3</sub> <sup>-</sup>	Tc-99	Cr
Cr500-1	0-10	98	52	--	0.5
	10-37	111	79	--	0.6
	37-53	97	84	--	<0.01
Tc2 Cured for 48 days and exposed for 50 days	0-10	90	75	40	--
	10-16	101	84	43	--
	16-38	94	89	15	--
Tc 4-1 Cured 10 days, No intentional exposure to air	1-85	Not measured	Not measured	5	Not measured

### 3.4 Leaching Results for 10 Day Old Cast Stone Spiked with Tc-99

The amount of Tc-99 leached from the 10 day old depth discrete samples containing  $7.79\text{E-}02\mu\text{Ci} / \text{g}$  ( $2.88 \text{ kBq} / \text{g}$ ) Tc-99 ranged from 4.3 to 6.2 percent of the amount in each subsample. The average amount of Tc-99 leached over the entire sample length was 5.0 % of the concentration in the as-cast cylinder. This is about 8X less that the average amount of Tc leached from the top 16 mm of Cast Stone sample Tc2 and about 3X less that the average amount of Tc leached over the depth interval 16 to 38 mm from the top exposed surface. (Tc2 was cured in a sealed container at ambient laboratory conditions for 48 days before being exposed to moist air for 50 days.) Leaching results for the 10 day old Cast Stone sample are tabulated in Table 3-8 and plotted in Figure 3-3.

**Table 3-8. Tc-99 Leaching Results for Depth Discrete Subsamples from Cast Stone Mix Tc 4-1 Cured for 10 days in Closed Container.**

Sub Sample No.	Length of Cylinder (mm)	Mean Distance from Top Surface (mm)	Sub sample mass (g)	DI Water Leachate Volume (ml)	Tc-99 Leachate Concentrations		Cast Stone Tc-99 Concentration ( $\mu\text{Ci Tc} / \text{1g}$ )	Tc-99 Leached ( $\mu\text{Ci}$ )	Tc-99 in Cast Stone Subsample Before leaching ( $\mu\text{Ci}$ )	Percent Tc-99 Leached (%)
					(dpm ml)	( $\mu\text{Ci/ml}$ )				
0	90	0								
1	88	1	2.1609	42.1633	4.25E+02	1.91E-04	7.79E-02	8.07E-03	1.68E-01	4.80
2	86	3	2.9084	41.5735	7.50E+02	3.38E-04	7.79E-02	1.40E-02	2.27E-01	6.20
3	83	5.5	2.909	41.1037	7.37E+02	3.32E-04	7.79E-02	1.36E-02	2.27E-01	6.02
4	80.5	8.25	3.4912	40.2783	9.23E+02	4.16E-04	7.79E-02	1.67E-02	2.72E-01	6.16
5	77	11.25	4.96	39.9538	1.06E+03	4.77E-04	7.79E-02	1.91E-02	3.86E-01	4.94
6	73	15	5.3442	39.6903	1.21E+03	5.45E-04	7.79E-02	2.16E-02	4.16E-01	5.20
7	69	19	5.423	39.726	1.12E+03	5.05E-04	7.79E-02	2.00E-02	4.22E-01	4.74
8	65.5	22.75	5.3715	40.1881	9.91E+02	4.46E-04	7.79E-02	1.79E-02	4.18E-01	4.29
9	61	26.75	4.9629	39.7714	9.85E+02	4.44E-04	7.79E-02	1.76E-02	3.87E-01	4.56
10	57	31	5.1564	40.0101	1.08E+03	4.86E-04	7.79E-02	1.95E-02	4.02E-01	4.85
11	54	34.5	4.872	40.1005	1.02E+03	4.59E-04	7.79E-02	1.84E-02	3.80E-01	4.85
12	49	38.5	6.9912	38.6415	1.50E+03	6.76E-04	7.79E-02	2.61E-02	5.45E-01	4.79
13	44	43.5	6.1877	39.1299	1.56E+03	7.03E-04	7.79E-02	2.75E-02	4.82E-01	5.70
14	39	48.5	6.5364	39.217	1.41E+03	6.35E-04	7.79E-02	2.49E-02	5.09E-01	4.89
15	34	53.5	6.8626	38.8619	1.49E+03	6.71E-04	7.79E-02	2.61E-02	5.35E-01	4.88
16	29	58.5	6.541	38.597	1.61E+03	7.25E-04	7.79E-02	2.80E-02	5.10E-01	5.49
17	24	63.5	6.9296	39.3212	1.56E+03	7.03E-04	7.79E-02	2.76E-02	5.40E-01	5.12
18	20	68	5.7246	39.1323	1.30E+03	5.86E-04	7.79E-02	2.29E-02	4.46E-01	5.14
19	10	75	8.1058	38.2424	1.65E+03	7.43E-04	7.79E-02	2.84E-02	6.31E-01	4.50
20	0	85	6.3431	39.4348	1.31E+03	5.90E-04	7.79E-02	2.33E-02	4.94E-01	4.71



**Figure 3-3. Comparison of Tc-99 Leachability / Solubility as a Function of Distance from the Top Surface for a 10 Day Old and a 98 Day Old (exposed to air) Cast Stone Sample.**

Based on these limited data, the kinetics of pertechnetate ( $TcO_4^{2-}$ ) reduction and re-oxidation in Cast Stone cannot be quantified. However, the majority (about 95%) of the Tc-99 in Cast Stone sample Tc4-1 which contained  $7.8E-02$  uCi Tc / g Cast Stone was reduced within the first 10 days of curing. These results suggest that Tc(VII) reduction to the much less soluble Tc(IV) oxidation state is relatively fast and is associated with setting and very early curing of Cast Stone containing GGBFS.

Re-oxidation of Tc(VII) to soluble Tc(IV) may also be relatively rapid upon exposure to air. Although depth discrete sample preparation in air (shaving, grinding, weighing) took only 5 minutes, these processes could have oxidized 5 % of the Tc in the subsamples. Other possible explanations for the results obtained from the 10 day old sample include: 1) the amount of Tc(VII) added to the Cast Stone exceeded the effective reduction capacity or 2) longer curing times may be necessary for additional or complete reduction. Additional testing is required to address these possibilities and other explanations of the results.

Comparison of the percent Tc-99 leached from a 10 day old sample in a sealed container and the percent Tc leached from a 98 day-old sample (cured for 45 days in a sealed container and then exposed to moist air for 50 days) supports the conclusion that re-oxidation occurred in the 98 day-old sample. Oxygen diffusion through the air and / or fluid in the pores of Tc2 appears to have converted between 59 and 6 percent (64 and 11 percent minus 5 %, i.e., baseline from Tc 4-1) of the Tc(IV) in low solubility solid phase(s) to soluble / leachable Tc(VII).

### 3.5 Reduction Capacity Result

#### 3.5.1 Reduction Capacity of Starting Materials and Fresh Cast Stone

The reduction capacities measured by the Ce(IV)–Fe(II) method for freshly ground (ground immediately before titration) Hanford slag, cement, and fly ash, and the 5 M Na HTWOS average simulant spiked with Tc-99 are provided in Table 3-9. The reduction capacity of freshly ground slag used in the SRS saltstone process is also reported for comparison.

The reduction capacity for Hanford Cast Stone prepared with 5M Na Average HTWOS simulant spiked with 1.72E-01  $\mu\text{Ci}$  Tc-99 / g simulant Tc was also calculated from the sum of fractions of the reduction capacities of the individual components. The calculated reduction capacity for Cast Stone containing the following mass fractions of ingredients: 5M Na simulant = 0.45; cement = 0.04; slag = 0.26, and fly ash = 0.25, is 687  $\mu\text{eq}$  / g which is in relatively good agreement with the measured value of 758  $\mu\text{eq}$  / g.

**Table 3-9. Reduction Capacities of Cement, Slag, and Fly Ash Supplied by PNNL and 5 M Na HTWOS Average Simulant.**

Material	Mass (g)	FAS Volume Titrated (mL)	Fe (mole e-)	(mole e-)	(mole e- / g)	Reduction Capacity (meq / g)	Reduction Measured Capacity ( $\mu\text{eq}$ / g)	Calculated Reduction Capacity ( $\mu\text{eq}$ / g)
no solids calibration	0.0001	31.65	0.001520	0.000000	0.000000	0.000000	0	--
SRS slag (lightly ground)*	0.5000	24.10	0.001157	0.000363	0.000725	0.725182	725	--
5M Na HTWOS Average Simulant Spiked with 1.72E-01 ( $\mu\text{Ci}$ Tc-99 / g simulant)	0.5026	21.66**	0.001070	0.000450	0.000895	0.894697	895	--
Hanford Fly Ash	0.5000	29.79	0.001431	0.000089	0.000179	0.178654	179	--
Hanford Slag (lightly ground)	0.5002	22.44	0.001078	0.000442	0.000884	0.884272	884	--
Hanford Cement	0.4992	28.85	0.001386	0.000134	0.000269	0.269373	269	--
Hanford Cast Stone prepared at SRNL from PNNL supplied material and cured for 35 days in a closed container (ground)	0.4996	23.76	0.001141	0.000379	0.000758	0.758446	758	687 Sum of Fractions of Ingredients

\*The freshly re-ground value for SRS slag measured in this study was less than the value of 820  $\mu\text{eq}$  e<sup>-</sup>/g previously measured by Roberts and Kaplan, 2008.<sup>10</sup> This may be attributed to less grinding or may reflect differences in the slag material tested.

\*\* The no solids calibration of the FAS solution was 30.76 ml of the FAS solution for the 5 M Na HTWOS simulant.

The reduction capacity measured by the Angus-Glasser Ce(IV)-Fe(II) method for the 5 M Na waste simulant is higher than any of the solid ingredients. Nitrite,  $\text{NO}_2^-$ , dissolved in the 5M Na HTWOS Average simulant is the most likely species contributing to the high reduction capacity measured by

this method. Cr and Tc in the 5 M Na simulant are present as the oxidized species, pertechnetate, Tc(VII), and chromate, Cr(VI). Consequently, the reduction capacity measurements made on salt solutions containing nitrite by the Ce(IV)-Fe(II) method do not correlate with and are not an indication of the speciation of the redox sensitive contaminants.

### *3.5.2 Reduction Capacity of Depth Discrete Cast Stone Samples*

Reduction capacity measurements were made on each depth discrete subsample collected from Cast Stone Cr 500-1 and Tc2. Results are tabulated in Tables 3-10 and 3-11 and are plotted as a function of distance from the exposed surface in Figure 3-4. The surface and near surface subsample of both of these Cast Stone specimens had the lowest reduction capacity values for their individual data sets. The averaged values of the Cr 500-1 and Tc2 subsamples were 595  $\mu\text{eq} / \text{g}$  and 567  $\mu\text{eq} / \text{g}$ , respectively, which are lower than 758  $\mu\text{eq} / \text{g}$ , the value measured for Cast Stone cured for 35 days in a closed container. The value for this sample calculated from the sum of fractions of the ingredients was 687  $\mu\text{eq} / \text{g}$ . Reduction capacity was not measured on depth discrete subsamples of Tc 4-1 from which only leaching data were collected.

Bulk reduction capacity measurements performed according to the Angus-Glasser method did not correlate with the fraction of redox sensitive contaminants (Cr and Tc) leached in de-aerated deionized leachate. Based on results obtained in this study and discussions with R. J. Serne, PNNL,<sup>11</sup> reduction capacity measured by the Ce(IV)-Fe(II) method is not an appropriate or meaningful parameter for determining or predicting Tc and Cr oxidation states, speciation, and / or solubilities in cementitious materials containing GGBFS such as Cast Stone. Furthermore, the use of reduction capacity as a general parameter in calculating long-term performance of a porous cementitious solid is not appropriate.

**Table 3-10. Reduction Capacity Results for Cast Stone Cr 500-1 Depth Discrete Subsamples.**

FAS Molarity (calc) 0.0478891 calculated

Ce Molarity 0.0608 theoretical

Ce total (mole e-) 0.001520

Sub Sample No.	Distance From Exposed Surface (Top) (mm)	Cast Stone Cr500-1 (g)	Volume Titrated FAS (mL)	Fe (mole e-)	Cast Stone (mole e-)	Cast Stone (mole e-/g)	Reduction Capacity (meq/g)	Reduction Capacity (µeq/g)	Comment
1	0.75	0.5062	27.67	0.001325	0.000195	0.000385	0.385043	385	air bubble in line
2	2.25	0.5097	25.42	0.001217	0.000303	0.000594	0.593799	594	
3	3.75	0.5102	25.63	0.001227	0.000293	0.000574	0.573505	574	
4	5.25	0.4929	25.88	0.001239	0.000281	0.000569	0.569345	569	
5	6.75	0.5036	25.98	0.001244	0.000276	0.000548	0.547739	548	
6	8.25	0.5010	26.13	0.001251	0.000269	0.000536	0.536243	536	
7	9.75	0.5082	25.57	0.001225	0.000295	0.000581	0.581416	581	
8	11.25	0.5013	26.10	0.001250	0.000270	0.000539	0.538788	539	
9	12.75	0.5102	25.52	0.001222	0.000298	0.000584	0.583830	584	
10	14.25	0.4976	24.95	0.001195	0.000325	0.000653	0.653471	653	
11	15.75	0.4983	25.18	0.001206	0.000314	0.000630	0.630449	630	
12	17.25	0.4937	24.16	0.001157	0.000363	0.000735	0.735263	735	auto titrator not zeroed
13	18.75	0.5052	25.53	0.001223	0.000297	0.000589	0.588661	589	
14	20.25	0.4995	34.24	0.001640	-0.000120	-0.000240	-0.239685	-240	no indicator
15	21.75	0.5041	25.32	0.001213	0.000307	0.000610	0.609895	610	
16	23.25	0.4988	25.32	0.001213	0.000307	0.000616	0.616375	616	
17	24.75	0.4933	25.32	0.001213	0.000307	0.000623	0.623248	623	
18	26.25	0.4986	25.32	0.001213	0.000307	0.000617	0.616623	617	
19	27.75	0.5010	25.32	0.001213	0.000307	0.000614	0.613669	614	
20-21	36.8	0.5058	25.32	0.001213	0.000307	0.000608	0.607845	608	
22-23	44.8	0.5047	25.32	0.001213	0.000307	0.000609	0.609170	609	
24-25	52.8	0.5034	25.32	0.001213	0.000307	0.000611	0.610743	611	
Average of Subsamples 2 to 11 plus 13 plus 15 to 25								595	

**Table 3-11. Reduction Capacity Results for Cast Stone Tc2 Depth Discrete Subsamples.**

FAS Molarity (calc) 0.05225163 calculated

Ce Molarity 0.0608 theoretical

Ce total (mole e-) 0.001520

Sub Sample No.	Distance From Exposed Surface (Top) (mm)	Cast Stone Tc2 (g)	Volume Titrated FAS (mL)	Fe (mole e-)	Cast Stone (mole e-)	Cast Stone (mole e-/g)	Reduction Capacity (meq/g)	Reduction Capacity (µeq/g)	Comment
1	0.5	0.5167	24.56	0.001283	0.000237	0.000458	0.458099	458	air bubble in line
2	2.5	0.5038	24.28	0.001269	0.000251	0.000499	0.498869	499	
3	4.5	0.5056	23.61	0.001234	0.000286	0.000566	0.566335	566	
4	5.5	0.4983	23.20	0.001212	0.000308	0.000618	0.617624	618	
5	6.5	0.5045	23.09	0.001206	0.000314	0.000621	0.621427	621	
6	8.5	0.5083	23.67	0.001237	0.000283	0.000557	0.557159	557	
7	9.5	0.5006	23.20	0.001212	0.000308	0.000615	0.614786	615	
8	10.5	0.5100	23.11	0.001208	0.000312	0.000613	0.612676	613	
9	11.5	0.5022	22.81	0.001192	0.000328	0.000653	0.653406	653	
10	13.5	0.5138	23.48	0.001227	0.000293	0.000571	0.570517	571	
11	14.5	0.5092	23.58	0.001232	0.000288	0.000565	0.565409	565	
12	15.5	0.5032	23.66	0.001236	0.000284	0.000564	0.563844	564	
13	16.5	0.5019	22.79	0.001191	0.000329	0.000656	0.655878	656	refilled titrator
14	18.5	0.5025	23.90	0.001249	0.000271	0.000540	0.539674	540	
15	21.5	0.5079	24.10	0.001259	0.000261	0.000513	0.513360	513	
16	24.5	0.5026	23.69	0.001238	0.000282	0.000561	0.561398	561	
17	27.5	0.5006	23.60	0.001233	0.000287	0.000573	0.573035	573	
18	29.5	0.5181	24.05	0.001257	0.000263	0.000508	0.508296	508	
19	32.5	0.4852	23.99	0.001254	0.000266	0.000549	0.549224	549	
20	37.5	0.4736	24.44	0.001277	0.000243	0.000513	0.513028	513	
Average of Subsamples 2 to 12 plus 14 to 20								567	

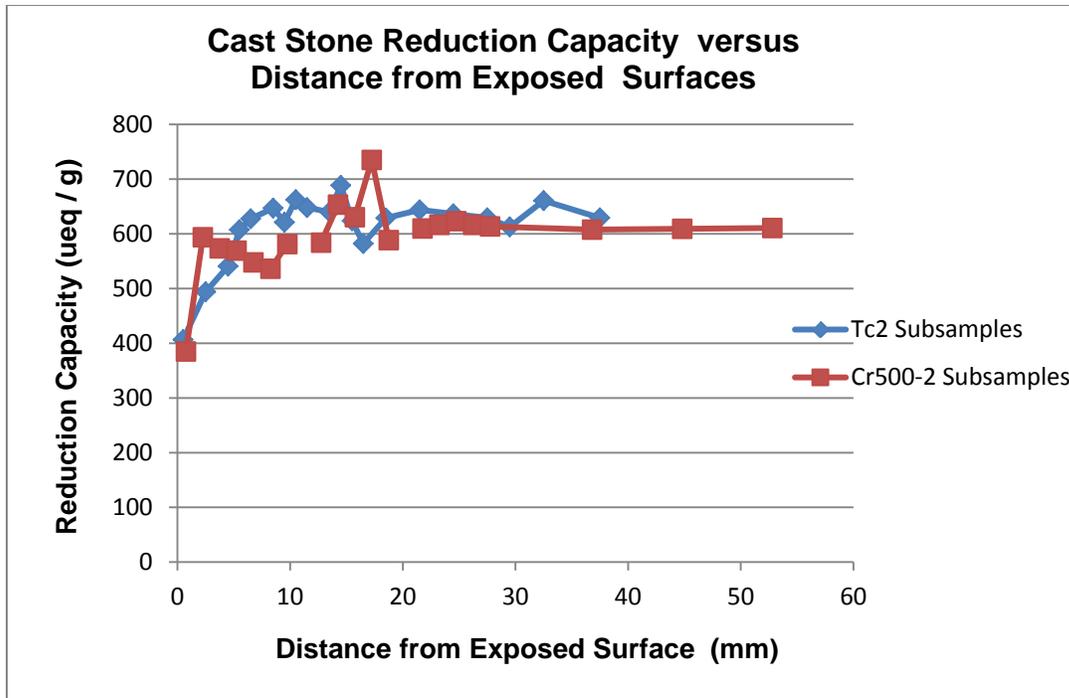


Figure 3-4. Reduction Capacity Results for Cr500-1 and Tc2 Depth Discrete Subsamples.

#### 4.0 Conclusions

Based on the results presented in this report, leaching crushed depth discrete subsamples appears to be a suitable method for evaluating the effects of exposure to moist air and other environmental conditions expected in disposal environments on solubility / leachability of redox sensitive contaminants in cementitious waste forms. The method is useful in identifying the contaminant specific oxidation fronts in cured chemically reducing cementitious waste forms and provides the mass fraction of soluble redox sensitive contaminants and other soluble contaminants as a function of distance from a surface exposed to moist air. The soluble fraction of a chemical species is that portion available to diffusion through the waste form pore solution. The effective diffusion coefficients for the soluble fractions of redox sensitive contaminants will be approximately the same as that for nitrate.

The top 10 mm of the samples tested appears to be slightly different than the bulk material based on visual appearance and leachability of soluble species. This suggests that the fresh samples may have experienced a minor amount of settling prior to setting and / or that exposure to moist air results in redistribution of the soluble species in the near-surface regions of the samples, or both.

Based on the preliminary depth discrete subsample leaching results, Tc(IV) appears to be more easily oxidized than Cr(III). Cr(III) can also substitute for Fe(III) in calcium alumino-ferrite monosulfate hydrate structures (AFm phases) or other hydrated crystalline and non-crystalline phases in the matrix. Tc(IV) and Tc(VII) have been reported to be present in cementitious waste forms as a sulfide (reduced environment) or oxide or hydrated oxide phases (oxidized or partially oxidized environments).<sup>12, 13</sup> However, due to the Tc (IV) and Tc(VII) ions size and charge, it is less likely that either will substitute for ions in calcium silicate or calcium aluminate cementitious phases.

The oxidation front and leachability of redox sensitive contaminants appears to be contaminant specific. The Cr(III)/Cr(VI) oxidation front in Cast Stone was identified at about 35mm below the top surface for a 111 day old sample which was exposed to moist air for 68 days. The oxidation front for Tc in Cast Stone was not identified. However, it appears to be more than 38 mm below the surface of a 98 day old Cast Stone sample exposed to moist air for 50 days. Based on the results presented in this report, other possible interpretations of the data include: 1) the possibility that about 5 wt. % of the Tc in the Cast Stone sample may have oxidized very quickly as a result of the subsampling process and / or 2) some of the Tc(VII) in the sample may never have been reduced. Additional testing is needed.

The reduction capacity measured for the 5 M Na waste simulant by the Angus-Glasser Ce(IV)-Fe(II) method was higher than any of the solid ingredients or resulting Cast Stone waste forms. Nitrite,  $\text{NO}_2^-$ , dissolved in the 5 M Na HTWOS Average simulant is the most likely reductant contributing to the high reduction capacity measured by the Ce(IV)-Fe(II) method. Reduction capacity measurements made on salt solutions containing nitrite using this method do not correlate with and are not an indication of the speciation of certain redox sensitive contaminants such as Tc and Cr because these ions are present in the nitrite containing solutions in the oxidized form, i.e., pertechnetate, Tc(VII), and chromate, Cr(VI).

Furthermore, reduction capacity measurements performed according to the Angus-Glasser method did not correlate with the fraction of redox sensitive contaminants (Cr and Tc) leached in de-aerated deionized leachate. Consequently, reduction capacity measured by this method is not predictive of the reducing effect on contaminants. Furthermore, it is not an appropriate or meaningful parameter for determining or predicting Tc and Cr oxidation states, speciation, and solubilities in cementitious materials such as Cast Stone.

Finally, the kinetics of Tc reduction in Cast Stone appears to be relatively fast. Ninety five percent of the soluble / leachable Tc in Cast Stone with Tc-99 concentration of  $7.79\text{E-}02\mu\text{Ci T / g}$  ( $2.88\text{ kBq / g}$ ) was insoluble (assumed to be as the result of chemical reduction and precipitation of a low solubility solid phase) after the sample was cured for 10 days at ambient laboratory conditions.

Measurement error was not evaluated in this study, which was intended to be a scoping study to determine the usefulness of depth discrete sampling to determine oxidation fronts of redox sensitive contaminants. The method developed for non rad redox sensitive contaminants was successfully adapted to work in a radioactive laboratory and hood.

## 5.0 Recommendations and Future Work

The results of this study demonstrate the effects of environmental exposure on the solubility and leachability of redox sensitive contaminants in cementitious waste forms such as Cast Stone. More specifically, they demonstrate that the redox sensitive species, Cr and Tc, are oxidized to soluble species when the Cast Stone is exposed to moist air (containing oxygen) at ambient temperatures.

Longer exposure times and additional exposure environments are needed to determine the rates of Cr and Tc oxidation in Cast Stone exposed to moist air and to the other conditions expected in the disposal container and disposal site. The bounding conditions for the realistic storage and disposal environments are required to develop test conditions that are relevant to actual disposal conditions. In addition, evaluation of the impact of fractures on the Cast Stone contaminant specific oxidation rates is also recommended.

Depth discrete sampling and leaching is recommended for evaluating Cast Stone and other chemically reducing waste forms containing GGBFS and / or other reduction / sequestration reagents to control redox sensitive contaminant chemistry and leachability in the near surface disposal environment.

Based on results presented in this report, reduction capacity measured by the Angus-Glasser Ce(IV)-Fe(II) method is not an appropriate or meaningful parameter for determining or predicting Tc and Cr oxidation states, speciation, or solubilities in cementitious materials such as Cast Stone. A model for predicting Tc(IV) oxidation to soluble Tc(VII) should consider the waste form porosity (pathway for oxygen ingress), oxygen source, and the contaminant specific oxidation rates and oxidation fronts.

Finally Tc(VII) reduction to Tc(IV) appears to occur relatively fast. Results demonstrated that about 95 percent of the Tc(VII) was reduced to Tc(IV) during the setting and very early stage setting for a Cast Stone sample cured 10 days. Additional testing at longer curing times is required to determine whether additional time is required to reduce 100 % of the Tc(VII) in Cast Stone or whether the Tc loading exceeded the ability of the waste form to reduce 100 % of the Tc(VII). Additional testing is required for samples cured for longer times. Depth discrete subsampling in a nitrogen glove box is also required to determine whether the 5 percent Tc extracted from the subsamples was the result of the sampling process which took place in air.

## 6.0 References

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**Appendix A. Analytical Data for 5 M Cast Stone Cr500-1 Depth Discrete Subsamples**

Units (mg/L)		Lab ID: 13-1017-1046											
<u>Sample ID</u>	<u>Lab ID</u>	<u>Al</u>	<u>As</u>	<u>Ba</u>	<u>Ca</u>	<u>Cr</u>	<u>Cu</u>	<u>Fe</u>	<u>Ga</u>	<u>K</u>	<u>Mg</u>	<u>Mn</u>	<u>Mo</u>
Caststone Cr500-1 Leach 1 (A)	13-1023	9.90	0.039	<0.010	2.52	0.119	<0.100	0.054	<0.010	18.1	<0.010	<0.010	0.014
Caststone Cr500-1 Leach 1 (B)	13-1023	9.90	0.034	<0.010	2.54	0.119	<0.100	0.054	<0.010	18.3	<0.010	<0.010	0.013
Caststone Cr500-1 Leach 2 (A)	13-1024	25.9	0.037	<0.010	3.92	0.242	<0.100	0.147	0.015	36.2	<0.010	<0.010	0.046
Caststone Cr500-1 Leach 2 (B)	13-1024	25.8	0.045	<0.010	3.93	0.242	<0.100	0.148	0.014	36.5	<0.010	<0.010	0.045
Caststone Cr500-1 Leach 3 (A)	13-1025	43.8	<0.010	<0.010	61.2	0.100	<0.100	0.042	0.032	53.3	<0.010	<0.010	0.046
Caststone Cr500-1 Leach 3 (B)	13-1025	44.8	<0.010	<0.010	63.1	0.099	<0.100	0.042	0.033	53.8	<0.010	<0.010	0.044
Caststone Cr500-1 Leach 4 (A)	13-1026	39.1	<0.010	<0.010	36.6	0.052	<0.100	0.119	0.030	53.7	<0.010	<0.010	0.035
Caststone Cr500-1 Leach 4 (B)	13-1026	39.4	0.011	<0.010	36.8	0.055	<0.100	0.120	0.031	53.6	<0.010	<0.010	0.036
Caststone Cr500-1 Leach 5 (A)	13-1027	41.5	<0.010	<0.010	36.6	0.066	<0.100	0.132	0.038	59.9	<0.010	<0.010	0.040
Caststone Cr500-1 Leach 5 (B)	13-1027	41.5	<0.010	<0.010	36.7	0.067	<0.100	0.131	0.040	60.1	<0.010	<0.010	0.041
Caststone Cr500-1 Leach 6 (A)	13-1028	44.5	<0.010	<0.010	35.6	0.096	<0.100	0.037	0.037	60.6	<0.010	<0.010	0.039
Caststone Cr500-1 Leach 6 (B)	13-1028	44.4	<0.010	<0.010	35.5	0.096	<0.100	0.037	0.037	60.2	<0.010	<0.010	0.039
Caststone Cr500-1 Leach 7 (A)	13-1029	43.1	<0.010	<0.010	12.4	0.085	<0.100	0.087	0.044	54.3	<0.010	<0.010	0.046
Caststone Cr500-1 Leach 7 (B)	13-1029	44.3	<0.010	<0.010	12.4	0.084	<0.100	0.085	0.047	55.0	<0.010	<0.010	0.045
Caststone Cr500-1 Leach 8 (A)	13-1030	42.5	<0.010	<0.010	35.0	0.080	<0.100	0.077	0.044	67.1	<0.010	<0.010	0.041
Caststone Cr500-1 Leach 8 (B)	13-1030	43.1	<0.010	<0.010	35.6	0.086	<0.100	0.082	0.047	65.9	<0.010	<0.010	0.043
Caststone Cr500-1 Leach 9 (A)	13-1031	64.7	0.025	<0.010	20.0	0.211	<0.100	0.084	0.067	67.8	<0.010	<0.010	0.059
Caststone Cr500-1 Leach 9 (B)	13-1031	65.1	0.019	<0.010	20.2	0.215	<0.100	0.086	0.070	67.6	<0.010	<0.010	0.055
Caststone Cr500-1 Leach 10 (A)	13-1032	59.5	0.019	<0.010	26.6	0.189	<0.100	0.076	0.066	64.3	<0.010	<0.010	0.049
Caststone Cr500-1 Leach 10 (B)	13-1032	60.1	0.019	<0.010	26.9	0.186	<0.100	0.077	0.066	65.8	<0.010	<0.010	0.049
Caststone Cr500-1 Leach 11 (A)	13-1033	58.7	0.018	<0.010	19.6	0.195	<0.100	0.079	0.062	61.6	<0.010	<0.010	0.047
Caststone Cr500-1 Leach 11 (B)	13-1033	57.9	0.014	<0.010	19.8	0.199	<0.100	0.083	0.068	61.7	<0.010	<0.010	0.050

<u>Sample ID</u>	<u>Lab ID</u>	<u>Na</u>	<u>S</u>	<u>Se</u>	<u>Si</u>	<u>Sn</u>	<u>Sr</u>	<u>V</u>	<u>W</u>	<u>Zn</u>
Caststone Cr500-1 Leach 1 (A)	13-1023	569	113	<0.010	27.3	<0.200	<0.100	0.310	0.111	<0.010
Caststone Cr500-1 Leach 1 (B)	13-1023	576	111	<0.010	27.3	<0.200	<0.100	0.309	0.106	<0.010
Caststone Cr500-1 Leach 2 (A)	13-1024	1140	230	<0.010	31.3	<0.200	<0.100	0.413	0.114	<0.010
Caststone Cr500-1 Leach 2 (B)	13-1024	1150	229	<0.010	31.6	<0.200	<0.100	0.418	0.115	<0.010
Caststone Cr500-1 Leach 3 (A)	13-1025	1270	229	<0.010	13.0	<0.200	0.304	0.183	0.116	<0.010
Caststone Cr500-1 Leach 3 (B)	13-1025	1280	228	<0.010	12.9	<0.200	0.304	0.183	0.114	<0.010
Caststone Cr500-1 Leach 4 (A)	13-1026	1110	188	<0.010	11.3	<0.200	0.438	0.143	0.116	<0.010
Caststone Cr500-1 Leach 4 (B)	13-1026	1100	187	<0.010	11.6	<0.200	0.451	0.146	0.114	<0.010
Caststone Cr500-1 Leach 5 (A)	13-1027	1200	193	<0.010	12.3	<0.200	0.475	0.156	0.117	<0.010
Caststone Cr500-1 Leach 5 (B)	13-1027	1210	193	<0.010	12.4	<0.200	0.479	0.157	0.115	<0.010
Caststone Cr500-1 Leach 6 (A)	13-1028	1200	195	<0.010	13.9	<0.200	0.450	0.163	0.116	<0.010
Caststone Cr500-1 Leach 6 (B)	13-1028	1200	193	<0.010	13.9	<0.200	0.438	0.163	0.115	<0.010
Caststone Cr500-1 Leach 7 (A)	13-1029	1070	221	<0.010	13.3	<0.200	0.393	0.174	0.116	<0.010
Caststone Cr500-1 Leach 7 (B)	13-1029	1070	220	<0.010	13.2	<0.200	0.392	0.175	0.118	<0.010
Caststone Cr500-1 Leach 8 (A)	13-1030	1300	189	<0.010	13.3	<0.200	0.472	0.169	0.119	<0.010
Caststone Cr500-1 Leach 8 (B)	13-1030	1290	193	<0.010	13.9	<0.200	0.507	0.177	0.115	<0.010
Caststone Cr500-1 Leach 9 (A)	13-1031	1470	261	<0.010	14.9	<0.200	0.205	0.312	0.156	<0.010
Caststone Cr500-1 Leach 9 (B)	13-1031	1470	261	<0.010	15.2	<0.200	0.208	0.315	0.126	<0.010
Caststone Cr500-1 Leach 10 (A)	13-1032	1370	239	<0.010	15.1	<0.200	0.223	0.295	0.120	<0.010
Caststone Cr500-1 Leach 10 (B)	13-1032	1390	236	<0.010	14.9	<0.200	0.222	0.294	0.120	<0.010
Caststone Cr500-1 Leach 11 (A)	13-1033	1320	232	<0.010	15.2	<0.200	0.215	0.302	0.117	<0.010
Caststone Cr500-1 Leach 11 (B)	13-1033	1310	234	<0.010	15.8	<0.200	0.225	0.310	0.119	<0.010

Sample ID	Lab ID	Al	As	Ba	Ca	Cr	Cu	Fe	Ga	K	Mg	Mn	Mo
Caststone Cr500-1 Leach 12 (A)	13-1034	68.2	0.018	<0.010	20.4	0.198	<0.100	0.109	0.084	73.0	<0.010	<0.010	0.062
Caststone Cr500-1 Leach 12 (B)	13-1034	68.4	0.022	<0.010	20.6	0.203	<0.100	0.113	0.087	73.5	<0.010	<0.010	0.064
Caststone Cr500-1 Leach 13 (A)	13-1035	65.2	0.021	<0.010	21.3	0.218	<0.100	0.098	0.085	65.9	<0.010	<0.010	0.060
Caststone Cr500-1 Leach 13 (B)	13-1035	66.2	0.025	<0.010	21.6	0.217	<0.100	0.099	0.084	65.4	<0.010	<0.010	0.059
Caststone Cr500-1 Leach 14 (A)	13-1036	70.0	0.030	<0.010	37.3	0.204	<0.100	0.121	0.096	70.1	<0.010	<0.010	0.068
Caststone Cr500-1 Leach 14 (B)	13-1036	70.5	0.031	<0.010	37.7	0.205	<0.100	0.124	0.097	70.2	<0.010	<0.010	0.068
Caststone Cr500-1 Leach 15 (A)	13-1037	62.8	0.024	<0.010	22.3	0.164	<0.100	0.098	0.083	56.9	<0.010	<0.010	0.055
Caststone Cr500-1 Leach 15 (B)	13-1037	61.0	0.021	<0.010	22.2	0.162	<0.100	0.094	0.080	57.2	<0.010	<0.010	0.055
Caststone Cr500-1 Leach 16 (A)	13-1038	70.3	0.025	<0.010	22.2	0.258	<0.100	0.116	0.093	67.9	<0.010	<0.010	0.067
Caststone Cr500-1 Leach 16 (B)	13-1038	69.5	0.024	<0.010	22.1	0.252	<0.100	0.111	0.089	68.5	<0.010	<0.010	0.064
Caststone Cr500-1 Leach 17 (A)	13-1039	65.7	0.037	<0.010	23.3	0.188	<0.100	0.094	0.082	61.0	<0.010	<0.010	0.058
Caststone Cr500-1 Leach 17 (B)	13-1039	65.9	0.021	<0.010	23.5	0.191	<0.100	0.094	0.081	59.8	<0.010	<0.010	0.056
Caststone Cr500-1 Leach 18 (A)	13-1040	72.1	0.023	<0.010	24.9	0.174	<0.100	0.129	0.090	63.4	<0.010	<0.010	0.064
Caststone Cr500-1 Leach 18 (B)	13-1040	71.2	0.016	<0.010	24.4	0.173	<0.100	0.124	0.088	65.2	<0.010	<0.010	0.061
Caststone Cr500-1 Leach 19 (A)	13-1041	71.8	0.024	<0.010	25.6	0.184	<0.100	0.124	0.09	66.7	<0.010	<0.010	0.066
Caststone Cr500-1 Leach 19 (B)	13-1041	72.7	0.022	<0.010	26.6	0.178	<0.100	0.117	0.086	67.6	<0.010	<0.010	0.062
Caststone Cr500-1 Leach 20/21 (A)	13-1042	128	0.085	<0.010	21.4	<0.010	<0.100	0.769	0.226	158	<0.010	<0.010	0.161
Caststone Cr500-1 Leach 20/21 (B)	13-1042	128	0.090	<0.010	21.4	<0.010	<0.100	0.817	0.237	154	<0.010	<0.010	0.167
Caststone Cr500-1 Leach 22/23 (A)	13-1043	124	0.073	<0.010	21.2	<0.010	<0.100	0.729	0.210	148	<0.010	<0.010	0.150
Caststone Cr500-1 Leach 22/23 (B)	13-1043	124	0.082	<0.010	21.1	<0.010	<0.100	0.760	0.220	145	<0.010	<0.010	0.153
Caststone Cr500-1 Leach 24/25 (A)	13-1044	125	0.097	<0.010	21.4	<0.010	<0.100	0.837	0.249	159	<0.010	<0.010	0.173
Caststone Cr500-1 Leach 24/25 (B)	13-1044	117	0.092	<0.010	18.9	<0.010	<0.100	0.823	0.232	156	<0.010	<0.010	0.166
Caststone Cr500-1 Leach 27 Blank (A)	13-1046	0.439	<0.010	<0.010	13.5	<0.010	<0.100	<0.010	<0.010	<0.100	<0.010	<0.010	<0.010
Caststone Cr500-1 Leach 27 Blank (B)	13-1046	0.355	<0.010	<0.010	12.1	<0.010	<0.100	<0.010	<0.010	<0.100	<0.010	<0.010	<0.010

Sample ID	Lab ID	Na	S	Se	Si	Sn	Sr	V	W	Zn
Caststone Cr500-1 Leach 12 (A)	13-1034	1550	278	<0.010	16.9	<0.200	0.239	0.347	0.119	<0.010
Caststone Cr500-1 Leach 12 (B)	13-1034	1560	277	<0.010	17.5	<0.200	0.243	0.354	0.120	<0.010
Caststone Cr500-1 Leach 13 (A)	13-1035	1390	242	<0.010	18.6	<0.200	0.256	0.355	0.120	<0.010
Caststone Cr500-1 Leach 13 (B)	13-1035	1390	241	<0.010	18.6	<0.200	0.259	0.356	0.121	<0.010
Caststone Cr500-1 Leach 14 (A)	13-1036	1510	268	<0.010	19.3	<0.200	0.263	0.385	0.124	<0.010
Caststone Cr500-1 Leach 14 (B)	13-1036	1520	265	<0.010	19.5	<0.200	0.271	0.389	0.128	<0.010
Caststone Cr500-1 Leach 15 (A)	13-1037	1240	220	<0.010	19.4	<0.200	0.255	0.347	0.118	<0.010
Caststone Cr500-1 Leach 15 (B)	13-1037	1240	224	<0.010	19.1	<0.200	0.254	0.345	0.116	<0.010
Caststone Cr500-1 Leach 16 (A)	13-1038	1470	265	<0.010	22.9	<0.200	0.259	0.381	0.120	<0.010
Caststone Cr500-1 Leach 16 (B)	13-1038	1500	259	<0.010	22.2	<0.200	0.244	0.371	0.122	<0.010
Caststone Cr500-1 Leach 17 (A)	13-1039	1320	232	<0.010	19.0	<0.200	0.246	0.355	0.121	<0.010
Caststone Cr500-1 Leach 17 (B)	13-1039	1300	235	<0.010	18.9	<0.200	0.246	0.355	0.118	<0.010
Caststone Cr500-1 Leach 18 (A)	13-1040	1380	253	<0.010	19.0	<0.200	0.250	0.376	0.120	<0.010
Caststone Cr500-1 Leach 18 (B)	13-1040	1410	246	<0.010	18.6	<0.200	0.248	0.368	0.121	<0.010
Caststone Cr500-1 Leach 19 (A)	13-1041	1440	258	<0.010	19.6	<0.200	0.257	0.382	0.121	<0.010
Caststone Cr500-1 Leach 19 (B)	13-1041	1450	257	<0.010	18.9	<0.200	0.248	0.369	0.124	<0.010
Caststone Cr500-1 Leach 20/21 (A)	13-1042	3220	608	0.094	22.1	<0.200	0.292	0.662	0.145	<0.010
Caststone Cr500-1 Leach 20/21 (B)	13-1042	3200	612	0.118	23.3	<0.200	0.311	0.688	0.141	<0.010
Caststone Cr500-1 Leach 22/23 (A)	13-1043	3030	584	0.085	21.4	<0.200	0.289	0.636	0.143	<0.010
Caststone Cr500-1 Leach 22/23 (B)	13-1043	2980	579	0.086	22.1	<0.200	0.302	0.650	0.140	<0.010
Caststone Cr500-1 Leach 24/25 (A)	13-1044	3240	621	0.113	23.9	<0.200	0.355	0.704	0.142	<0.010
Caststone Cr500-1 Leach 24/25 (B)	13-1044	3220	623	0.105	22.8	<0.200	0.329	0.683	0.142	<0.010
Caststone Cr500-1 Leach 27 Blank (A)	13-1046	4.00	<0.100	<0.010	0.591	<0.200	<0.100	<0.010	0.104	<0.010
Caststone Cr500-1 Leach 27 Blank (B)	13-1046	3.97	<0.100	<0.010	0.557	<0.200	<0.100	<0.010	0.100	<0.010

Sample ID	Lab ID	NO2	NO3	SO4	pH
Caststone Cr500-1 Leach 1 (A)	13-1023	138	536	<100	12.4
Caststone Cr500-1 Leach 1 (B)	13-1023	137	535	<100	
Caststone Cr500-1 Leach 2 (A)	13-1024	290	1080	117	12.9
Caststone Cr500-1 Leach 2 (B)	13-1024	288	1080	119	
Caststone Cr500-1 Leach 3 (A)	13-1025	330	1210	<100	13.3
Caststone Cr500-1 Leach 3 (B)	13-1025	328	1210	<100	
Caststone Cr500-1 Leach 4 (A)	13-1026	272	1020	<100	13.4
Caststone Cr500-1 Leach 4 (B)	13-1026	272	1020	<100	
Caststone Cr500-1 Leach 5 (A)	13-1027	286	1080	<100	13.5
Caststone Cr500-1 Leach 5 (B)	13-1027	285	1080	<100	
Caststone Cr500-1 Leach 6 (A)	13-1028	281	1050	<100	13.5
Caststone Cr500-1 Leach 6 (B)	13-1028	279	1050	<100	
Caststone Cr500-1 Leach 7 (A)	13-1029	331	1250	100	13.6
Caststone Cr500-1 Leach 7 (B)	13-1029	329	1250	101	
Caststone Cr500-1 Leach 8 (A)	13-1030	292	1100	<100	13.6
Caststone Cr500-1 Leach 8 (B)	13-1030	290	1100	<100	
Caststone Cr500-1 Leach 9 (A)	13-1031	353	1330	184	13.5
Caststone Cr500-1 Leach 9 (B)	13-1031	352	1330	185	
Caststone Cr500-1 Leach 10 (A)	13-1032	335	1250	158	13.5
Caststone Cr500-1 Leach 10 (B)	13-1032	332	1250	156	
Caststone Cr500-1 Leach 11 (A)	13-1033	313	1180	140	13.4
Caststone Cr500-1 Leach 11 (B)	13-1033	311	1180	136	
Caststone Cr500-1 Leach 12 (A)	13-1034	369	1400	222	13.5
Caststone Cr500-1 Leach 12 (B)	13-1034	369	1400	222	
Caststone Cr500-1 Leach 13 (A)	13-1035	328	1250	173	13.5
Caststone Cr500-1 Leach 13 (B)	13-1035	327	1250	158	
Caststone Cr500-1 Leach 14 (A)	13-1036	351	1340	202	13.5
Caststone Cr500-1 Leach 14 (B)	13-1036	350	1340	196	
Caststone Cr500-1 Leach 15 (A)	13-1037	297	1130	125	13.4
Caststone Cr500-1 Leach 15 (B)	13-1037	296	1130	119	
Caststone Cr500-1 Leach 16 (A)	13-1038	345	1310	198	13.5
Caststone Cr500-1 Leach 16 (B)	13-1038	345	1320	189	
Caststone Cr500-1 Leach 17 (A)	13-1039	306	1170	142	13.4
Caststone Cr500-1 Leach 17 (B)	13-1039	306	1170	135	
Caststone Cr500-1 Leach 18 (A)	13-1040	333	1270	180	13.5
Caststone Cr500-1 Leach 18 (B)	13-1040	332	1270	177	
Caststone Cr500-1 Leach 19 (A)	13-1041	340	1300	193	13.5
Caststone Cr500-1 Leach 19 (B)	13-1041	339	1300	183	
Caststone Cr500-1 Leach 20/21 (A)	13-1042	816	3180	865	13.7
Caststone Cr500-1 Leach 20/21 (B)	13-1042	816	3190	863	
Caststone Cr500-1 Leach 22/23 (A)	13-1043	777	3040	808	13.7
Caststone Cr500-1 Leach 22/23 (B)	13-1043	776	3040	800	
Caststone Cr500-1 Leach 24/25 (A)	13-1044	830	3240	884	13.7
Caststone Cr500-1 Leach 24/25 (B)	13-1044	829	3240	882	
Caststone Cr500-1 Leach 27 Blank (A)	13-1046	<100	<100	<100	6.42
Caststone Cr500-1 Leach 27 Blank (B)	13-1046	<100	<100	<100	

**Appendix B. Analytical Data for 5 M Cast Stone Tc2 Depth Discrete Subsamples**

## ICP-OES Results

Description: CAST STONE TC-99 LEACH

Travel Copy: 64958

Instrument: Leeman Prodigy ICP-ES

Reviewer: John Young

Comments:

Method Detection Limit (MDL) = Instrument Detection Limit (IDL) x Dilution Factor.  
 Uncertainty is the RMS of the method uncertainty and the sample uncertainty.

USER_SAMPLEID: SAMPLE_ID: UNITS:	TC2 LEACH 1 300306611 mg/L	TC2 LEACH 2 300306612 mg/L	TC2 LEACH 3 300306613 mg/L	TC2 LEACH 4 300306614 mg/L	TC2 LEACH 5 300306615 mg/L
Element					
Ag	< 0.212 (N/A %RSD)				
Al	38.2 (10 %RSD)	32.5 (10 %RSD)	30.9 (10 %RSD)	28.9 (10 %RSD)	34.2 (10 %RSD)
B	1.75 (15.1 %RSD)	1.96 (17.6 %RSD)	1.95 (15.5 %RSD)	1.69 (11.4 %RSD)	2.03 (12.9 %RSD)
Ba	< 0.106 (N/A %RSD)				
Be	< 0.02 (N/A %RSD)				
Ca	22.7 (10 %RSD)	30.1 (10 %RSD)	33.6 (10 %RSD)	37 (10 %RSD)	30.5 (10 %RSD)
Cd	< 0.244 (N/A %RSD)				
Ce	< 2.01 (N/A %RSD)				
Co	< 0.422 (N/A %RSD)				
Cr	< 0.326 (N/A %RSD)				
Cu	< 0.18 (N/A %RSD)				
Fe	< 0.452 (N/A %RSD)				
Gd	< 0.47 (N/A %RSD)				
K	50.7 (10.1 %RSD)	58.4 (10.1 %RSD)	56.4 (10.2 %RSD)	51.9 (10 %RSD)	72 (10.6 %RSD)
La	< 0.156 (N/A %RSD)				
Li	< 0.19 (N/A %RSD)				
Mg	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)	0.068 (10.1 %RSD)	< 0.02 (N/A %RSD)	0.021 (10 %RSD)
Mn	< 0.046 (N/A %RSD)				
Mo	< 1.66 (N/A %RSD)				
Na	1120 (10 %RSD)	1210 (10 %RSD)	1160 (10 %RSD)	1040 (10 %RSD)	1490 (10 %RSD)
Ni	< 2.39 (N/A %RSD)				
P	< 7.09 (N/A %RSD)				
Pb	< 10.6 (N/A %RSD)				
S	< 160 (N/A %RSD)	179 (25.3 %RSD)	< 160 (N/A %RSD)	< 160 (N/A %RSD)	167 (19.4 %RSD)
Sb	< 5.79 (N/A %RSD)				
Si	14.7 (10.1 %RSD)	12.5 (10 %RSD)	12.1 (10 %RSD)	10.2 (10.2 %RSD)	12.5 (10.3 %RSD)
Sn	< 5.37 (N/A %RSD)				
Sr	0.207 (10 %RSD)	0.337 (10.1 %RSD)	0.365 (10.2 %RSD)	0.41 (10.2 %RSD)	0.369 (10 %RSD)
Th	< 1.13 (N/A %RSD)				
Ti	< 0.138 (N/A %RSD)				
U	< 8.8 (N/A %RSD)				
V	0.22 (16.8 %RSD)	0.202 (17.8 %RSD)	0.212 (12.6 %RSD)	0.162 (20.2 %RSD)	0.232 (19.1 %RSD)
Zn	< 0.16 (N/A %RSD)				
Zr	< 4.38 (N/A %RSD)				

Method Detectic  
Uncertainty is

USER_SAM SAMP	TC2 LEACH 6 300306616 mg/L	TC2 LEACH 7 300306617 mg/L	TC2 LEACH 8 300306618 mg/L	TC2 LEACH 9 300306619 mg/L	TC2 LEACH 10 300306620 mg/L
Element					
Ag	< 0.212 (N/A %RSD)				
Al	30.4 (10 %RSD)	24 (10 %RSD)	25.4 (10.1 %RSD)	30.4 (10 %RSD)	29.6 (10 %RSD)
B	1.65 (14.1 %RSD)	1.56 (34.3 %RSD)	1.42 (13.2 %RSD)	1.53 (16 %RSD)	1.63 (15.2 %RSD)
Ba	< 0.106 (N/A %RSD)				
Be	< 0.02 (N/A %RSD)				
Ca	34.2 (10 %RSD)	40 (10 %RSD)	37.4 (10 %RSD)	31.7 (10 %RSD)	33 (10 %RSD)
Cd	< 0.244 (N/A %RSD)				
Ce	< 2.01 (N/A %RSD)				
Co	< 0.422 (N/A %RSD)				
Cr	< 0.326 (N/A %RSD)				
Cu	< 0.18 (N/A %RSD)				
Fe	< 0.452 (N/A %RSD)				
Gd	< 0.47 (N/A %RSD)				
K	59.2 (10.4 %RSD)	42.4 (10.1 %RSD)	48.5 (10.1 %RSD)	59.4 (10 %RSD)	60.5 (10.7 %RSD)
La	< 0.156 (N/A %RSD)				
Li	< 0.19 (N/A %RSD)				
Mg	< 0.02 (N/A %RSD)				
Mn	< 0.046 (N/A %RSD)				
Mo	< 1.66 (N/A %RSD)				
Na	1160 (10 %RSD)	845 (10 %RSD)	982 (10 %RSD)	1190 (10 %RSD)	1210 (10 %RSD)
Ni	< 2.39 (N/A %RSD)				
P	< 7.09 (N/A %RSD)				
Pb	< 10.6 (N/A %RSD)				
S	< 160 (N/A %RSD)	< 160 (N/A %RSD)	< 160 (N/A %RSD)	162 (10.9 %RSD)	194 (14.3 %RSD)
Sb	< 5.79 (N/A %RSD)				
Si	12 (10 %RSD)	9.82 (10.2 %RSD)	10.8 (10.1 %RSD)	12.5 (10 %RSD)	11.8 (10.4 %RSD)
Sn	< 5.37 (N/A %RSD)				
Sr	0.391 (10.1 %RSD)	0.416 (10 %RSD)	0.423 (10.1 %RSD)	0.366 (10.1 %RSD)	0.398 (10 %RSD)
Th	< 1.13 (N/A %RSD)				
Ti	< 0.138 (N/A %RSD)				
U	< 8.8 (N/A %RSD)				
V	0.192 (13.6 %RSD)	0.146 (21.9 %RSD)	0.146 (17.9 %RSD)	0.24 (14.5 %RSD)	0.206 (10.7 %RSD)
Zn	< 0.16 (N/A %RSD)				
Zr	< 4.38 (N/A %RSD)				

## ICP-OES Results

Description: CAST STONE TC-99 LEACH

Travel Copy: 64959

Instrument: Leeman Prodigy ICP-ES

Reviewer: John Young

Comments:

Method Detection Limit (MDL) = Instrument Detection Limit (IDL) x Dilution Factor.  
 Uncertainty is the RMS of the method uncertainty and the sample uncertainty.

USER_SAMPLEID: SAMPLE_ID: UNITS:	TC2 LEACH 11 300306621 mg/L	TC2 LEACH 12 300306622 mg/L	TC2 LEACH 13 300306623 mg/L	TC2 LEACH 14 300306624 mg/L	TC2 LEACH 15 300306625 mg/L
Element					
Ag	< 0.212 (N/A %RSD)				
Al	27.9 (10.1 %RSD)	27.8 (10 %RSD)	46.8 (10 %RSD)	50.6 (10 %RSD)	51.7 (10 %RSD)
B	1.41 (11.3 %RSD)	1.42 (27.7 %RSD)	3.19 (11.3 %RSD)	3.43 (14.4 %RSD)	3.46 (10.3 %RSD)
Ba	< 0.106 (N/A %RSD)				
Be	< 0.02 (N/A %RSD)				
Ca	34.3 (10 %RSD)	33.8 (10 %RSD)	20.9 (10 %RSD)	20.8 (10 %RSD)	19.9 (10 %RSD)
Cd	< 0.244 (N/A %RSD)				
Ce	< 2.01 (N/A %RSD)				
Co	< 0.422 (N/A %RSD)				
Cr	< 0.326 (N/A %RSD)				
Cu	< 0.18 (N/A %RSD)				
Fe	< 0.452 (N/A %RSD)	< 0.452 (N/A %RSD)	0.987 (10.4 %RSD)	1.54 (10.2 %RSD)	1.04 (10.2 %RSD)
Gd	< 0.47 (N/A %RSD)				
K	51 (10.2 %RSD)	51.1 (10.2 %RSD)	133 (10.1 %RSD)	137 (10 %RSD)	144 (10 %RSD)
La	< 0.156 (N/A %RSD)				
Li	< 0.19 (N/A %RSD)				
Mg	< 0.02 (N/A %RSD)				
Mn	< 0.046 (N/A %RSD)				
Mo	< 1.66 (N/A %RSD)				
Na	1080 (10 %RSD)	1030 (10 %RSD)	2600 (10 %RSD)	2700 (10 %RSD)	2830 (10 %RSD)
Ni	< 2.39 (N/A %RSD)				
P	< 7.09 (N/A %RSD)				
Pb	< 10.6 (N/A %RSD)				
S	< 160 (N/A %RSD)	< 160 (N/A %RSD)	359 (10.7 %RSD)	371 (10.1 %RSD)	451 (11.8 %RSD)
Sb	< 5.79 (N/A %RSD)				
Si	11.6 (10.3 %RSD)	11.7 (10.1 %RSD)	15.8 (10.1 %RSD)	16.7 (10.1 %RSD)	16.1 (10.1 %RSD)
Sn	< 5.37 (N/A %RSD)				
Sr	0.377 (10.1 %RSD)	0.365 (10.1 %RSD)	0.354 (10 %RSD)	0.325 (10.2 %RSD)	0.34 (10.2 %RSD)
Th	< 1.13 (N/A %RSD)				
Ti	< 0.138 (N/A %RSD)				
U	< 8.8 (N/A %RSD)				
V	0.2 (22.2 %RSD)	0.196 (14.3 %RSD)	0.396 (10.1 %RSD)	0.45 (10.3 %RSD)	0.46 (10.4 %RSD)
Zn	< 0.16 (N/A %RSD)				
Zr	< 4.38 (N/A %RSD)				

## ICP-OES Re:

Description:

Travel Copy:

Instrument:

Reviewer:

Comments:

Method Detectio  
Uncertainty is

USER_SAM SAMP	TC2 LEACH 16 300306626 mg/L	TC2 LEACH 17 30 0306627 mg/L	TC2 LEACH 18 300306628 mg/L	TC2 LEACH 19 300306629 mg/L	TC2 LEACH 20 300306630 mg/L
Element					
Ag	< 0.212 (N/A %RSD)	< 0.212 (N/A %RSD)	< 0.212 (N/A %RSD)	< 0.212 (N/A %RSD)	< 0.212 (N/A %RSD)
Al	51.9 (10 %RSD)	47.6 (10 %RSD)	52.8 (10 %RSD)	46.6 (10 %RSD)	51.5 (10 %RSD)
B	3.59 (11.1 %RSD)	3.16 (10.2 %RSD)	3.65 (10.1 %RSD)	3.14 (12.3 %RSD)	3.19 (11.2 %RSD)
Ba	< 0.106 (N/A %RSD)	< 0.106 (N/A %RSD)	< 0.106 (N/A %RSD)	< 0.106 (N/A %RSD)	< 0.106 (N/A %RSD)
Be	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)
Ca	18.9 (10 %RSD)	19.9 (10 %RSD)	18.9 (10 %RSD)	20.9 (10 %RSD)	21.5 (10 %RSD)
Cd	< 0.244 (N/A %RSD)	< 0.244 (N/A %RSD)	< 0.244 (N/A %RSD)	< 0.244 (N/A %RSD)	< 0.244 (N/A %RSD)
Ce	< 2.01 (N/A %RSD)	< 2.01 (N/A %RSD)	< 2.01 (N/A %RSD)	< 2.01 (N/A %RSD)	< 2.01 (N/A %RSD)
Co	< 0.422 (N/A %RSD)	< 0.422 (N/A %RSD)	< 0.422 (N/A %RSD)	< 0.422 (N/A %RSD)	< 0.422 (N/A %RSD)
Cr	< 0.326 (N/A %RSD)	< 0.326 (N/A %RSD)	< 0.326 (N/A %RSD)	< 0.326 (N/A %RSD)	< 0.326 (N/A %RSD)
Cu	< 0.18 (N/A %RSD)	< 0.18 (N/A %RSD)	< 0.18 (N/A %RSD)	< 0.18 (N/A %RSD)	< 0.18 (N/A %RSD)
Fe	1.66 (10.1 %RSD)	0.962 (10.2 %RSD)	3.09 (10 %RSD)	1.31 (10.3 %RSD)	1.7 (10 %RSD)
Gd	< 0.47 (N/A %RSD)	< 0.47 (N/A %RSD)	< 0.47 (N/A %RSD)	< 0.47 (N/A %RSD)	< 0.47 (N/A %RSD)
K	149 (10.1 %RSD)	135 (10.1 %RSD)	161 (10 %RSD)	133 (10 %RSD)	150 (10.1 %RSD)
La	< 0.156 (N/A %RSD)	< 0.156 (N/A %RSD)	< 0.156 (N/A %RSD)	< 0.156 (N/A %RSD)	< 0.156 (N/A %RSD)
Li	< 0.19 (N/A %RSD)	< 0.19 (N/A %RSD)	< 0.19 (N/A %RSD)	< 0.19 (N/A %RSD)	< 0.19 (N/A %RSD)
Mg	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)	< 0.02 (N/A %RSD)
Mn	< 0.046 (N/A %RSD)	< 0.046 (N/A %RSD)	< 0.046 (N/A %RSD)	< 0.046 (N/A %RSD)	< 0.046 (N/A %RSD)
Mo	< 1.66 (N/A %RSD)	< 1.66 (N/A %RSD)	< 1.66 (N/A %RSD)	< 1.66 (N/A %RSD)	< 1.66 (N/A %RSD)
Na	2890 (10 %RSD)	2630 (10 %RSD)	3040 (10 %RSD)	2590 (10 %RSD)	2870 (10 %RSD)
Ni	< 2.39 (N/A %RSD)	< 2.39 (N/A %RSD)	< 2.39 (N/A %RSD)	< 2.39 (N/A %RSD)	< 2.39 (N/A %RSD)
P	< 7.09 (N/A %RSD)	< 7.09 (N/A %RSD)	< 7.09 (N/A %RSD)	< 7.09 (N/A %RSD)	< 7.09 (N/A %RSD)
Pb	< 10.6 (N/A %RSD)	< 10.6 (N/A %RSD)	< 10.6 (N/A %RSD)	< 10.6 (N/A %RSD)	< 10.6 (N/A %RSD)
S	422 (11.7 %RSD)	392 (10.1 %RSD)	460 (10.3 %RSD)	383 (10 %RSD)	433 (11.2 %RSD)
Sb	< 5.79 (N/A %RSD)	< 5.79 (N/A %RSD)	< 5.79 (N/A %RSD)	< 5.79 (N/A %RSD)	< 5.79 (N/A %RSD)
Si	14.2 (10 %RSD)	14.2 (10.3 %RSD)	14.7 (10.1 %RSD)	15.9 (10 %RSD)	14.4 (10.2 %RSD)
Sn	< 5.37 (N/A %RSD)	< 5.37 (N/A %RSD)	< 5.37 (N/A %RSD)	< 5.37 (N/A %RSD)	< 5.37 (N/A %RSD)
Sr	0.339 (10 %RSD)	0.351 (10 %RSD)	0.374 (10 %RSD)	0.348 (10.2 %RSD)	0.396 (10 %RSD)
Th	< 1.13 (N/A %RSD)	< 1.13 (N/A %RSD)	< 1.13 (N/A %RSD)	< 1.13 (N/A %RSD)	< 1.13 (N/A %RSD)
Ti	< 0.138 (N/A %RSD)	< 0.138 (N/A %RSD)	< 0.138 (N/A %RSD)	< 0.138 (N/A %RSD)	< 0.138 (N/A %RSD)
U	< 8.8 (N/A %RSD)	< 8.8 (N/A %RSD)	< 8.8 (N/A %RSD)	< 8.8 (N/A %RSD)	< 8.8 (N/A %RSD)
V	0.402 (11.7 %RSD)	0.396 (10 %RSD)	0.416 (11 %RSD)	0.39 (12.3 %RSD)	0.392 (11 %RSD)
Zn	< 0.16 (N/A %RSD)	< 0.16 (N/A %RSD)	< 0.16 (N/A %RSD)	< 0.16 (N/A %RSD)	< 0.16 (N/A %RSD)
Zr	< 4.38 (N/A %RSD)	< 4.38 (N/A %RSD)	< 4.38 (N/A %RSD)	< 4.38 (N/A %RSD)	< 4.38 (N/A %RSD)



**Tc-99 data**  
C Diprete to: Christine Langton  
Cc: David Diprete

ADS LIMS #	Tc-99(dpm/ml)	1 Sigma %Unc
300306611	2.39E+03	5.00%
300306612	1.72E+03	5.00%
300306613	1.39E+03	5.00%
300306614	2.20E+03	5.00%
300306615	1.48E+03	5.00%
300306616	1.99E+03	5.00%
300306617	1.44E+03	5.00%
300306618	1.33E+03	5.00%
300306619	1.42E+03	5.00%
300306620	1.46E+03	5.00%



**Tc-99 data**  
C Diprete to: Christine Langton  
Cc: David Diprete

ADS LIMS #	Tc-99(dpm/ml)	1 Sigma %Unc
300306621	1.39E+03	5.00%
300306622	1.37E+03	5.00%
300306623	1.44E+03	5.00%
300306624	1.79E+03	5.00%
300306625	1.97E+03	5.00%
300306626	1.84E+03	5.00%
300306627	1.65E+03	5.00%
300306628	1.60E+03	5.00%
300306629	1.59E+03	5.00%
300306630	1.81E+03	5.00%

SAMPLE_ID	USER_SAMPLEID	OPERATION	COMPONENT	TEXT_VALUE	UNITS
300306611	TC2 LEACH 1	IC ANIONS (B134)	Bromide	<10	UG/ML
300306612	TC2 LEACH 2	IC ANIONS (B134)	Bromide	<10	UG/ML
300306613	TC2 LEACH 3	IC ANIONS (B134)	Bromide	<10	UG/ML
300306614	TC2 LEACH 4	IC ANIONS (B134)	Bromide	<10	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Bromide	<10	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Bromide	<10	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Bromide	<10	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Bromide	<10	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Bromide	<10	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Bromide	<10	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Bromide	<10	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Bromide	<10	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Bromide	<10	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Bromide	<10	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Bromide	<10	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Bromide	<10	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Bromide	<10	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Bromide	<10	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Bromide	<10	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Bromide	<10	UG/ML
300306611	TC2 LEACH 1	IC ANIONS (B134)	Chloride	17 (10)	UG/ML
300306612	TC2 LEACH 2	IC ANIONS (B134)	Chloride	18 (10)	UG/ML
300306613	TC2 LEACH 3	IC ANIONS (B134)	Chloride	17 (10)	UG/ML
300306614	TC2 LEACH 4	IC ANIONS (B134)	Chloride	16 (10)	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Chloride	22 (10)	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Chloride	17 (10)	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Chloride	12 (10)	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Chloride	13 (10)	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Chloride	16 (10)	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Chloride	16 (10)	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Chloride	14 (10)	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Chloride	14 (10)	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Chloride	33 (10)	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Chloride	35 (10)	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Chloride	37 (10)	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Chloride	38 (10)	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Chloride	34 (10)	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Chloride	39 (10)	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Chloride	33 (10)	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Chloride	38 (10)	UG/ML

300306611	TC2 LEACH 1	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306612	TC2 LEACH 2	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306613	TC2 LEACH 3	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306614	TC2 LEACH 4	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Fluoride	<10	UG/ML
300306611	TC2 LEACH 1	IC ANIONS (B134)	Formate	<10	UG/ML
300306612	TC2 LEACH 2	IC ANIONS (B134)	Formate	<10	UG/ML
300306613	TC2 LEACH 3	IC ANIONS (B134)	Formate	<10	UG/ML
300306614	TC2 LEACH 4	IC ANIONS (B134)	Formate	<10	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Formate	<10	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Formate	<10	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Formate	<10	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Formate	<10	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Formate	<10	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Formate	<10	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Formate	<10	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Formate	<10	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Formate	<10	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Formate	<10	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Formate	<10	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Formate	<10	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Formate	<10	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Formate	<10	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Formate	<10	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Formate	<10	UG/ML
300306611	TC2 LEACH 1	IC ANIONS (B134)	Nitrate	682 (10)	UG/ML

300306612	TC2 LEACH 2	IC ANIONS (B134)	Nitrate	908 (10)	UG/ML
300306613	TC2 LEACH 3	IC ANIONS (B134)	Nitrate	913 (10)	UG/ML
300306614	TC2 LEACH 4	IC ANIONS (B134)	Nitrate	867 (10)	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Nitrate	1310 (10)	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Nitrate	1030 (10)	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Nitrate	766 (10)	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Nitrate	799 (10)	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Nitrate	990 (10)	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Nitrate	1007 (10)	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Nitrate	889 (10)	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Nitrate	854 (10)	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Nitrate	2414 (10)	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Nitrate	2571 (10)	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Nitrate	2715 (10)	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Nitrate	2777 (10)	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Nitrate	2503 (10)	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Nitrate	2926 (10)	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Nitrate	2412 (10)	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Nitrate	2691 (10)	UG/ML
300306611	TC2 LEACH 1	IC ANIONS (B134)	Nitrite	186 (10)	UG/ML
300306612	TC2 LEACH 2	IC ANIONS (B134)	Nitrite	234 (10)	UG/ML
300306613	TC2 LEACH 3	IC ANIONS (B134)	Nitrite	231 (10)	UG/ML
300306614	TC2 LEACH 4	IC ANIONS (B134)	Nitrite	212 (10)	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Nitrite	330 (10)	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Nitrite	261 (10)	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Nitrite	174 (10)	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Nitrite	202 (10)	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Nitrite	248 (10)	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Nitrite	251 (10)	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Nitrite	219 (10)	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Nitrite	211 (10)	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Nitrite	589 (10)	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Nitrite	625 (10)	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Nitrite	658 (10)	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Nitrite	678 (10)	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Nitrite	604 (10)	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Nitrite	709 (10)	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Nitrite	579 (10)	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Nitrite	649 (10)	UG/ML
300306611	TC2 LEACH 1	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306612	TC2 LEACH 2	IC ANIONS (B134)	Oxalate	<10	UG/ML

300306613	TC2 LEACH 3	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306614	TC2 LEACH 4	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Oxalate	<10	UG/ML
300306611	TC2 LEACH 1	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306612	TC2 LEACH 2	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306613	TC2 LEACH 3	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306614	TC2 LEACH 4	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Phosphate	<10	UG/ML
300306611	TC2 LEACH 1	IC ANIONS (B134)	Sulfate	323 (10)	UG/ML
300306612	TC2 LEACH 2	IC ANIONS (B134)	Sulfate	387 (10)	UG/ML
300306613	TC2 LEACH 3	IC ANIONS (B134)	Sulfate	357 (10)	UG/ML

300306614	TC2 LEACH 4	IC ANIONS (B134)	Sulfate	332 (10)	UG/ML
300306615	TC2 LEACH 5	IC ANIONS (B134)	Sulfate	483 (10)	UG/ML
300306616	TC2 LEACH 6	IC ANIONS (B134)	Sulfate	371 (10)	UG/ML
300306617	TC2 LEACH 7	IC ANIONS (B134)	Sulfate	284 (10)	UG/ML
300306618	TC2 LEACH 8	IC ANIONS (B134)	Sulfate	297 (10)	UG/ML
300306619	TC2 LEACH 9	IC ANIONS (B134)	Sulfate	369 (10)	UG/ML
300306620	TC2 LEACH 10	IC ANIONS (B134)	Sulfate	373 (10)	UG/ML
300306621	TC2 LEACH 11	IC ANIONS (B134)	Sulfate	329 (10)	UG/ML
300306622	TC2 LEACH 12	IC ANIONS (B134)	Sulfate	319 (10)	UG/ML
300306623	TC2 LEACH 13	IC ANIONS (B134)	Sulfate	890 (10)	UG/ML
300306624	TC2 LEACH 14	IC ANIONS (B134)	Sulfate	957 (10)	UG/ML
300306625	TC2 LEACH 15	IC ANIONS (B134)	Sulfate	1007 (10)	UG/ML
300306626	TC2 LEACH 16	IC ANIONS (B134)	Sulfate	1022 (10)	UG/ML
300306627	TC2 LEACH 17	IC ANIONS (B134)	Sulfate	920 (10)	UG/ML
300306628	TC2 LEACH 18	IC ANIONS (B134)	Sulfate	1066 (10)	UG/ML
300306629	TC2 LEACH 19	IC ANIONS (B134)	Sulfate	886 (10)	UG/ML
300306630	TC2 LEACH 20	IC ANIONS (B134)	Sulfate	986 (10)	UG/ML

**Distribution:**

P. M. Almond, 773-43A  
D. R. Best, 999-W  
T. B. Brown, 773-A  
D. R. Click, 999-W  
A. D. Cozzi, 999-W  
D. A. Crowley, 773-43A  
R. E. Eibling, 999-W  
S. D. Fink, 773-A  
G. P. Flach, 773-42A  
K. M. Fox, 999-W  
E. K. Hansen, 999-W  
C. C. Herman, 773-A  
E. N. Hoffman, 999-W  
V. Jain, 766-H  
C. A. Langton, 773-43A  
S. L. Marra, 773-A  
F. M. Pennebaker, 773-42A  
M. M. Reigel, 999-W  
W. T. Riley, 999-W  
K. A. Roberts, 773-43A  
S. P. Simner, 249-8H  
S. A. Smith, 705-1C  
A. V. Staub, 704-Z  
K. H. Subramanian, 241-156H  
W. R. Wilmarth, 773-A  
Records Administration (EDWS)

J. A. Diediker, DOE-ORP  
T. W. Fletcher, DOE-ORP  
B. J. Harp, DOE-ORP  
C. C. Harrington, DOE-ORP  
S. Pfaff, DOE-ORP  
A. V. Arakali, WTP  
S. M. Barnes, WTP  
P. A. Benson, WTP  
G. M. Duncan, WTP  
S. T. Arm, WRPS  
C. Burrows, WRPS  
P. Cavanah, WRPS  
T. W. Crawford, WRPS  
W. G. Ramsey, WRPS  
R. A. Robbins, WRPS  
P. L. Rutland, WRPS  
S. A. Saunders, WRPS  
D. J. Swanberg, WRPS  
M. G. Thien, WRPS  
C.A. Brown, PNNL  
R. D. Peterson, PNNL  
R. J. Serne, PNNL  
J. H. Westsik, PNNL