SRNL-STI-2012-00558 Revision 0

Process Formulations and Curing Conditions that Affect Saltstone Properties

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September 2012

Savannah River National Laboratory Savannah River Nuclear Solutions, LLC Aiken, SC 29808

Prepared for the U.S. Department of Energy under contract number DE-AC09-08SR22470.



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Printed in the United States of America

Prepared for U.S. Department of Energy

Keywords: Saltstone Performance Assessment Hydraulic Conductivity Water to Premix Ratio

Retention: Permanent

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9-28-12 Date

<u>9/28/12</u> Date

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9-28-12 Date

9/28/12 Date

EXECUTIVE SUMMARY

The Saltstone Production Facility (SPF) receives low level waste (LLW) salt solution from Tank 50H for treatment and disposal. Although Tank 50 receives transfers from multiple processes at the Savannah River Site (SRS), the primary influent is from the Actinide Removal Process/Modular Caustic Side Solvent Extraction Unit (ARP/MCU). At the SPF, the LLW is mixed with premix (a cementitious mixture of portland cement, blast furnace slag and Class F fly ash) in a Readco continuous mixer to produce fresh (uncured) saltstone that is transferred to the Saltstone Disposal Facility (SDF) for permanent disposition in the Saltstone Disposal Units (SDU) (previously referred to a vaults). Admixtures, if needed, are also added to the formulation to ensure the grout can be processed through the facility but not adversely affect the curing of saltstone in the SDU. Throughout processing operations, the mixer, grout hopper, grout pump, and piping to the SDU are flushed with water every 15 minutes to mitigate process upsets. However, this extra water (which also includes drain water, and sources other than the Tank 50 salt solution) is not accounted for in the water to premix (w/p) calculation for the saltstone formulation. Analysis of a continuous day of processing shows that during a routine operational flush, an instantaneous w/p ratio of 0.73 is being made. During nominal steady-state processing, the w/p ratio target is 0.59 or 0.60. Data from a typical (no setbacks or process upsets) processing day at the SPF were analyzed to calculate instantaneous and cumulative saltstone water to premix ratios over the given period of processing. A series of saltstone formulations were developed and analyzed based on these data. The fresh properties (set, bleed, flowability, viscosity, yield stress, and gel) and cured properties (porosity, density, compressive strength, hydraulic conductivity, and contaminant retention) of these formulations were analyzed.

The first objective of this study was to analyze saltstone fresh properties to determine the feasibility of reducing the formulation w/p ratio while varying the amount of extra water and admixtures used during processing at the SPF. The 8 wt % extra water in the formulation causes saltstone to generate more bleed water as it cures than the samples with 4 wt % extra water or salt solution since the water dilutes the salt solution molarity which delays the reaction of the slag and fly ash. Adding extra water to the formulations with w/p ratios of 0.58 or lower does not increase the set time past three days and the bleed water is reabsorbed after three days. Therefore, only considering the properties of bleed water and set time, it is feasible to process w/p ratios below 0.58 by adding extra water or admix. Adding Daratard to w/p ratios greater than 0.66 causes long set times and in some cases, standing bleed water after three days; therefore its use should be limited or avoided at these formulations.

However, the gel time is affected by the extra water, even at the low w/p ratios. Saltstone formulations with low w/p ratios (0.56 or lower) without any admixture or extra water, gel too quickly (< 20 min) to ensure reliable processing through the facility; however, if admix is used in these formulations, the gel time is extended into the acceptable range of 20 - 60 minutes. Adding 8 wt % extra water to the low w/p formulations increases the gel time past 60 minutes. Therefore, for gel time it may be feasible to use Daratard to process low w/p ratios as long as the dosage is very well controlled and can remain at 0.05 wt %. However, using 8 wt % extra water extends the gel time past the acceptable limits for these formulations. The vane rheology method for determining the gel time of a sample provides insight into the structure development of the sample; however it is difficult to distinguish the gel time from settling or other reactions based on the results obtained thus far. If this method is further developed, it could be less subjective than the pour test and provide more understanding into the hydration and structure development of fresh saltstone.

Based on the fresh property data presented in this report, mixes with w/p ratios in the range of 0.57 - 0.66 formulated with 4 wt % extra water or salt solution only, produce fresh saltstone grout that satisfies acceptance criteria for set time, bleed, gel, and rheology. All of the formulations with 8 wt % extra water

or formulations with w/p ratios above 0.65 that include Daratard, have fresh properties outside the acceptance criteria.

The second part of this study was to provide information for understanding the impact of curing conditions (cure temperature, relative humidity (RH)) and processing formulation on the performance properties of cured saltstone. Samples were cured under profiles based on temperature data from cell K and cell F in SDU 4. Half of the samples were kept under saturated conditions by adding liquid on top of the grout to maintain complete saturation. The other half had the grout surface exposed to the humid environment. Based on the results of this study, the relative humidity of the curing environment has the most impact on the performance (cured) properties of saltstone. If the drying of the sample is eliminated or at least minimized, higher cure temperatures can be beneficial for saltstone due to increased reaction rates, formation of denser hydration products, and reduction in porosity.

The final w/p ratio of the formulation does affect the density, porosity, and compressive strength of the cured saltstone. The saltstone formulations with 0.59 w/p ratios and lower had higher densities, lower porosities, and higher compressive strength than samples formulated at a higher w/p ratio. There is a general trend that the samples formulated at high w/p ratios (0.67 and higher) have the lowest density, highest porosity and lowest strength. However, compared to data from previous studies, the density, porosity, and compressive strength of saltstone are more controlled by curing in a high humidity environment rather than the w/p ratio of the saltstone formulation. The Leachability index for Na, NO₃⁻ and NO₂⁻ was calculated for the cured samples using the ANS/ANSI 16.1 standard. Based on the formulations tested in this study, the initial formulation of the samples did not have an effect on the rate at which contaminants are released from the saltstone.

Samples cured for 28 days in controlled humidity environments under temperature profiles up to 55 and 80°C had hydraulic conductivities of 4.5E-8 to 4.3E-10 cm/s. On average, the cell K samples had slightly higher hydraulic conductivities for samples at higher w/p ratios than the cell F samples; however, the majority of the samples have hydraulic conductivities within the range of E-9 or E-10 cm/s. Of the 72 samples analyzed, only three samples had hydraulic conductivities on the order of 2.8E-7 cm/s. In a previous study, samples cured isothermally in an uncontrolled environment had hydraulic conductivities ranging from 1.04E-9 to 1.90E-6 cm/s, with the majority of the 60 °C samples having hydraulic conductivities greater than 1.0E-7 cm/s. Comparing the results presented in this study to the results from the previous study, the curing conditions have a greater effect on the hydraulic conductivity of saltstone than the w/p ratio. The saltstone in the current study was cured in a high humidity environment which allowed the microstructure to develop without being dehydrated, reducing the formation of microcracks. Therefore, the combination of curing saltstone in a saturated or high humidity environment and under a ramped temperature profile results in slower hydraulic conductivities than saltstone dried out during curing and cured at high isothermal temperatures.

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LIST OF ABBREVIATIONS

ANS	American Nuclear Society
ANSI	American National Standards Institute
ARP	Actinide Removal Process
ASTM	American Society for Testing and Materials
C&WDA	Closure and Waste Disposal Authority
gpm	Gallons per minute
НОН	Heat of Hydration
IC	Ion Chromatography
ICP-ES	Inductively Coupled Plasma Emission Spectroscopy
LLW	Low Level Waste
MCU	Modular Caustic Side Solvent Extraction Unit
mL	milliliters
PA	Performance Assessment
Psi	Pounds per square inch
RH	Relative Humidity
S	second
SDF	Saltstone Disposal Facility
SDU	Saltstone Disposal Unit
SPF	Saltstone Production Facility
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
TTQAP	Task Technical and Quality Assurance Plan
TTR	Technical Task Request
w/p	Water to premix
WSE	Waste Solidification Engineering
wt %	Weight percent

1.0 Introduction and Background

The Saltstone Facility at the Savannah River Site (SRS) is comprised of two facilities, the Saltstone Production Facility (SPF) and the Saltstone Disposal Facility (SDF). The SPF receives low level waste (LLW) salt solution from Tank 50H for treatment. Although Tank 50 receives transfers from multiple processes at SRS, the primary influent is from the Actinide Removal Process/Modular Caustic Side Solvent Extraction Unit (ARP/MCU). At the SPF, the LLW is mixed with premix (a cementitious mixture of portland cement, blast furnace slag and Class F fly ash) in a Readco continuous mixer to produce fresh (uncured) saltstone that is transferred to the Saltstone Disposal Facility (SDF) for permanent disposition in a Saltstone Disposal Unit (SDU) (previously referred to as a vault). Admixtures, if needed,¹⁻³ are also added to the formulation to ensure the grout can be processed through the facility but not adversely affect the curing of saltstone in the SDU. Figure 1-1 shows a simplified process flow diagram for the entire Saltstone Facility.



Figure 1-1. Simplified Saltstone Facility process flow diagram

Throughout processing operations, the mixer, grout hopper, grout pump, and piping to the SDU are flushed with water every 15 minutes to mitigate process upsets. However, this extra water is not accounted for in the water to premix (w/p) calculation for the saltstone formulation. Analysis of a continuous day of processing shows that during a routine operational flush, an instantaneous w/p ratio of 0.73 is being made.^{*} During nominal steady-state processing, the w/p ratio target is 0.59 or 0.60. Previous testing has shown that saltstone formulated at a higher w/p ratio has longer gel and set times, more bleed water generated, and generally poor performance properties compared to saltstone made at w/p ratios of 0.6 or lower.⁴⁻⁸ The yield stress and plastic viscosity of fresh saltstone, properties which are indicative of the flowability and thickness of the slurry, are dependent on the final w/p ratio of the formulation.⁹

^{*} Using PI ProcessBook and PI DataLink to analyze instantaneous and cumulative w/p ratios throughout processing on Nov. 23, 2011.

Adding Daratard or extra water or both to the initial saltstone formulation to increase the w/p ratio lowers the yield stress and plastic viscosity of that mixture. Saltstone formulations with extra water or admixtures often have longer gel times than samples at lower w/p ratios.²

Admixtures, specifically Daratard-17, a set retarder and long range water reducer, have a significant impact on the fresh and cured properties of saltstone. Historically, Daratard-17 has been added to a saltstone formulation to extend the gel time past 20 minutes, improve flowability, and to optimize processing.¹⁰⁻¹² Recent studies have shown that it causes a significant amount of drain water to be returned from the SDU, especially at higher w/p ratios¹³ which dilutes the Tank 50 material held in the Salt Feed Tank (SFT) prior to being processed (Figure 1-1).¹⁴ In addition, the extra drain water causes an increased amount of cementitious solids¹⁵ to pass through the sheet drain that lines the inside of the SDU and plug the drain water return lines.¹³ This buildup of cementious material causes failure of the drain water return system and associated equipment, and causes delays in processing material from Tank 50H.¹⁶ Therefore, the feasibility of altering the flowsheet to reduce the w/p ratio while varying the Daratard concentration and maintaining the current operational strategy (flushing sequence) is investigated in this report as requested in the Technical Task Request (TTR) from Waste Solidification Engineering (WSE).¹⁶ As described in the Task Technical and Quality Assurance Plan (TTQAP), the fresh properties (density, rheology, flowability, etc.) of these grout mixes are evaluated to facilitate development of acceptance criteria for saltstone to ensure processability through the SPF.¹⁷

Static gel time has been used to estimate the amount of time available for recovery in the event of a process upset. It is an indication of the maximum amount of time the pumps could be inoperable before the slurry developed enough structure to prevent fluid response to pressure.¹¹ Static gel time has also been used along with operating experience to estimate saltstone flowability in the SDUs. The shorter the gel time, the less flow achieved in the SDUs.¹¹ The traditional pour test provides a gel time of the fresh grout; however, the rate the sample is poured, disturbing the sample prior to analysis, and the judgment of the researcher all factor into the final determination of the gel time. Replacing the pour test with a less subjective measurement such as monitoring the development of structure in the saltstone by measuring shear stress using vane rheology would be a more definitive method that could be related to processing of grout through the SPF.

In a separate TTR, Closure and Waste Disposal Authority (C&WDA) requested the cured properties of the saltstone formulations requested by WSE be analyzed to further define the SPF/SDF operating conditions required to meet the materials performance properties used in the SDF Performance Assessment (PA).¹⁸ As a continuation of previous work,⁸ samples were cured under a temperature profile rather than cured isothermally. As described in the TTQAP, historical data from SDU 4 were used as a basis for the temperature profile; however it is important to note that temperatures recorded in previous disposal units vary due to many factors including processing strategy and saltstone formulation. The temperatures that will be recorded in future SDUs will vary from those in SDU 4 due to pour strategy, formulation, and other factors. The intention of this study was to trend saltstone performance properties as a function of curing conditions (relative humidity, temperature) rather than provide information on the performance of material residing in SDU 4 or previous disposal units. Since field conditions vary in the disposal units depending on pour strategy, etc., two curing conditions were investigated. The first was curing saltstone under a defined temperature profile with the surface exposed to a humid environment. The second was saltstone cured under the sample temperature profile while keeping the surface covered with liquid to maintain saturated conditions as described in the PA.¹⁹ The cured properties of these samples are compared to results from a previous study⁸ where samples were cured under the same temperature profile in an environment with uncontrolled humidity.

The objective of this report is twofold: first, analyze saltstone fresh properties to determine the feasibility of reducing the formulation w/p ratio while varying the amount of extra water and admixtures used during

processing; and second, provide information for understanding the impact of curing conditions (cure temperature, relative humidity (RH)) and processing formulation (w/p, extra water, admixtures) on the performance properties (hydraulic conductivity, porosity, density, contaminant retention) of cured saltstone.

2.0 Experimental Procedure

Data from a typical (no setbacks or process upsets) processing day at the SPF were analyzed to determine the average amount of extra water added to the system (Appendix A). Extra water is defined for this work as the water from flushes, drain water, and sources other than the Tank 50 salt solution. The data were used to calculate instantaneous and cumulative w/p ratios over the given period of processing. Both points were tested to provide snapshots of actual conditions at a given time in the facility.

The w/p ratio, amount of extra water, and admixture dosage are the formulation variables. The premix composition and the salt solution chemistry were held constant. The test matrix is listed in Table 2-1. Cured properties were measured on selected samples that encompass the ranges of w/p ratios investigated in this study. The starting w/p was selected by assuming that only the LLW salt solution is being processed with premix to make saltstone slurry. The baseline formulation 0.59 w/p ratio with no admix or extra water (WP001) since this is how the SPF has been processing since the third quarter of 2011. The extra water added is the total calculated amount of water added during a full day of processing (8 wt % of the LLW processed) or half of that amount (4 wt %). The final w/p ratio is calculated after the extra water is added to the formulation. The amount of admix added is an average of admix dosage to the saltstone formulations from 2008 – 2010 (approximately 0.08 gpm in the SPF).^{20,21} The formulations selected for cured property measurements are representative of the range of starting and final w/p ratios analyzed in this report.

Samula	Starting	Extra Water	Final	Admix	Measure Cured
Sample	w/p	(wt %)	w/p	(wt %)	Properties?
WP001	0.59	0	0.59	0.00	Y
WP002	0.56	0	0.56	0.00	Ν
WP003	0.67	0	0.67	0.00	Y
WP004	0.59	0	0.59	0.05	Ν
WP005	0.56	8	0.70	0.05	Y
WP006	0.52	0	0.52	0.00	Ν
WP007	0.53	0	0.53	0.05	Ν
WP008	0.55	4	0.62	0.00	Ν
WP009	0.69	0	0.69	0.05	Ν
WP010	0.58	8	0.72	0.05	Y
WP011	0.57	4	0.64	0.00	Y
WP012	0.54	0	0.54	0.05	Ν
WP013	0.54	8	0.68	0.00	Ν
WP014	0.55	0	0.55	0.00	Y
WP015	0.61	0	0.61	0.00	Ν
WP016	0.58	0	0.58	0.05	Ν
WP017	0.57	0	0.57	0.05	Ν
WP018	0.73	0	0.73	0.00	Y
WP019	0.71	0	0.71	0.05	Ν
WP020	0.63	0	0.63	0.00	Ν
WP021	0.53	4	0.60	0.05	Y
WP022	0.65	0	0.65	0.05	Ν
WP023	0.51	0	0.51	0.00	Y
WP024	0.59	4	0.66	0.05	N
WP025	0.59	8	0.74	0.00	Ν

 Table 2-1. Experimental design for saltstone mixes tested

2.1 Salt Solution Simulant

The mid-aluminate molarity salt solution (Table 2-2) is used in this task since its composition is comparable to what is currently being processed from Tank 50H and cured property results from previous studies can be compared to results from this report.^{8,22} The salt solution was spiked with chromium (300 mg/L) to facilitate contaminant retention testing of cured samples.

	Mid Aluminate				
Compound	Molarity [mol/L]	Molecular Weight [g/mol]			
50% by Weight NaOH	2.460	40.00			
NaNO ₃	1.865	84.99			
NaNO ₂	0.550	68.99			
Na ₂ CO ₃	0.176	105.99			
Na ₂ SO ₄	0.059	142.04			
Aluminum Nitrate (9 H ₂ O)	0.165	375.13			
Sodium Phosphate (12 H ₂ O)	0.012	380.12			
Total Salt mass (g)	359	9.06			
Total sodium Molarity	Total sodium Molarity5.38				

Table 2-2. Composition of mid-aluminate ARP/MCU salt solution simulant

2.2 Sample Preparation

The nominal premix distribution used in this testing is 45 wt % slag, 45 wt % thermally beneficiated Class F fly ash, and 10 wt % portland cement. The premix materials (Table 2-3) were received in five gallon containers from the vendors during delivery of the bulk materials to the SPF. The premix materials were stored such that the exposure to humid air and hydration was limited prior to use.

Material	Category	Vendor
Portland Cement	ASTM C 150/ C 150M-11	Holcim
Blast Furnace Slag	ASTM C 989-10	Holcim
Fly Ash	ASTM C 618-08a	SEFA

Table 2-3. Saltstone premix materials

The salt solution, premix, and admix materials were mixed for approximately three minutes using a paddle blade mixer. The mixing was paused for approximately five seconds after 30 seconds of mixing to allow entrained air to escape from the grout. All mixes included ACP-3183 (previously called Q2-3183A), an antifoam admixture, at a dose of 0.03 wt %, since it is included during daily processing at the SPF. Daratard-17 was added to selected mixes (Table 2-1) at the specified dosage of 0.05 wt %. All admixture dosages are on a wt % basis of the premix in the formulation. The admixtures were added to the salt solution first and then the dry feeds were added. After mixing was complete, the fresh grout was poured into sample molds and cups for property testing. Table 2-4 lists the approximate sample size for each analysis performed in this study.

Property or Analysis Method	Sample Container	Approximate Sample Mass
Bleed water	1 x 4 inch vials	75 grams
Set	1 x 4 inch vials	50 grams
Fresh density	1 x 4 inch vials	15 grams
Gel time	1 x 4 inch vials	50 grams
Rheology Flow Curve	Rheology cup	100 grams
Vane rheology	Rheology cup	300 grams
Flow Cone	2 x 4 inch cylinder	150 grams
Heat of Hydration	15 mL vial	7 grams
Compression	3 x 6 inch cylinder	1200 grams
Bulk Density	3 x 6 inch cylinder	1200 grams
Hydraulic Conductivity	3 x 6 inch cylinder	1200 grams
Leachability Index	1 x 4 inch vials	130 grams
Porosity	Not applicable	100 grams

Table 2	2-4.	Sample	size ar	nd c	ontainers	for	specific	property	analyses
		~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~					- Person	Property.	

# 2.3 Fresh Property Measurements

Fresh properties were measured after the grout had mixed for the designated three minutes. These properties include: bleed water, set time, fresh density, gel time, yield stress, plastic viscosity, flowability, and heat of hydration. The standing (bleed) water was measured on duplicate samples of fresh grout placed in sealed cylinders and left undisturbed for 24 hours. A Vicat needle²³ was used to determine the final set time of each mix. The fresh density of each mix was determined by pouring the fresh mixture into a density cup with a volume of 8.38 milliliters (mL). The density is determined by dividing the sample mass by the volume of the cup.

Gel Time was determined by two methods: 1) Vane measurement using the Haake rotoviscometer equipped with a four-blade vane which was rotated at one revolution per hour⁹ and 2) the pour test. There have been multiple methods for interpreting the stress versus time curve as a result of vane measurements. Previous tests have used the change in slope as the gel time; however, with this method it can be difficult to distinguish between settling and gelling.¹¹ Since gelling is a continuous process, the time of the first indication of increased resistance or the change of the slope of the curve will be used as the gel time.¹¹ The pour test consists of letting the grout sit undisturbed and pouring a sample every five or ten minutes from a cylinder into an empty container. This pouring is repeated until the fresh grout had developed sufficient structure so it does not flow as a result of its own mass. The time at which the grout does not flow from the cylinder is designated as the gel time.

Rheological flow curves were obtained using a Haake VT550 rotoviscometer equipped with a stationary sample cup and a rotating MVII bob. This instrument has a smooth wall coaxial cylindrical geometry.^{9,11} The flow between the two concentric cylinders is characterized by measuring the torque and speed of the inner cylinder. The torque readings were converted to shear stress and the speed to shear rate. Flow curves (up and down) were generated over a shear rate range of 0 to 300 sec⁻¹. Each curve took 150 seconds to accelerate/decelerate. After accelerating to 300 sec⁻¹, the shear rate was held for 30 seconds prior to decelerating. Based on the shapes of the down curves, a Bingham Plastic rheological model was used for regression of the data to calculate the plastic viscosity and yield stress of the fresh grout. Equations used to calculate the plastic viscosity and yield stress using the flow curve data are presented elsewhere.⁹ The plastic viscosities and yield stresses were calculated from the data on the decreasing shear rate (down curve) of the flow curves to minimize the impact of any thixotropic behavior of the saltstone on the measurements.

A modified slump test was used to measure the flowability of the fresh saltstone due to gravity.²⁴ Immediately after mixing, the slurry is poured into an open cylinder with one end sitting on a smooth, flat surface. Approximately 30 seconds after the cylinder was filled, it was lifted vertically approximately one inch off the surface to let the grout flow out and spread due to gravity. The diameter of the flowed grout was measured.

The heat of hydration (HOH) for the mixes was determined using an isothermal calorimeter. Mixes with varying extra water amounts and admixture dosages were tested to determine the effect on the heat generated during curing. The isothermal calorimeter was kept at 25 °C and tests continued for at least three days after mixing.

#### 2.4 Sample Curing

Samples were cured under profiles based on temperature data from cell K and cell F in SDU 4. To date, the temperatures in cell K are the highest recorded in any of the SDUs and the temperature profile of cell F is typical of temperatures recorded in multiple cells in SDU 4. The data used are actual temperature data recorded by thermocouples in the cells[†] over several months and approximated to a 28 day cure time. Since the processing conditions cannot be replicated and the cure time for simulated saltstone is significantly decreased compared to actual saltstone in cells K and F, the cure profiles in Figure 2-1 are only meant to approximate two different processing conditions (pour strategy, etc.) in the SPF. The actual thermocouple data as recorded in the cells K and F are provided in Appendix A. A programmable convection oven was used to achieve the curing profiles (Figure 2-1). At the end of the curing profile, the oven temperature was ramped down to room temperature over several days while keeping the humidity high to prevent any drying and cracking of the samples during cooling. Once removed from the oven, samples were kept in a plastic bag with a wet sponge to maintain humid conditions until the cured properties were analyzed.

Modifications to the oven were made to provide a high humidity curing environment while the samples were heated. A control program was used to keep the chamber of the oven at a high relative humidity. Since field conditions vary in the disposal units depending on pour strategy, etc., two curing conditions were investigated. Quadruplicate samples of each formulation were cured in the oven for 28 days. Two of the samples were kept under saturated conditions by adding liquid on top of the grout to maintain complete saturation. The other two samples had the grout surface exposed to the humid environment.

[†] Using PI ProcessBook and PI DataLink to analyze the thermocouple readings in SDU 4, cells F and K.



Figure 2-1. Modified cell F and cell K curing profiles

# 2.5 Cured Property Measurements

Cured properties were measured after samples cured for 28 days. Grout is considered "cured" after 28 days since the major hydration reactions are complete within that time. Although hydration reactions continue indefinitely, the ratio of strength gain compared to cure time is minimal after 28 days. The cured properties measured on these samples are hydraulic conductivity, compression strength, porosity, cured density, and leachability index. After the 28 days, the quadruplicate samples were removed from the oven and one sample from each of the curing conditions was used for hydraulic conductivity analysis. The remaining samples were used for the analyzing the other cured properties.

# 2.5.1 Hydraulic Conductivity

The saturated hydraulic conductivity,²⁵ dry bulk density, and porosity were measured by an offsite laboratory[‡] after the samples had cured for 28 days at SRNL. At the offsite laboratory, each 3x6 inch cylindrical sample was cut in half to create duplicate 3x3 inch samples and labeled as the top and bottom, respectively. The ends of each sample were shaved to ensure parallel surfaces according to the ASTM standard. The saturated hydraulic conductivity of the saltstone samples was determined by ASTM D 5084 method F, the constant-volume falling head using a flexible wall permeameter.²⁵ The fluid used for testing was the simple salt solution that did not include reactive species such as aluminum nitrate or phosphates. This was used to avoid interactions of the test fluid and the sample during testing. Saturated hydraulic conductivity (K) is a function of the porous medium and the properties of the test fluid as described by Darcy's law in Equation (2-1).²⁶

[‡] AMEC E&I (formerly Mactec), 396 Plasters Ave, Atlanta, GA, 30324

$$\frac{dq}{dt} = K \frac{\Delta HA}{L\mu}$$
(2-1)

Where dq/dt is the rate of fluid flow,  $\mu$  is the viscosity of the fluid,  $\Delta H$  is the pressure gradient, A is the surface area, and L is the thickness of the solid. Additional details of the testing and calculations for dry bulk density, porosity and hydraulic conductivity results provided by the offsite laboratory are documented in previous reports.^{6,27,28}

#### 2.5.2 Compressive strength, porosity, cured density

The bulk density of the cured samples was measured immediately after the samples were removed from the oven. The cured density of each sample was calculated by dividing the sample mass by the volume of the sample. The compression strength of the cured samples was measured using a Humboldt CM-3000-LXI compression tester according to ASTM C39 "Compressive strength of Cylindrical Specimens".²⁹ According to the procedure, compression testing of moist-cured specimens shall be made as soon as practical after removal from moist storage.²⁹ Prior to testing, each sample was removed from the plastic cylinder, weighed and the dimensions were measured. Each sample was centered on the lower block and the load was applied to the sample at a constant rate of  $35 \pm 7$  psi/s. The compressive load was applied to each sample until the indicator showed a steady decrease in the load and the sample was visibly cracked.²⁹ The compressive strength was calculated by dividing the maximum load by the cross-sectional area of the sample.

After completion of compressive strength measurements, two fractured pieces from the center of each sample were used for porosity measurements. The porosity was determined by the mass loss³⁰ upon heating samples to 105 °C in a convection oven. The mass loss was monitored until no further mass loss occurred (approximately 24 hours). The water released during heating  $(m_w)$  is converted to a volume of simulant ( $V_{sim}$ ) by dividing by the ratio of the mass of water to the mass of the simulant (0.710 for the mid-point ARP/MCU simulant) then dividing again by the density of the simulant  $(\rho_{sim})^{30}$  as shown in Equation (2-2). The reported porosity (P) of the cured samples is the simulant volume ( $V_{sim}$  [cm³]) divided by the sample volume ( $V_s$  [cm³]) as shown in Equation (2-3).³⁰

$$V_{sim} = \frac{\left(\frac{m_w}{0.710}\right)}{\rho_{sim}} \tag{2-2}$$

$$P = \frac{V_{sim}}{V_s} \tag{2-3}$$

#### 2.5.3 Leachability Index

Before the cured saltstone samples could be leach tested using the American National Standard Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure,³¹ they had to be removed from their sample vials (P/N VL25H from Lavials.com). A miter saw was used to cut the bottom edge off the vial, the sample was turned over, and then the sample was pressed out of the vial using a hand press. After the sample was out of the vial, it was cut to approximately 6 cm using a clean miter saw. The cut dimensions (height and diameter) of the saltstone samples are shown in Table 2-5. The total external surface area (S) of each cut saltstone sample was calculated using Equation (2-4) as follows.

$$S = \pi * D * H + 2 * \pi * \left(\frac{D}{2}\right)^2$$
(2-4)

Where D is the diameter of the cut sample [cm], H is the height of the cut sample [cm], and S is the external surface area of the cut sample [cm²]. The volume of leachant (deionized water) needed for each cut saltstone sample was calculated using Equation (2-5) where  $V_L$  is the volume of leachate [cm³ = mL]. Note that the deionized water was assumed to have a density of 1 g/ml so the volume of leachant calculated in Equation (2-5) was treated as the mass of leachant needed as shown in Table 2-5.

$$V_L = 10 * S$$
 (2-5)

The saltstone samples were cut so the volume of leachant to surface area of the saltstone sample was  $10 \pm 0.2$  cm per ANSI/ANS 16.1-2003.³¹ The samples were cut to approximately 6 cm long (Table 2-5) rather than using the entire 10 cm sample in order to reduce the volume of leachate generated with this test. The cut saltstone samples were suspended in the 1 L bottles with the leachant amounts as shown in Table 2-5. The suspended samples were lowered into each bottle with fresh leachant then lifted out at set times and placed in new bottles with fresh leachant. A blank with just deionized water was created and sampled at each time period to ensure that the leachant was not introducing contaminants into the leachates.³¹ The time periods for the leachate testing were 2, 7, 24, 48, 72, and 96, and 120 hours.³¹ At the end of each elapsed time period, two leachate samples were collected from each bottle. A 125-ml sample was collected for Ion Chromatography (IC) measurements of nitrite (NO₂⁻) and nitrate (NO₃⁻) concentrations and Inductively Coupled Plasma Emission Spectroscopy (ICP-ES) measurements of chromium and sodium concentrations. A 30-60 ml sample was collected from each bottle for pH and conductivity measurements.

Sample	Height (H) [cm]	Diameter (D) [cm]	Side Circumferential Area [cm²]	End Circular Area [cm ² ]	Total External Surface Area of Cut Section (S) [cm ² ]	Volume of Cut Section (V) [cm ³ ]	Leachant Needed for V ₁ /S=10 cm
WP001A	5.93	3.31	61.59	17.16	78.75	101.78	787.5 g
WP001B	5.98	3.29	61.79	16.97	78.76	101.56	787.6 g
WP003A	5.93	3.31	61.71	17.25	78.96	102.25	789.6 g
WP003B	5.88	3.30	60.98	17.11	78.09	100.62	780.9 g
WP005A	5.98	3.39	63.69	18.03	81.72	107.89	817.2 g
WP005B	5.87	3.30	60.76	17.06	77.83	100.13	778.3 g
WP010A	5.88	3.31	61.21	17.23	78.44	101.37	784.4 g
WP010B	5.84	3.30	60.52	17.11	77.63	99.86	776.3 g
WP011A	5.84	3.30	60.47	17.05	77.53	99.63	775.3 g
WP011B	5.86	3.29	60.66	17.03	77.70	99.88	777.0 g
WP014A	5.89	3.28	60.67	16.91	77.58	99.53	775.8 g
WP014B	5.87	3.29	60.61	16.99	77.60	99.68	776.0 g
WP018A	5.88	3.29	60.81	17.00	77.81	100.03	778.1 g
WP018B	5.79	3.29	59.81	16.98	76.79	98.32	767.9 g
WP021A	5.97	3.32	62.15	17.26	79.41	103.02	794.1 g
WP021B	5.98	3.30	61.92	17.06	78.99	102.05	789.9 g
WP023A	5.79	3.28	59.69	16.91	76.60	97.92	766.0 g
WP023B	5.82	3.32	60.80	17.35	78.14	101.02	781.4 g

Table 2-5. Cut Saltstone sample dimensions and calculations for surface area and leachant volume

# **3.0 Results and Discussion**

## 3.1 Fresh Properties

The fresh properties of the grout mixes are important to ensure the product can be processed through the facility and transferred to the SDF SDU. In the SDU, the grout should be formulated such that it sets within three days and produces less than three volume percent of bleed water. The gel time of the fresh grout should be between 20 and 60 minutes.^{11,32} A gel time of less than 20 minutes limits the workability of the grout during a process upset while it is in the facility and a gel time of longer than 60 minutes can lead to settling and segregation of the grout. The specified gel range is to ensure processability through the facility and to ensure that sufficient microstructure develops once the grout is placed in the SDU. Development of the structure over a short time period helps prevent segregation of the grout components (liquid from solids). Bleed water on top of the set grout is an indication that segregation is occurring.²⁶

## 3.1.1 Fresh Density, Heat of Hydration, Flowability, Bleed and Set Time

The fresh density, flowability, set time, bleed, and heat of hydration data for all mixes are listed in Table 3-1.

Gammala	w/p ı	ratio	Fresh Density	Flowability	Set Time	Bleed (vol %)		НОН
Sample	Start	Finish	(g/mL)	(cm)	(days)	Day 1	Day 3	( <b>J</b> /g)
WP001	0.59	0.59	1.728	23.02	1	wet surface		162.19
WP002	0.56	0.56	1.730	21.75	1	dry si	ırface	175.64
WP003	0.67	0.67	1.678	22.38	3	0.41	0.00	149.09
WP004	0.59	0.59 ^a	1.730	21.35	1	1.49	0.00	153.84
WP005	0.56	$0.7^{a,b}$	1.651	25.00	2	0.73	0.00	142.65
WP006	0.52	0.52	1.802	18.73	1	1.14	0.00	151.24
WP007	0.53	0.53 ^a	1.784	20.56	2	1.42	0.00	162.36
WP008	0.55	0.62 ^c	1.698	22.07	3	1.21	0.00	165.58
WP009	0.69	0.69 ^a	1.680	25.72	3	2.91	1.57	141.36
WP010	0.58	0.72 ^{a,b}	1.639	27.31	2	1.19	0.00	148.58
WP011	0.57	0.64 ^c	1.684	24.37	3	1.66	0.79	134.56
WP012	0.54	0.54 ^a	1.751	21.59	2	wet s	urface	136.67
WP013	0.54	0.68 ^b	1.658	24.92	2	1.72	0.78	145.14
WP014	0.55	0.55	1.742	21.43	1	0.33	0.00	172.32
WP015	0.61	0.61	1.701	24.69	1	1.02	0.00	n/a
WP016	0.58	0.58 ^a	1.723	22.38	1	wet s	urface	140.98
WP017	0.57	0.57 ^a	1.730	22.23	2	dry si	urface	159.00
WP018	0.73	0.73	1.653	28.10	4	1.99	1.84	151.28
WP019	0.71	0.71 ^a	1.693	28.26	2	1.16	0.00	161.27
WP020	0.63	0.63	1.710	24.45	2	0.80	0.00	160.72
WP021	0.53	0.6 ^{a,c}	1.707	21.35	2	0.28	0.00	133.89
WP022	0.65	0.65 ^a	1.691	25.80	2	0.22	0.00	158.00
WP023	0.51	0.51	1.794	18.57	1	dry surface		132.92
WP024	0.59	0.66 ^{a,c}	1.668	26.51	3	1.04	0.00	157.07
WP025	0.59	0.74 ^b	1.637	26.59	3	1.77	1.46	169.85

Table 3-1. Fresh density, flowability, set, bleed, and heat of hydration for all mixes

^a Samples with admix included in the formulation

^b Formulation with 8 wt % extra water

^c Formulation with 4 wt % extra water

As shown in Table 3-1, all mixes were set within three days except for mix WP018 which set in four days. This is expected since this mix has a w/p of 0.73, which is the highest of all the mixes tested using only salt solution. In general, the mixes with Daratard had longer set times. This is expected since this admixture is designed to be a set retarder. The extra water did have an effect on the set time of the samples and made the material flow out from the cylinder in a larger diameter. The set time of samples with extra water was extended since fly ash and slag are activated by alkali solutions and have very slow reactions with water.³³⁻³⁷ Therefore, diluting the salt solution with water will delay the hydration reactions of the slag and fly ash, leading to longer set times. The delayed reaction is indicated by the lower HOH values for mixes with extra water or admix compared to mixes without any additions.

Mixes that are more fluid are also less dense and generally had a higher volume percentage of bleed than the mixes that were denser. The data show that admixtures had no effect on the bleed water of a mix except for mix WP009, which had the most bleed after one day of all the samples. Mixes with starting w/p ratios of 0.59 or lower with no extra water or admixes had little (wet surface, no measurable bleed) or no bleed water (dry surface). Samples that were formulated low starting w/p ratios (below 0.59) plus 4% extra water did generate bleed after one day; however, all the bleed was reabsorbed after three days. The samples that were formulated with high starting w/p ratios or 8 % extra bleed water had excess (standing) bleed water after three days.

In summary, the 8 wt % extra water in the formulation causes the saltstone to generate more bleed water as it cures than the samples with 4 wt % extra water or salt solution only. Limiting the amount of extra water in the SPF would limit the amount of settling and excess bleed water of the saltstone in the SDUs. Adding Daratard to w/p ratios greater than 0.66 causes longer set times and in some cases, standing bleed water after three days; therefore its use should be limited or avoided. Adding extra water to the formulations with w/p ratios of 0.58 or lower does not increase the set time past three days and the bleed water is reabsorbed after three days (with the exception of WP011). Therefore, only considering the properties of bleed water and set time, it is feasible to process w/p ratios below 0.58 by adding extra water or admix.

## 3.1.2 Gel Time

As shown in Table 3-2 as well as the figures in Appendix B, extra water or admixtures in the saltstone formulation delays the gel time of the sample. Gel times from 20 - 60 minutes are acceptable as long as there is no settling occurring.^{11,32} Long gel times are beneficial for the SPF to ensure processability through the facility; however, longer gel times often lead to settling and higher bleed water generation in the SDF SDUs. The vane rheology method for determining the gel time of a sample provides insight into the structure development of the sample; however, it is difficult to distinguish the gel time from settling or other reactions based on the results obtained thus far. If this method is further developed, it could be less subjective than the pour test and provide more understanding into the hydration and structure development of fresh saltstone.

Based on the results presented in Table 3-2, saltstone formulations with low w/p ratios (0.56 or lower) without any admixture or extra water, gel too quickly (< 20 min) to ensure reliable processing through the facility; however, if admix is used in these formulations, the gel time is extended into the acceptable range of 20 - 60 minutes. However, adding 8 wt % extra water to the low w/p formulations increases the gel time past 60 minutes. Therefore, for gel time it may be feasible to use Daratard to process low w/p ratios as long as the dosage is very well controlled and can remain at 0.05 wt %; however, using 8 wt % extra water extends the gel time past the acceptable limits for these formulations.

Gammala	Starting Extra Water		Final	Gel Time (min)		
Sample	w/p ratio	(wt %)	w/p ratio	Pour	Vane	
WP001	0.59	0	0.59	30	35	
WP002	0.56	0	0.56	15	18	
WP003	0.67	0	0.67	40	55	
WP004	0.59	0	0.59 ^a	40	55	
WP005	0.56	8	$0.7^{\mathrm{a}}$	35	60+	
WP006	0.52	0	0.52	20	13	
WP007	0.53	0	0.53 ^a	40	21	
WP008	0.55	4	0.62	50	34	
WP009	0.69	0	0.69 ^a	120	60+	
WP010	0.58	8	0.72 ^a	45	55	
WP011	0.57	4	0.64	60	44	
WP012	0.54	0	0.54 ^a	50	43	
WP013	0.54	8	0.68	60	60+	
WP014	0.55	0	0.55	20	14	
WP015	0.61	0	0.61	50	37	
WP016	0.58	0	0.58 ^a	20	37	
WP017	0.57	0	0.57 ^a	13	30	
WP018	0.73	0	0.73	70	60+	
WP019	0.71	0	0.71 ^a	43	60+	
WP020	0.63	0	0.63	32	33	
WP021	0.53	4	$0.60^{a}$	20	40	
WP022	0.65	0	0.65 ^a	36	45	
WP023	0.51	0	0.51	15	13	
WP024	0.59	4	0.66 ^a	17	42	
WP025	0.59	8	0.74	20	58	

 Table 3-2. Comparison of gel time measurements for all mixes

^a Samples with admix included in the formulation

#### 3.1.3 Rheology Flow Curve

The yield stress and plastic viscosity results, calculated using a Bingham plastic model, are listed in Table 3-3 in order of lowest to highest yield stress. Saltstone slurries with higher flowability (Table 3-1) also have lower yield stress and plastic viscosities. The yield stress and plastic viscosity of a mixture is dependent on the final w/p ratio of the formulation. As shown in Figure 3-1, adding Daratard or extra water or both to the initial saltstone formulation lowers the yield stress and plastic viscosity of that mixture. The rheological properties can also be changed by increasing or decreasing the w/p ratio of the saltstone formulation without adding any admixtures or extra water (Figure 3-2).

All of the mixes analyzed in this study have rheological properties that meet acceptance criteria (< 8.5 Pa) except for mix WP023.^{11,32} Another part of the acceptance criteria for yield stress is "as low as possible without bleed."^{11,32} As shown in Table 3-3, mixes with high final w/p ratios (greater than 0.66) have low yield stresses and plastic viscosities which meets the criteria for processing through the SPF and increased flowability in the SDU; however, these mixes also have high bleed water and long gel and set times.

Sample	w/p start	flush water (wt%)	w/p final	Yield Stress (Pa)	Plastic Viscosity (cP)
WP019	0.71	0	0.71 ^a	2.03	44.12
WP009	0.69	0	0.69 ^a	2.06	49.83
WP018	0.73	0	0.73	2.11	42.90
WP013	0.54	8	0.68	3.09	48.12
WP015	0.61	0	0.61	3.18	58.50
WP010	0.58	8	0.72 ^a	3.20	41.56
WP005	0.56	8	0.70 ^a	3.25	43.65
WP003	0.67	0	0.67	3.32	49.43
WP022	0.65	0	0.65 ^a	3.36	53.81
WP025	0.59	8	0.74	3.59	40.59
WP024	0.59	4	0.66 ^a	3.64	52.24
WP004	0.59	0	0.59 ^a	3.89	80.79
WP020	0.63	0	0.63	3.97	67.10
WP008	0.55	4	0.62	4.07	64.88
WP012	0.54	0	0.54 ^a	5.04	121.81
WP011	0.57	4	0.64	5.08	62.21
WP007	0.53	0	0.53 ^a	5.19	116.15
WP016	0.58	0	0.58 ^a	5.29	84.20
WP021	0.53	4	0.60 ^a	5.43	70.40
WP001	0.59	0	0.59	5.51	76.07
WP017	0.57	0	0.57 ^a	5.72	109.08
WP002	0.56	0	0.56	6.66	95.02
WP014	0.55	0	0.55	7.37	101.63
WP006	0.52	0	0.52	7.58	135.32
WP023	0.51	0	0.51	10.03	138.19

Table 3-3. Yield stress and plastic viscosity results for all mixes from lowest to highest

^a Samples with admix included in the formulation



Figure 3-1. Flow curves (decreasing shear rate) for samples starting at 0.59 w/p and adding admixture or extra water or both



Figure 3-2. Flow curves (decreasing shear rate) for all mixes made without admix or extra water

# 3.2 <u>Cured Properties</u>

The impact of curing temperature on the performance (cured) properties of saltstone has been studied extensively.^{5-8,38} All of the samples analyzed in these previous studies were cured in an environment with uncontrolled humidity, resulting in drying out of the saltstone. These studies showed that the performance properties of saltstone decreased with increasing curing temperatures; however it was unclear whether the deleterious effects of curing at high temperatures were solely due to drying out of the grout or whether there were other effects of curing at high temperatures that caused the poor performance properties.⁸ The results presented in this report investigate the impact of curing environment (saturated conditions and exposure to high humidity) and temperature on the cured properties of saltstone formulated with various amounts of extra water as well as with and without admixtures. It should be noted that the samples in the previous study were cured for a total of 90 days prior to analysis whereas the samples in this study cured for 28 days. It is accepted in the grout industry that the majority of the hydration reactions have occurred after 28 days and although curing continues, the performance properties do not change significantly between 28 and 90 days of curing.^{26,39}

## 3.2.1 Porosity and Bulk Density

Grout porosity is generally defined as the percentage of total volume of cured grout that is not occupied by either the starting cementitious materials (in this case, portland cement, blast furnace slag, and Class F fly ash) or the products that results from reaction of these cementitious materials with water (calcium silicate hydrate, calcium hydroxide crystals, etc.).²⁶ For saltstone mixes, the pore volume is occupied by a salt solution.^{7,8} The cured bulk density is calculated from the cured weight and dimensions of each sample after curing for 28 days. The porosity and bulk density are reported in Table 3-4 in order of samples formulated with the lowest to highest final w/p ratio. These results support the conclusions from previous studies and literature that the porosity and bulk density of saltstone have an inverse relationship primarily controlled by the w/p ratio.^{6,8} Samples formulated at low w/p ratios are denser and have lower porosity than samples at higher w/p ratios (Table 3-4) and have higher compressive strength and lower hydraulic conductivity as discussed in the following sections.

	~		Cell K				Cell F			
Sample	Start	Final w/p ratio	Saturated		Exposed Surface		Saturated		Exposed Surface	
	w/p ratio		Density (g/cm ³ )	P (%)						
WP023	0.51	0.51	1.787	63.5	1.796	57.5	1.793	55.1	1.789	55.3
WP014	0.55	0.55	1.792	60.9	1.771	61.2	1.789	58.4	1.767	57.1
WP001	0.59	0.59	1.755	64.1	1.736	61.3	1.759	69.9	1.752	59.9
WP021	0.53	$0.60^{a,c}$	1.741	64.9	1.719	62.3	1.732	60.5	1.733	61.3
WP011	0.57	0.64 ^c	1.700	65.3	1.686	60.3	1.722	61.3	1.700	62.5
WP003	0.67	0.67	1.715	67.3	1.692	62.5	1.705	62.1	1.715	63.2
WP005	0.56	$0.70^{a,b}$	1.669	69.6	1.660	64.3	1.659	64.1	1.685	65.8
WP010	0.58	0.72 ^{a,b}	1.677	69.1	1.671	64.7	1.673	68.1	1.642	66.6
WP018	0.73	0.73	1.691	66.2	1.677	64.8	1.703	66.6	1.679	66.8

 Table 3-4. Bulk density and porosity of samples, listed from lowest to highest final w/p ratio, cured under cell K and cell F temperature profiles

^a Samples with admix included in the formulation

^b Formulation with 8 wt % extra water

^c Formulation with 4 wt % extra water

In a previous study, it was reported that curing temperature has an effect on the porosity and density of saltstone; however, the mass loss of the sample as a result of curing at elevated temperatures would affect

the cured density and therefore the grout porosity, since the cured density is used to calculate the grout porosity.⁸ Therefore, since these prior study samples were cured in an environment with uncontrolled humidity, the cured properties, specifically density, were more affected by drying than cure temperature alone. In fact, the rapid hydration at higher temperatures leads to encapsulation of porous cementitious materials by a layer of dense hydrated product, causing an overall decrease in grout porosity, assuming that the cementitious materials are completely hydrated.^{26,39} Therefore, since the samples in this study were cured in a high humidity environment and not dehydrated, the densities of these samples are higher than those cured in an uncontrolled environment. For example, samples in the previous study formulated at 0.55 w/p with no admixtures and cured at 60 °C had densities of 1.711 and 1.769 g/mL for the low and high aluminate solutions, respectively.⁸ In this study, the same formulation was cured to 55 and 80 °C under saturated conditions resulting in densities of 1.789 and 1.792 g/cm³, respectively (Table 3-4).

#### 3.2.2 Compressive Strength

Although the compressive strength of saltstone is not an input in to the PA, it is a property that provides insight into other properties of the material, such as hydraulic conductivity, porosity and density. Higher density grouts will have higher compressive strength and lower hydraulic conductivity. This is due to lower void spaces and lower porosity in the grout resulting in a higher strength material.³⁹ There are other factors that determine the compressive strength of a grout that include the microstructure, w/p ratio (including admixtures), the mixing conditions, the curing conditions (especially temperature and RH), the age, and the matter of testing (including water content in the sample.³⁹ Table 3-5 lists the compressive strength of all the samples in order of lowest to highest final w/p ratio. The general trend of the data is that low w/p samples have higher compressive strengths.

A previous report on the compressive strength of saltstone has shown a decrease in strength as a result of higher curing temperatures (approximately 1240 psi at 20 °C to 1090 psi at 40 °C);⁷ however, these samples may have been dried out, which would lead to the lower compressive strength. As shown in Table 3-5, the compressive strength of samples cured under high RH and temperature profiles up to 80 and 55 °C are greater than or equal to the 1240 psi recorded from room temperature cured samples,⁷ indicating that higher cure temperatures benefit the saltstone cured properties, provided the material is not dried out. The data in Table 3-5 shows that curing in a high humidity environment is important, than if the sample is saturated or has the surface exposed to the environment. The cell K samples have a higher average compressive strength than the cell F samples but the standard deviation indicates the two cure profiles result in saltstone with comparable compressive strengths.

	G4 4	E, 1	Compressive strength (psi)					
Sample	Starting w/n ratio	ig Final		Cell K	Cell F			
_	w/p ratio	w/p1auo	Saturated	Exposed Surface	Saturated	Exposed Surface		
WP023	0.51	0.51	3039	2978	2696	2645		
WP014	0.55	0.55	2994	3238	2372	2447		
WP001	0.59	0.59	3072	3033	1636	2193		
WP021	0.53	$0.60^{a,c}$	1743	1837	1837	1754		
WP011	0.57	0.64 ^c	1988	1737	1994	2016		
WP003	0.67	0.67	1635	2307	1491	1380		
WP005	0.56	$0.70^{a,b}$	1418	1441	1229	1266		
WP010	0.58	0.72 ^{a,b}	1168	1399	1264	1277		
WP018	0.73	0.73	1368	1475	1005	1036		
Average		2047	2161	1725	1779			
S	tandard Deviat	ion	777	747	557	576		

# Table 3-5. Compressive strength of samples, listed from lowest to highest final w/p ratio, cured under cell K and cell F temperature profiles

^a Samples with admix included in the formulation

^b Formulation with 8 wt % extra water

^c Formulation with 4 wt % extra water

## 3.2.3 Hydraulic Conductivity

Permeability is defined as the property that governs the rate of flow of a fluid into a porous solid. For steady-state flow, the coefficient of permeability, also known as hydraulic conductivity, is determined by Darcy's Equation (2-1).²⁶ The saturated hydraulic conductivity (Table 3-6) was measured on the top and bottom of each sample after curing for 28 days under each temperature profile. The samples were split to provide duplicate measurements on a single sample. All of the samples had a hydraulic conductivity of 4.5E-8 to 4.3E-10 cm/s. Of the 72 samples analyzed, only three samples had hydraulic conductivities on the order of 1E-7 cm/s (the saturated WP010 sample and both WP003 samples, all cured under the cell K profile). On average, the cell K samples had slightly higher hydraulic conductivities for samples at higher w/p ratios than the cell F samples; however, the majority of the samples have hydraulic conductivities within the range of E-9 or E-10 cm/s.

As shown in literature, there is an exponential relationship between porosity and the hydraulic conductivity or strength of grout. As the porosity increases, the strength decreases and hydraulic conductivity increases.^{26,39} Comparing the results in Table 3-4 and Table 3-5, there is a general trend that the samples with the highest porosity (higher w/p ratios) have the lowest strength and therefore highest hydraulic conductivity which indicates the w/p ratio of the formulation effects the cured properties of saltstone. However, for the hydraulic conductivity results reported in Table 3-6, it is difficult to find a trend in the data based on w/p ratio.

The results from a previous study showed a slight trend of increasing hydraulic conductivity with increasing w/p ratio, but the primary trend was higher temperatures resulted in higher hydraulic conductivities.⁸ In that study, a range of sample formulations were cured isothermally at 20, 40 and 60 °C for 28 days followed by a 62 day cure at room temperature. These samples had hydraulic conductivities ranging from 1.04E-9 to 1.90E-6 cm/s, with the majority of the 60 °C samples having hydraulic conductivities greater than 1.0E-7.⁸ It was determined that those samples were dried out, causing microcracks in the sample and higher hydraulic conductivities.^{8,39} Comparing the results presented in Table 3-6 to the results from a previous study, the curing conditions have a greater effect on the hydraulic conductivity of saltstone than the w/p ratio. The saltstone in the current study was cured in a high

humidity environment which allowed the microstructure to develop without being dehydrated, reducing the formation of microcracks.

Another previous study showed the impact of initial cure temperature on cured saltstone properties. This study showed that saltstone cured at room temperature followed by a high temperature cure had better performance properties than the same saltstone formulation cured at high temperature followed by curing at room temperature.⁷ Therefore, the combination of curing saltstone in a saturated or high humidity environment and under a ramped temperature profile results in slower hydraulic conductivities than saltstone dried out during curing and cured at high isothermal temperature.

				Hydraulic Conductivity (cm/s)					
Sampla	Starting	Final w/p	Sample	Cel	l K	Cel	ll F		
Sample	w/p ratio	ratio	Section	Saturated	Exposed Surface	Saturated	Exposed Surface		
WP023	0.51	0.51	top	9.50E-10	5.40E-10	5.80E-09	1.00E-09		
WP023 0.51	0.31	bottom	1.50E-09	3.70E-10	7.70E-10	7.20E-10			
	0.55	0.55	top	4.30E-10	3.70E-09	1.30E-09	1.80E-09		
WF014	0.55	0.55	bottom	3.90E-10	4.30E-09	3.70E-09	9.80E-10		
WD001	0.50	0.50	top	1.70E-09	4.50E-09	1.40E-09	4.30E-09		
WFUUI	0.39	0.39	bottom	1.90E-09	3.90E-10	3.60E-09	1.60E-09		
WD021	0.53	0 60 ^{a,c}	top	1.70E-09	1.70E-09	4.10E-09	2.10E-09		
WP021	0.55	0.60	bottom	2.10E-09	2.20E-09	3.70E-09	1.30E-09		
WD011	0.57	0.64°	top	3.20E-08	4.50E-08	7.00E-09	1.30E-09		
WFUII	0.57	0.04	bottom	9.60E-09	1.30E-08	5.00E-09	3.10E-09		
WD003	0.67	ZP003 0.67	0.67	top	6.20E-07	4.00E-07	3.50E-09	3.70E-09	
WF003	0.07	0.07	bottom	1.90E-09	9.80E-10	3.00E-09	1.50E-09		
WD005	0.56	0.70 ^{a,b}	top	4.30E-10	1.00E-09	7.70E-10	5.20E-10		
WF003	0.50	0.70	bottom	3.10E-10	3.30E-10	7.70E-10	5.20E-10		
WD010	0.58	0.72 ^{a,b}	top	2.80E-07	3.60E-09	4.00E-09	3.10E-09		
WPUIU	0.38	0.72	bottom	3.50E-09	2.60E-09	2.20E-09	2.50E-09		
WD019	0.72	0.72	top	7.80E-09	8.00E-09	2.10E-09	4.50E-09		
WP018	0.73	0.73	bottom	2.90E-08	3.30E-09	5.70E-09	7.60E-09		

Table 3-6. Hydraulic conductivity of samples, listed from lowest to highest final w/p ratio, cured
under cell K and cell F temperature profiles

^a Samples with admix included in the formulation

^b Formulation with 8 wt % extra water

^c Formulation with 4 wt % extra water

# 3.2.4 *Leachability Index*

The leachability indices³¹ for chromium, sodium, nitrate, and nitrite were determined for the saltstone formulations cured under the cell K profile. Note that the duplicate samples of each formulation were used to determine the leachability index and the samples were separated into two sets for convenience.

The first set of saltstone samples was WP001-A/B, WP003-A/B, WP005-A/B, WP010-A/B, WP011-A/B, Blank-1 and the second set was WP014-A/B, WP018-A/B, WP021-A/B, WP023-A/B, Blank-2.

The ANSI/ANS 16.1 standard requires many results to be reported as part of determining the effective diffusivity and leachability index of samples; however, since many of those intermediate data are not pertinent to the results of this study, they are reported in Appendix C of this report. The pH, conductivity and sodium,  $NO_2^-$  and  $NO_3^-$  concentrations for the leachate samples are given in Appendix C. Note that the concentration of Cr in the leachate was below detection limits of the analytical equipment.

The ANSI/ANS 16.1 standard specifies how to calculate the effective diffusivity of a leached species when less than 20% of the species leaches using Equation (3-1).

$$D_{e,n}^{i} = \pi * \left[\frac{\frac{a_{n}}{A_{0}}}{(\Delta t)_{n}}\right]^{2} * \left[\frac{V}{S}\right]^{2} * T$$
(3-1)

Where:

 $a_n$  = Amount of species in leachate for leaching interval n [g]

 $A_0$  = Total amount of species in the Saltstone Sample initially [g]

 $(\Delta t)_n = t_n - t_{n-1} = duration of the nth leaching interval [s]$ 

 $t_n = \sum_{k=1}^n (\Delta t)_n$  = cumulative leaching time for the nth leaching interval [s]

 $D_{e,n}^i$  = Effective diffusivity of leaching species i for leaching interval n [cm²/s]

V = Volume of cut saltstone sample [cm³]

S = External surface area of cut Saltstone sample [cm²]

T = 
$$\left[\frac{1}{2} * \left(t_n^{1/2} + t_{n-1}^{1/2}\right)\right]^2$$
 = Mean time of leaching interval [s]

Inserting the leachate masses at the various time intervals  $(a_n)$ , the initial species masses  $(A_0)$ , the time intervals  $(\Delta t)_n$  and mean leaching times (T), and the volume to surface area ratios (V/S) into Equation (3-1), the effective diffusivity of the leaching species for each time interval, n, can be calculated. Once the effective diffusivities for the Cr, Na, NO₃⁻, and NO₂⁻ species for the various leaching time intervals  $(D_{e,n}^i)$  for the saltstone samples are defined, the Leachability Index of each species (L_i) can be calculated using Equation (3-2).

$$L_{i} = \frac{1}{7} * \sum_{n=1}^{7} \log_{10} \left( \frac{1}{D_{e,n}^{i}} \right)$$
(3-2)

Where:

 $D_{e,n}^{i}$  = Effective diffusivity of leaching species i for leaching interval n [cm²/s]

# L_i = Leachability Index of species i [unitless]

The Leachability Index based on the ANSI 16.1 standard test for the Cr, Na,  $NO_3^-$ , and  $NO_2^-$  species in the saltstone samples were calculated using Equation (3-2) above using the effective diffusivities defined in Appendix C. The Leachability Indices for the saltstone samples with respect to Cr (both excluding and including the Cr in the premix) are shown in Table 3-7. The Leachability Indices for the saltstone samples with respect to Na,  $NO_3^-$  and  $NO_2^-$  are shown in Table 3-8 and Table 3-9, respectively. The last column in each of these tables shows the average Leachability Indices for the various species. Detailed inputs for evaluating Equations (3-1) and (3-2) are shown in Appendix C.

# Table 3-7. Leachability Index of Cr (excluding and *including Cr in Pre-mix) for saltstone cured under the cell K profile

Gammla	Starting	Final	Excluding Cr in Premix			Including Cr in Premix			
Sample	w/p ratio	w/p ratio	Α	В	Average	Α	В	Average	
WP001	0.59	0.59	> 10.0	> 10.0	> 10.0	> 10.6	> 10.6	> 10.6	
WP003	0.67	0.67	> 10.0	> 10.0	> 10.0	> 10.6	> 10.6	> 10.6	
WP005	0.56	$0.70^{a,b}$	> 9.8	> 9.8	> 9.8	> 10.4	> 10.5	> 10.4	
WP010	0.58	0.72 ^{a,b}	> 9.8	> 9.9	> 9.9	> 10.5	> 10.5	> 10.5	
WP011	0.57	0.64 ^c	> 9.9	> 9.9	> 9.9	> 10.5	> 10.6	> 10.6	
WP014	0.55	0.55	> 9.9	> 10.0	> 9.9	> 10.6	> 10.6	> 10.6	
WP018	0.73	0.73	> 10.0	> 10.1	> 10.0	> 10.6	> 10.6	> 10.6	
WP021	0.53	$0.60^{a,c}$	> 9.9	> 9.9	> 9.9	> 10.6	> 10.6	> 10.6	
WP023	0.51	0.51	> 9.9	> 9.9	> 9.9	> 10.6	> 10.6	> 10.6	

^a Samples with admix included in the formulation

^b Formulation with 8 wt % extra water

^c Formulation with 4 wt % extra water

*Includes Cr(III) or Cr2O3 in pre-mix along with salt solution Cr(VI) or Na2CrO4

Table 3-8. L	eachability I	ndex of Na fo	r saltstone cured	l under the cell	K profile
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Sample	Starting w/p ratio	Final w/p ratio	Α	В	Average
WP001	0.59	0.59	7.7	7.8	7.7
WP003	0.67	0.67	7.8	7.9	7.9
WP005	0.56	$0.70^{a,b}$	7.0	7.0	7.0
WP010	0.58	$0.72^{a,b}$	7.6	7.7	7.7
WP011	0.57	0.64 ^c	7.2	7.3	7.3
WP014	0.55	0.55	7.7	8.0	7.8
WP018	0.73	0.73	7.1	7.8	7.4
WP021	0.53	$0.60^{a,c}$	7.2	7.1	7.2
WP023	0.51	0.51	7.3	7.5	7.4

^a Samples with admix included in the formulation

^b Formulation with 8 wt % extra water

^c Formulation with 4 wt % extra water

Samula	Starting	Final	NO ₃			NO ₂		
Sample	w/p ratio	w/p ratio	Α	В	Average	Α	В	Average
WP001	0.59	0.59	7.0	6.9	7.0	7.1	7.0	7.0
WP003	0.67	0.67	7.3	7.4	7.3	7.3	7.4	7.4
WP005	0.56	$0.70^{a,b}$	7.0	6.9	7.0	7.0	7.0	7.0
WP010	0.58	0.72 ^{a,b}	7.3	7.3	7.3	7.1	7.1	7.1
WP011	0.57	0.64 ^c	6.9	6.9	6.9	7.3	7.3	7.3
WP014	0.55	0.55	7.3	7.5	7.4	7.3	7.5	7.4
WP018	0.73	0.73	6.6	7.3	7.0	6.7	7.4	7.0
WP021	0.53	$0.60^{a,c}$	6.8	6.6	6.7	6.9	6.7	6.8
WP023	0.51	0.51	6.8	7.0	6.9	6.8	7.1	6.9

Table 3-9. Leachability Index of NO₃⁻ and NO₂⁻ for saltstone cured under the cell K profile

^a Samples with admix included in the formulation

^b Formulation with 8 wt % extra water

^c Formulation with 4 wt % extra water

The Leachability Indices for Na ranged from 7.0 to 7.9. The Leachability Indices for  $NO_3^-$  and  $NO_2^-$  ranged from 6.9 to 7.4. A smaller the Leachability Index indicates that a constituent is more easily removed from the saltstone. Based on the formulations tested in this study, the initial formulation of the samples did not influence the rate at which contaminants were released from the saltstone. However, comparing these results to samples that were dried out, the curing environment may have an impact on the leachability of some constituents in saltstone. Based on results from another study, samples that were dried out leached more  $NO_3^-$  and  $NO_2^-$  than the samples that were kept in a moist environment.⁴⁰

# 4.0 Conclusions

Data from a typical (no setbacks or process upsets) processing day at the SPF was analyzed to calculate instantaneous and cumulative saltstone water to premix ratios over the given period of processing. A series of saltstone formulations were developed and analyzed based on these data. The fresh properties (gel, set time, bleed, viscosity, yield stress) and cured properties (hydraulic conductivity, porosity, density, and contaminant retention) of these formulations were analyzed.

The first objective of this study was to analyze saltstone fresh properties to determine the feasibility of reducing the formulation w/p ratio and while varying the amount of extra water and admixtures used during processing at the SPF. The 8 wt % extra water in the formulation causes saltstone to generate more bleed water as it cures than the samples with 4 wt % extra water or salt solution only since the water dilutes the salt solution molarity, delaying the reaction of the slag and fly ash. Adding extra water to the formulations with w/p ratios of 0.58 or lower does not increase the set time past three days and the bleed water is reabsorbed after three days, therefore, only considering the properties of bleed water and set time, it is feasible to process w/p ratios below 0.58 by adding extra water or admix. Adding Daratard to w/p ratios greater than 0.66 causes long set times and in some cases, standing bleed water after three days; therefore its use should be limited or avoided at these formulations.

The gel time is affected by extra water, even at the low w/p ratios. Saltstone formulations with low w/p ratios (0.56 or lower) without any admixture or extra water, gel too quickly (< 20 min) to ensure reliable processing through the facility. However, if admix is used in these formulations, the gel time is extended into the acceptable range of 20 - 60 minutes. Adding 8 wt % extra water to the low w/p formulations increases the gel time past 60 minutes. Therefore, for gel time it may be feasible to use Daratard to process low w/p ratios as long as the dosage is very well controlled and can remain at 0.05 wt %; however, using 8 wt % extra water extends the gel time past the acceptable limits for these formulations. The vane rheology method for determining the gel time of a sample provides insight into the structure development of the sample; however it is difficult to distinguish the gel time from settling or other reactions based on the results obtained thus far. If this method is further developed, it could be less subjective than the pour test and provide more understanding into the hydration and structure development of fresh saltstone.

Based on the fresh property data presented in this report, mixes with w/p ratios in the range of 0.57 - 0.66 formulated with 4 wt % extra water or salt solution only, produce fresh saltstone grout that satisfies acceptance criteria for set time, bleed, gel, and rheology. All of the formulations with 8 wt % extra water or formulations with w/p ratios above 0.65 that include Daratard have fresh properties outside the acceptance criteria.

The second part of this study was to provide information for understanding the impact of curing conditions (cure temperature, relative humidity (RH)) and processing formulation on the performance properties of cured saltstone. Samples were cured under profiles based on temperature data from cell K and cell F in SDU 4. Half of the samples were kept under saturated conditions by adding liquid on top of the grout to maintain complete saturation. The other half had the grout surface exposed to the humid environment. Based on the results of this study, the relative humidity of the curing environment has the most impact on the performance (cured) properties of saltstone. If the drying of the sample is eliminated or at least minimized, higher cure temperatures can be beneficial for saltstone due to increased reaction rates, formation of denser hydration products, and reduction in porosity.

The final w/p ratio of the saltstone formulation does affect the density, porosity, and compressive strength of the cured saltstone. The saltstone formulations with 0.59 w/p ratios and lower had higher densities, lower porosities, and higher compressive strength than samples formulated at a higher w/p ratio. There is a general trend that the samples formulated at high w/p ratios (0.67 and higher) have the lowest density,

highest porosity and lowest strength. However, compared to data from previous studies, the density, porosity, and compressive strength of saltstone are more controlled by curing in a high humidity environment rather than the w/p ratio of the saltstone formulation. The Leachability index for Na,  $NO_3^-$  and  $NO_2^-$  were calculated for the cured samples using the ANS/ANSI 16.1 standard. Based on the formulations tested in this study, the initial formulation of the samples did not affect the rate at which contaminants are released from the saltstone.

Samples cured for 28 days in controlled humidity environments under temperature profiles up to 55 and 80°C had hydraulic conductivities of 4.5E-8 to 4.3E-10 cm/s. On average, the cell K samples had slightly higher hydraulic conductivities for samples at higher w/p ratios than the cell F samples; however, the majority of the samples have hydraulic conductivities within the range of E-9 or E-10 cm/s. Of the 72 samples analyzed, only three samples had hydraulic conductivities on the order of 2.8E-7 cm/s. In a previous study, samples cured isothermally in an uncontrolled environment had hydraulic conductivities ranging from 1.04E-9 to 1.90E-6 cm/s, with the majority of the 60 °C samples having hydraulic conductivities greater than 1.0E-7 cm/s. Comparing the results presented in this study to the results from the previous study, the curing conditions have a greater effect on the hydraulic conductivity of saltstone than the w/p ratio. The saltstone in the current study was cured in a high humidity environment which allowed the microstructure to develop without being dehydrated, reducing the formation of microcracks. Therefore, the combination of curing saltstone in a saturated or high humidity environment and under a ramped temperature profile results in slower hydraulic conductivities than saltstone dried out during curing and cured at high isothermal temperatures.

# 5.0 Recommendations, Path Forward or Future Work

The formulations used in this study represent instantaneous water to premix ratios that are processed through the SPF. While these formulations provide valuable insight into the properties of fresh and cured saltstone, it is not representative of the entire bulk of material in the SDU. Therefore, a scaled Saltstone Facility should be used to examine cumulative effects of flush water and varying w/p ratios of a saltstone formulation throughout a day of processing. The material produced using the scaled facility can be cured in a humid environment (using saturated conditions and an exposed surface) under a temperature profile to mimic the conditions recorded in the SDU. The hydraulic conductivity and other cured properties of these samples should be measured.

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Appendix A. SDU Temperature Profiles and Facility Processing Data





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Appendix B. Vane Rheology Results

The results of the vane rheology measurements are shown in this appendix. The gel time for each sample is marked with an "X" and recorded in Table 2-1.



























Appendix C. Inputs for Calculating Leachability Index

Saltstone Bottle ID	Leachate Sample Elapsed Time						
	2 hr	7 hr	24 hr	48 hr	72 hr	96 hr	120 hr
WP001-A	9.66	9.9	10.25	10.56	10.34	10.36	10.2
WP001-B	9.48	9.91	10.29	10.65	10.25	10.4	10.24
WP003-A	9.49	9.8	10.33	10.63	10.26	10.34	10.17
WP003-B	9.3	9.88	10.18	10.55	10.36	10.26	10.32
WP005-A	10.11	10.22	10.69	10.95	10.72	10.73	10.68
WP005-B	9.46	10.3	10.71	10.93	10.72	10.69	10.65
WP010-A	9.32	9.83	10.41	10.48	10.25	10.42	10.23
WP010-B	9.26	9.9	10.31	10.49	10.2	10.39	10.23
WP011-A	9.98	9.86	10.56	10.66	10.64	10.54	10.47
WP011-B	9.68	10.01	10.52	10.7	10.33	10.37	10.57
Blank-1	7.97	8.26	8.59	8.59	8.11	7.52	8.84
WP014-A	9.62	10.25	10.57	10.51	10.49	10.63	10.64
WP014-B	10.31	9.58	10.51	10.48	10.41	10.22	10.52
WP018-A	10.58	10.12	10.86	10.87	10.69	10.63	10.82
WP018-B	10.38	10.35	10.74	10.67	10.58	10.86	10.82
WP021-A	10.4	10.37	10.6	10.52	10.5	10.52	10.59
WP021-B	10.4	10.31	10.67	10.69	10.42	10.47	10.61
WP023-A	10.3	10.17	10.64	10.71	10.39	10.54	10.72
WP023-B	10.17	10.29	10.6	10.52	10.44	10.6	10.52
Blank-2	8.49	8.81	8.98	8.98	8.85	8.62	9

Table C-1. Leachate pH

Saltstone	Leachate Sample Elapsed Time						
Bottle ID	2 hr	7 hr	24 hr	48 hr	72 hr	96 hr	120 hr
1-A	179.6	353	693	763	498	392	385
1-B	143	337	721	878	458	460	401
3-A	123	272.1	748	865	486	404	374
3-B	93.1	315	557	754	559	356	491
5-A	542	608	1316	1367	907	717	674
5-B	199.1	800	1430	1346	942	568	696
10-A	137.5	325	787	580	445	436	334
10-B	114.1	357	641	616	403	390	330
11-A	574	535	1393	1115	963	736	632
11-B	231	502	1140	1118	683	481	580
Blank-1	1.04	1.1	1.14	1.26	1.21	1.21	1.12
14-A	63.2	327	693	632	409	393	431
14-B	350	71.1	584	572	438	149.1	332
18-A	773	302	1337	1370	919	402	640
18-B	360	413	910	711	617	602	583
21-A	568	626	883	736	632	372	457
21-B	545	534	1057	984	504	396	492
23-A	436	424	957	1014	528	433	639
23-В	329	488	949	758	647	519	461
Blank-2	1.1	1.09	1.22	1.22	1.22	1.13	0.99

Table C-2. Leachate conductivities [µS/cm]

Saltstone	Leachate Sample Elapsed Time						
Bottle ID	2 hr	7 hr	24 hr	48 hr	72 hr	96 hr	120 hr
1-A	48.3	38.3	85.5	90.5	65.8	49.6	53.7
1-B	40.1	31.1	76.3	81.8	60.7	48.3	40.2
3-A	30.1	31.4	93.6	94.7	55.5	56.0	49.4
3-В	33.6	31.5	69.6	73.3	47.8	43.8	41.0
5-A	67.6	85.4	198	140	103	87.6	64.7
5-B	76.6	87.1	178	151	130	83.3	75.7
10-A	34.2	40.0	85.7	85.8	58.7	55.9	39.3
10-B	42.8	35.6	68.5	72.6	56.2	50.9	47.7
11-A	60.6	70.6	161	146	115	75.6	73.6
11-B	62.2	65.6	144	134	102	77.6	41.5
Blank-1	< 1.00	< 1.00	< 1.00	< 1.00	< 1.00	< 1.00	< 1.00
14-A	44.0	34.0	81.7	72.4	62.3	52.0	46.0
14-B	26.1	34.9	61.4	60.0	45.5	48.9	33.5
18-A	73.0	123	193	182	132.0	118	79.0
18-B	30.2	42.0	96.9	111	66.7	60.1	54.8
21-A	52.0	58.1	124	105	196	83.4	58.4
21-B	83.9	60.0	155	132	120	107	84.1
23-A	51.2	61.5	141	156	110	104	67.9
23-B	40.5	38.9	121	95.8	71.7	73.6	59.5
Blank-2	< 1.00	< 1.00	1.25	< 1.00	< 1.00	< 1.00	< 1.00

Table C-3. Leachate sodium concentrations [mg/L]

Saltstone	Leachate Sample Elapsed Time						
Bottle ID	2 hr	7 hr	24 hr	48 hr	72 hr	96 hr	120 hr
1-A	43.5	51.8	27.9	11.1	21.0	16.2	16.4
1-B	40.6	57.4	26.0	25.9	19.3	16.1	13.0
3-A	24.9	22.8	10.4	22.4	17.3	17.9	16.2
3-B	31.4	30.0	10.2	< 10.00	15.3	14.5	12.7
5-A	27.4	25.8	< 10.00	12.7	27.8	24.3	17.9
5-B	29.4	27.9	12.4	15.3	36.2	23.2	19.1
10-A	47.6	22.5	< 10.00	27.8	17.2	17.0	11.6
10-B	53.3	24.4	< 10.00	23.6	16.8	15.7	13.8
11-A	21.9	11.8	20.8	48.3	3.38	23.5	23.6
11-B	27.1	13.6	21.0	43.4	3.07	24.2	22.9
Blank-1	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00
14-A	13.2	10.7	27.6	25.0	19.8	16.7	14.1
14 <b>-</b> B	< 10.00	11.0	22.0	19.5	14.5	15.7	10.4
18-A	24.5	40.6	60.5	56.7	39.1	36.8	24.0
18-B	< 10.00	13.2	29.6	33.2	21.4	17.9	16.0
21-A	17.5	19.0	43.2	39.1	28.8	25.8	18.6
21-B	27.0	20.4	50.3	42.9	39.3	34.3	26.9
23-A	16.6	21.9	48.7	51.2	35.3	34.0	20.6
23-B	13.2	13.0	43.3	34.0	24.7	23.0	20.2
Blank-2	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00

Table C-4. Leachate NO₂⁻ Concentrations [mg/L]

Saltstone	Leachate Sample Elapsed Time						
Bottle ID	2 hr	7 hr	24 hr	48 hr	72 hr	96 hr	120 hr
1-A	297	344	182	65.5	129	99.9	96.5
1-B	273	384	174	165	118	97.8	75.8
3-A	149	136	62.7	144	109	113	102
3-B	190	178	62.4	63.4	95.0	86.6	75.7
5-A	162	152	57.1	54.8	192	168	125
5-B	176	167	72.2	74.8	248	160	132
10-A	287	127	< 10.00	173	110	110	71.4
10-B	323	141	11.4	145	108	99.5	84.9
11-A	134	68.8	127	301	219	153	157
11-B	165	81.1	123	270	197	156	150
Blank-1	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00
14-A	78.2	64.0	158	143	114	99.1	85.6
14-B	43.4	62.1	130	112	84.0	94.9	60.2
18-A	145	258	384	362	250	237	153
18-B	58.5	81.3	189	214	138	116	103
21-A	111	113	265	241	175	163	113
21-B	163	119	308	265	250	217	169
23-A	98.0	132	293	315	219	209	126
23-B	76.0	75.7	252	205	145	141	122
Blank-2	< 10.00	10.2	< 10.00	< 10.00	< 10.00	< 10.00	< 10.00

Table C-5. Leachate NO₃⁻ concentrations [mg/L]

To calculate the amount of species in the Saltstone sample initially ( $A_0$ ), the composition of the various components going into the Saltstone sample had to be identified. The salt solution was mixed with the pre-mix, an admixture, and some additional water for each Saltstone sample as shown in Table C-6. The wt% of the species Cr, Na, NO₃⁻, and NO₂⁻ in the salt solution are shown in Table C-7. For the pre-mix components (Cement, Slag, Fly Ash,) the weight percent of the Cr, Na, NO₃⁻, and NO₂⁻ species are shown in Table C-8. For the Saltstone samples in this study the pre-mix components were mixed as 10 wt% cement, 45 wt% Slag, and 45 wt% fly ash. Using the mass and component concentration data for the salt solution, pre-mix, admixture, and additional water, the wt% of the species Cr, Na, NO₃⁻, and NO₂⁻ in the final Saltstone mix were calculated as shown in Table C-9. The calculations showed that the amount of Cr(III) from the Cr₂O₃ in the pre-mix was about equal to the amount of Cr(VI) from Na₂CrO₄ as the Cr* column in Table C-9 illustrates. Normally the amount of Cr(III) from the Cr₂O₃ in pre-mix is treated as insoluble or not leachable once in the Saltstone final form. To be conservative, the total amount of Cr in the initial Saltstone sample (A₀) is calculated using the concentration of Cr with and without the pre-mix contribution. Note that the NO₃⁻ and NO₂⁻ concentrations for the pre-mix components are zero and the Cr, Na, NO₃⁻, and NO₂⁻ concentrations in the admix are considered zero.

Saltstone Sample ID	Salt Solution	Pre- mix	Admixture	Additional Water
WP001	2490	3009.5	0	0
WP003	305.6	325	0.1	0
WP005	255.4	325	0.1	45.5
WP010	2281	2815	2.25	402.6
WP011	1540.4	1924.4	0.58	134.7
WP014	2394.5	3100	0.93	0
WP018	2783.4	2715	0.811	0
WP021	1476	1983.8	1.59	138.6
WP023	1502	2096.1	0.63	0

Table C-6	Saltstone	<b>Component Masses</b>	[g]	
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 Table C-7. Salt Solution Component Concentrations [wt%]

Cr	Na	NO ₃	NO ₂
0.024	9.93	11.75	2.03

Table C 0, TTC mix Component Concentrations ( wt/v	Table C-8.	Pre-mix Com	ponent Conce	entrations [wt%	1
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Pre-mix Component	Cr	Na	NO ₃ ⁻	NO ₂ ⁻
Cement	0.019	0.183	0	0
Slag	0.016	0.176	0	0
Fly Ash	0.027	0.349	0	0

 Table C-9. Saltstone Mix Component Concentrations [wt%]

Saltstone Sample ID	Cr	Cr*	Na	NO ₃ -	NO ₂
WP001	0.0109	0.0227	4.6363	5.3190	0.9198
WP003	0.0117	0.0228	4.9438	5.6922	0.9843
WP005	0.0098	0.0211	4.1844	4.7929	0.8288
WP010	0.0100	0.0210	4.2488	4.8713	0.8424
WP011	0.0103	0.0219	4.3859	5.0266	0.8692
WP014	0.0105	0.0227	4.4713	5.1188	0.8852
WP018	0.0122	0.0228	5.1529	5.9460	1.0282
WP021	0.0099	0.0218	4.2125	4.8165	0.8329
WP023	0.0101	0.0226	4.2937	4.9031	0.8479

*Includes Cr(III) or Cr₂O₃ in pre-mix along with salt solution Cr(VI) or Na₂CrO₄

Now that the Saltstone mix concentrations in Table C-9 are defined, the masses of the initial cut Saltstone Samples are needed as shown in Table C-10. Note that there were multiple vials of each Saltstone mix that were cured and only the masses of the vials leached are shown in Table C-10.

Using the concentrations of Table C-9 and the masses in Table C-10, the initial amount of the various species in the Saltstone Samples  $(A_0)$  can be calculated. Excluding the Cr in the pre-mix, the initial

amounts of Cr in the Saltstone Samples are shown in Table C-11. Including the Cr in the pre-mix, the initial amounts of Cr in the Saltstone Samples are shown in Table C-12. The initial amounts of Na,  $NO_3^-$ , and  $NO_2^-$  in the cut Saltstone Samples are shown in Table C-13, Table C-14 and Table C-15, respectively.

Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
WP001		86.8277	87.318	
WP003	84.5618			84.1677
WP005	81.5039			79.4580
WP010	82.8712		82.4526	
WP011			82.8827	84.2573
WP014	86.2742			87.4940
WP018			82.4400	84.5803
WP021	86.9141		86.8276	
WP023		87.3019	88.3543	

Table C-10. Initial masses of cut saltstone samples

Table C-11. Cut Saltstone Sample Initial Cr Masses [g]

Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
WP001		0.009	0.010	
WP003	0.010			0.010
WP005	0.008			0.008
WP010	0.008		0.008	
WP011			0.009	0.009
WP014	0.009			0.009
WP018			0.010	0.010
WP021	0.009		0.009	
WP023		0.009	0.009	

Table C-12. Cut Saltstone Sample Initial Cr* Masses [g]

Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
WP001		0.020	0.020	
WP003	0.019			0.019
WP005	0.017			0.017
WP010	0.017		0.017	
WP011			0.018	0.018
WP014	0.020			0.020
WP018			0.019	0.019
WP021	0.019		0.019	
WP023		0.020	0.020	

*Includes Cr(III) or Cr₂O₃ in pre-mix along with salt solution Cr(VI) or Na₂CrO₄

Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
WP001		4.026	4.048	
WP003	4.181			4.161
WP005	3.410			3.325
WP010	3.521		3.503	
WP011			3.635	3.695
WP014	3.858			3.912
WP018			4.248	4.358
WP021	3.661		3.658	
WP023		3.748	3.794	

Table C-13. Cut Saltstone Sample Initial Na Masses [g]

Table C-14. Cut Saltstone Sample Initial NO₃⁻ Masses [g]

Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
WP001		4.618	4.644	
WP003	4.813			4.791
WP005	3.906			3.808
WP010	4.037		4.017	
WP011			4.166	4.235
WP014	4.416			4.479
WP018			4.902	5.029
WP021	4.186		4.182	
WP023		4.281	4.332	

Table C-15. Cut Saltstone Sample Initial NO₂⁻ Masses [g]

Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
WP001		0.799	0.803	
WP003	0.832			0.828
WP005	0.676			0.659
WP010	0.698		0.695	
WP011			0.720	0.732
WP014	0.764			0.774
WP018			0.848	0.870
WP021	0.724		0.723	
WP023		0.740	0.749	

The volume to surface area ratios for the cut Saltstone samples are shown in Table C-16. Note that since the volumes are in  $cm^3$  and the surface areas are in  $cm^2$  then the ratios are in cm.

Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
WP001		1.292	1.289	
WP003	1.295			1.289
WP005	1.320			1.287
WP010	1.292		1.286	
WP011			1.285	1.286
WP014	1.283			1.284
WP018			1.286	1.280
WP021	1.297		1.292	
WP023		1.278	1.293	

Table C 10. Cat ballstone ballple volume to ballace filea Rados (v/b) [em	<b>m</b> ]
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Since the mass or volume of leachant used and the concentration of each species in the leachate are known for each Saltstone sample at each time interval, the mass of each species in the leachant at each time interval can be calculated. The time intervals in the ANSI/ANS 16.1 standard are shown in Table C-17. Table C-18 shows the leachate masses for Cr if the Cr in the pre-mix is excluded for each time interval and Table C-19 shows the leachate masses for Cr if the Cr in the pre-mix is included. Table C-20, Table C-21, and Table C-22show the leachate masses for Na, NO₃⁻, and NO₂⁻ for each time interval, respectively.

	-		
Time Interval n	t _n	$(\Delta t)_n$	Т
1	7,200	7,200	1,800
2	18,000	25,200	14,835
3	86,400	61,200	51,231
4	172,800	86,400	125,894
5	259,200	86,400	213,818
6	345,600	86,400	300,849
7	432,000	86,400	387,596

Table C-17. Leaching Time Parameters [s]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
1	WP011			< 7.75E-06	<7.77E-06
	WP014	< 7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	<7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
2	WP011			< 7.75E-06	< 7.77E-06
	WP014	< 7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
3	WP011		_	< 7.75E-06	< 7.77E-06
	WP014	<7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
4	WP011			< 7.75E-06	<7.77E-06
	WP014	<7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
5	WP011			< 7.75E-06	<7.77E-06
	WP014	< 7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	

Table C-18. Leachate Masses  $(a_n)$  for Cr [g]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
6	WP011			< 7.75E-06	< 7.77E-06
7	WP014	< 7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
	WP011			< 7.75E-06	< 7.77E-06
	WP014	< 7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	

Table C-18. Leachate Masses  $(a_n)$  for Cr [g]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
1	WP011			< 7.75E-06	<7.77E-06
	WP014	<7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	<7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			<7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
2	WP011			< 7.75E-06	<7.77E-06
	WP014	< 7.76E-06			< 7.76E-06
	WP018	+7.04E.06		< 7.78E-06	< /.68E-06
	WP021	< 7.94E-06	47.60E.06	< 7.90E-06	
	WP023		< 7.87E.06	< 7.81E-06	
2	WP001	< 7.00E.06	< /.8/E-00	< 7.88E-00	< 7.91E.06
	WP005	< 7.90E-00			< 7.81E-00
	WP010	< 3.17E-00		< 7.76E.06	< 7.78E-00
	WP010	< 7.84L-00		< 7.76E-06	< 7.77E-06
5	WP014	< 7.76E-06		< 1.15E 00	< 7.77E-06
	WP014	< 7.76£ 00		< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
4	WP011			< 7.75E-06	<7.77E-06
	WP014	< 7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
5	WP011			< 7.75E-06	<7.77E-06
	WP014	< 7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	

Table C-19. Leachate Masses  $(a_n)$  for  $Cr^*$  [g]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
6	WP011			< 7.75E-06	< 7.77E-06
7	WP014	< 7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	
	WP001		< 7.87E-06	< 7.88E-06	
	WP003	< 7.90E-06			< 7.81E-06
	WP005	< 8.17E-06			< 7.78E-06
	WP010	< 7.84E-06		< 7.76E-06	
	WP011			< 7.75E-06	< 7.77E-06
	WP014	<7.76E-06			< 7.76E-06
	WP018			< 7.78E-06	< 7.68E-06
	WP021	< 7.94E-06		< 7.90E-06	
	WP023		< 7.66E-06	< 7.81E-06	

Table C-19. Leachate Masses (a_n) for Cr* [g]

*Includes Cr(III) or  $Cr_2O_3$  in pre-mix along with salt solution Cr(VI) or  $Na_2CrO_4$ 

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
1	WP001		0.0380	0.0316	
	WP003	0.0238			0.0262
	WP005	0.0552			0.0596
	WP010	0.0268		0.0332	
	WP011			0.0470	0.0483
	WP014	0.0341			0.0203
	WP018			0.0568	0.0232
	WP021	0.0413		0.0663	
	WP023		0.0392	0.0316	
	WP001		0.0302	0.0245	
	WP003	0.0248			0.0246
	WP005	0.0698			0.0678
2	WP010	0.0314		0.0276	
	WP011			0.0547	0.0510
	WP014	0.0264			0.0271
	WP018		-	0.0957	0.0323
	WP021	0.0461		0.0474	
	WP023		0.0471	0.0304	
	WP001		0.0673	0.0601	
	WP003	0.0739			0.0543
3	WP005	0.1618			0.1385
	WP010	0.0672		0.0532	
	WP011			0.1248	0.1119
	WP014	0.0634			0.0476
	WP018			0.1499	0.0744
	WP021	0.0987		0.1227	
	WP023		0.1078	0.0942	
4	WP001		0.0713	0.0644	
	WP003	0.0748	0		0.0572
	WP005	0.1144		0.054	0.1175
	WP010	0.0673		0.0564	0.10.11
	WP011	0.05.60	—	0.1132	0.1041
	WP014	0.0562		0.1416	0.0466
	WP018	0.0024		0.1416	0.0852
	WP021	0.0834	0.1105	0.1043	
	WP023		0.1195	0.0749	
	WP001	0.0429	0.0518	0.0478	0.0272
	WP005	0.0438			0.03/3
	WP005	0.0842		0.0426	0.1012
	WP010 WD011	0.0460		0.0436	0.0702
	WP011	0.0492		0.0892	0.0793
	WP014	0.0483		0.1027	0.0353
	WP018	0.1555		0.1027	0.0512
	WP021	0.1557		0.0948	

Table C-20. Leachate Masses (a_n) for Na [g]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP023		0.0843	0.0560	
6	WP001		0.0391	0.0380	
	WP003	0.0442			0.0342
	WP005	0.0716			0.0648
	WP010	0.0438		0.0395	
	WP011			0.0586	0.0603
	WP014	0.0403			0.0379
	WP018			0.0918	0.0461
	WP021	0.0662		0.0847	
	WP023		0.0794	0.0575	
7	WP001		0.0423	0.0317	
	WP003	0.0390			0.0320
	WP005	0.0529			0.0589
	WP010	0.0308		0.0370	
	WP011			0.0571	0.0322
	WP014	0.0357			0.0260
	WP018			0.0615	0.0421
	WP021	0.0464		0.0664	
	WP023		0.0520	0.0465	

Table C-20. Leachate Masses  $\left(a_{n}\right)$  for Na  $\left[g\right]$ 

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
1	WP001		0.2339	0.2150	
	WP003	0.1176			0.1484
	WP005	0.1324			0.1370
	WP010	0.2251	·	0.2507	
	WP011			0.1039	0.1282
	WP014	0.0607			0.0337
	WP018			0.1128	0.0449
	WP021	0.0882		0.1287	
	WP023		0.0751	0.0594	
	WP001		0.2709	0.3025	
	WP003	0.1074			0.1390
	WP005	0.1242			0.1300
	WP010	0.0996		0.1095	
2	WP011			0.0533	0.0630
	WP014	0.0496			0.0482
	WP018			0.2007	0.0624
	WP021	0.0897		0.0940	
	WP023		0.1010	0.0592	
	WP001		0.1433	0.1371	
3	WP003	0.0495			0.0487
	WP005	0.0467			0.0562
	WP010	< 0.0078		0.0088	
	WP011			0.0985	0.0956
	WP014	0.1225			0.1007
	WP018			0.2986	0.1450
	WP021	0.2101		0.2432	
	WP023		0.2246	0.1968	
4	WP001		0.0516	0.1300	
	WP003	0.1137			0.0495
	WP005	0.0448			0.0582
	WP010	0.1357		0.1126	
	WP011			0.2334	0.2098
	WP014	0.1109			0.0867
	WP018			0.2819	0.1642
	WP021	0.1915		0.2094	
	WP023		0.2414	0.1602	
5	WP001		0.1016	0.0929	
	WP003	0.0861			0.0742
	WP005	0.1569			0.1930
	WP010	0.0863		0.0838	
	WP011			0.1698	0.1531
	WP014	0.0885			0.0652
	WP018			0.1942	0.1059
	WP021	0.1389		0.1971	
	WP023		0.1677	0.1130	

Table C-21. Leachate Masses (a_n) for NO₃⁻ [g]
Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		0.0787	0.0770	
	WP003	0.0892			0.0676
	WP005	0.1373			0.1245
	WP010	0.0863	·	0.0772	
6	WP011			0.1186	0.1212
	WP014	0.0769			0.0736
	WP018			0.1847	0.0892
	WP021	0.1296		0.1716	
	WP023		0.1601	0.1102	
	WP001		0.0760	0.0597	
	WP003	0.0805			0.0591
	WP005	0.1022			0.1027
	WP010	0.0560		0.0659	
7	WP011			0.1217	0.1165
	WP014	0.0664			0.0467
	WP018			0.1190	0.0791
	WP021	0.0897		0.1335	
	WP023		0.0965	0.0953	

Table C-21. Leachate Masses  $(a_n)$  for  $NO_3^-[g]$ 

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		0.0343	0.0320	
	WP003	0.0197			0.0245
	WP005	0.0224			0.0229
ĺ	WP010	0.0373		0.0414	
1	WP011			0.0170	0.0211
	WP014	0.0102			< 0.0078
	WP018			0.0191	< 0.0077
	WP021	0.0139		0.0213	
	WP023		0.0127	0.0103	
	WP001		0.0408	0.0452	
	WP003	0.0180			0.0234
	WP005	0.0211			0.0217
	WP010	0.0176		0.0189	
2	WP011			0.0091	0.0106
	WP014	0.0083			0.0085
	WP018			0.0316	0.0101
	WP021	0.0151		0.0161	
	WP023		0.0168	0.0102	
	WP001		0.0220	0.0205	
	WP003	0.0082			0.0080
	WP005	< 0.0082			0.0097
	WP010	< 0.0078		< 0.0078	
3	WP011			0.0161	0.0163
	WP014	0.0214			0.0170
	WP018			0.0471	0.0227
	WP021	0.0343		0.0397	
	WP023		0.0373	0.0338	
	WP001		0.0087	0.0204	
	WP003	0.0177			< 0.0078
	WP005	0.0104			0.0119
	WP010	0.0218		0.0183	
4	WP011			0.0374	0.0337
	WP014	0.0194			0.0151
	WP018			0.0441	0.0255
	WP021	0.0311		0.0339	
	WP023		0.0392	0.0265	
	WP001		0.0165	0.0152	
	WP003	0.0137			0.0119
	WP005	0.0227			0.0282
	WP010	0.0135		0.0130	
5	WP011			0.0026	0.0024
	WP014	0.0154			0.0113
	WP018			0.0304	0.0164
	WP021	0.0229		0.0310	
	WP023		0.0270	0.0193	

Table C-22. Leachate Masses  $(a_n)$  for  $NO_2^-[g]$ 

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		0.0128	0.0127	
	WP003	0.0141			0.0113
	WP005	0.0199			0.0181
	WP010	0.0133		0.0122	
6	WP011			0.0182	0.0188
	WP014	0.0130			0.0122
	WP018			0.0287	0.0137
	WP021	0.0205		0.0271	— — —
	WP023		0.0260	0.0180	
	WP001		0.0129	0.0102	
	WP003	0.0128			0.0099
	WP005	0.0146			0.0149
	WP010	0.0091		0.0107	
7	WP011			0.0183	0.0178
	WP014	0.0109			0.0081
	WP018			0.0187	0.0123
	WP021	0.0148		0.0212	
	WP023		0.0158	0.0158	

Table C-22. Leachate Masses (a_n) for NO₂⁻[g]

Inserting the leachate masses at the various time intervals ( $a_n$ ) from Table C-18 through Table C-22, the initial species masses ( $A_0$ ) from Table C-11 through Table C-15, the time intervals ( $\Delta t$ )_n and mean leaching times (T) from Table C-17, and the volume to surface area ratios (V/S) from Table C-16 into Equation (3-1), the effective diffusivity of the leaching species for each time interval n can be calculated. Table C-23 shows the effective diffusivity for Cr if the Cr in the pre-mix is excluded for each time interval and Table C-24 shows the effective diffusivity for Cr if the Cr in the pre-mix is included. Table C-25, Table C-26, and Table C-27 show the effective diffusivity for Na, NO₃⁻, and NO₂⁻, respectively.

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		< 1.26E-10	< 1.24E-10	
	WP003	<1.17E-10			<1.14E-10
	WP005	<1.97E-10			< 1.79E-10
	WP010	<1.64E-10		< 1.60E-10	
1	WP011			< 1.48E-10	< 1.44E-10
	WP014	<1.32E-10			< 1.29E-10
	WP018			< 1.08E-10	< 9.92E-11
	WP021	<1.57E-10		< 1.55E-10	
	WP023		< 1.36E-10	< 1.41E-10	
	WP001		< 1.66E-10	< 1.63E-10	1 202 10
	WP003	<1.54E-10			< 1.50E-10
	WP005	< 2.60E-10		<b>2 12 1</b> 0	< 2.36E-10
	WP010	< 2.16E-10		< 2.12E-10	1.005 10
2	WP011	· 1 74E 10		< 1.95E-10	< 1.90E-10
	WP014	< 1.74E-10		× 1 42E 10	< 1.70E-10
	WP018	< 2.07E 10		< 1.43E-10	< 1.31E-10
	WP021	< 2.07E-10	< 1.70E 10	< 2.04E-10	
	WP025		< 1.79E-10	< 1.80E-10	
	WP001	< 4 50E 11	< 4.94E-11	< 4.0/E-11	< 4 40E 11
	WP005	< 4.39E-11			< 4.49E-11
	WP010	< 6.45F-11		< 6 32E-11	< 7.04E-11
3	WP011	< 0.45E 11		< 5.82E-11	< 5 66E-11
5	WP014	< 5 19E-11		< 5.02E 11	< 5.06E-11
	WP018			< 4.26E-11	< 3.91E-11
	WP021	< 6.19E-11		< 6.09E-11	
	WP023		< 5.35E-11	< 5.56E-11	
	WP001		< 6.10E-11	< 6.00E-11	
	WP003	< 5.66E-11		_	< 5.54E-11
	WP005	< 9.58E-11			< 8.68E-11
	WP010	< 7.95E-11		< 7.79E-11	
4	WP011			< 7.18E-11	< 6.98E-11
	WP014	< 6.40E-11			< 6.24E-11
	WP018			< 5.25E-11	< 4.82E-11
	WP021	< 7.63E-11		< 7.51E-11	
	WP023		< 6.60E-11	< 6.85E-11	
	WP001		< 1.04E-10	< 1.02E-10	
	WP003	< 9.62E-11			< 9.40E-11
	WP005	< 1.63E-10			< 1.47E-10
_	WP010	< 1.35E-10		< 1.32E-10	
5	WP011	1.057		< 1.22E-10	< 1.19E-10
	WP014	< 1.09E-10		0.017.11	< 1.06E-10
	WP018	.1.005.10		< 8.91E-11	< 8.18E-11
	WP021	< 1.30E-10	. 1 105 10	< 1.2/E-10	
	WP023		< 1.12E-10	< 1.16E-10	

Table C-23. Effective Diffusivity  $(D_{e,n}^{Na})$  for Cr [cm²/s]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		< 1.46E-10	< 1.43E-10	
	WP003	<1.35E-10			< 1.32E-10
	WP005	< 2.29E-10			< 2.07E-10
	WP010	< 1.90E-10		< 1.86E-10	
6	WP011			< 1.72E-10	< 1.67E-10
	WP014	<1.53E-10			< 1.49E-10
	WP018			< 1.25E-10	< 1.15E-10
	WP021	< 1.82E-10		< 1.79E-10	
	WP023		< 1.58E-10	< 1.64E-10	
	WP001		< 1.88E-10	< 1.85E-10	
	WP003	<1.74E-10			< 1.70E-10
	WP005	< 2.95E-10			< 2.67E-10
	WP010	< 2.45E-10		< 2.40E-10	
7	WP011			< 2.21E-10	< 2.15E-10
· · ·	WP014	<1.97E-10			< 1.92E-10
	WP018			< 1.62E-10	< 1.48E-10
	WP021	< 2.35E-10		< 2.31E-10	
	WP023		< 2.03E-10	< 2.11E-10	

Table C-23. Effective Diffusivity  $(D_{e,n}^{Na})$  for Cr [cm²/s]

*Includes Cr(III) or Cr₂O₃ in pre-mix along with salt solution Cr(VI) or Na₂CrO₄

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		< 2.90E-11	< 2.85E-11	
	WP003	< 3.06E-11			< 2.99E-11
	WP005	< 4.31E-11			< 3.91E-11
	WP010	< 3.69E-11		< 3.62E-11	
1	WP011			< 3.30E-11	< 3.21E-11
	WP014	< 2.82E-11			< 2.75E-11
	WP018			< 3.08E-11	< 2.82E-11
	WP021	< 3.23E-11		< 3.18E-11	
	WP023		< 2.68E-11	< 2.79E-11	
	WP001		< 3.82E-11	< 3.76E-11	
	WP003	< 4.04E-11			< 3.95E-11
	WP005	< 5.69E-11			< 5.15E-11
	WP010	< 4.86E-11		< 4.77E-11	
2	WP011			< 4.35E-11	< 4.23E-11
	WP014	< 3.72E-11			< 3.63E-11
	WP018			< 4.06E-11	< 3.72E-11
	WP021	< 4.26E-11		< 4.19E-11	
	WP023		< 3.54E-11	< 3.67E-11	
	WP001		<1.14E-11	<1.12E-11	
	WP003	<1.21E-11			<1.18E-11
	WP005	<1.70E-11			<1.54E-11
	WP010	<1.45E-11		< 1.42E-11	
3	WP011			< 1.30E-11	< 1.26E-11
	WP014	<1.11E-11			< 1.09E-11
	WP018			< 1.21E-11	<1.11E-11
	WP021	<1.27E-11		< 1.25E-11	
	WP023		< 1.06E-11	< 1.10E-11	
	WP001		<1.41E-11	< 1.39E-11	
	WP003	<1.49E-11			<1.45E-11
	WP005	< 2.09E-11			< 1.90E-11
	WP010	<1.79E-11		< 1.76E-11	
4	WP011			< 1.60E-11	<1.56E-11
	WP014	<1.37E-11			<1.34E-11
	WP018			< 1.49E-11	<1.37E-11
	WP021	<1.57E-11		< 1.54E-11	
	WP023		< 1.30E-11	< 1.35E-11	
	WP001		< 2.39E-11	< 2.35E-11	
	WP003	< 2.53E-11			< 2.47E-11
	WP005	< 3.56E-11			< 3.22E-11
5	WP010	< 3.03E-11		< 2.98E-11	
5	WP011			< 2.72E-11	< 2.65E-11
	WP014	< 2.33E-11			< 2.27E-11
	WP018			< 2.54E-11	< 2.33E-11
	WP021	< 2.67E-11		< 2.62E-11	

Table C-24. Effective Diffusivity  $(D_{e,n}^{Na})$  for Cr* [cm²/s]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP023		< 2.21E-11	< 2.30E-11	
	WP001		< 3.36E-11	< 3.31E-11	
	WP003	< 3.56E-11			< 3.48E-11
	WP005	< 5.01E-11			< 4.54E-11
	WP010	< 4.28E-11		< 4.20E-11	
6	WP011			< 3.83E-11	< 3.72E-11
	WP014	< 3.28E-11			< 3.20E-11
	WP018			< 3.57E-11	< 3.28E-11
	WP021	< 3.75E-11		< 3.69E-11	
	WP023		< 3.11E-11	< 3.23E-11	
	WP001		< 4.33E-11	< 4.27E-11	
	WP003	< 4.58E-11			< 4.48E-11
	WP005	< 6.45E-11			< 5.84E-11
	WP010	< 5.52E-11		< 5.41E-11	
7	WP011			< 4.93E-11	< 4.79E-11
	WP014	< 4.22E-11			< 4.12E-11
	WP018			< 4.60E-11	< 4.22E-11
	WP021	< 4.84E-11		< 4.75E-11	
	WP023		< 4.01E-11	< 4.17E-11	

Table C-24. Effective Diffusivity  $(D_{e,n}^{Na})$  for Cr* [cm²/s]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		1.63E-08	1.10E-08	
	WP003	5.91E-09			7.20E-09
	WP005	4.99E-08			5.81E-08
	WP010	1.06E-08		1.62E-08	
1	WP011			3.01E-08	3.08E-08
	WP014	1.41E-08			4.82E-09
	WP018			3.22E-08	5.06E-09
	WP021	2.34E-08		5.98E-08	
	WP023		1.95E-08	1.27E-08	
	WP001		1.35E-08	8.76E-09	
	WP003	8.48E-09			8.35E-09
	WP005	1.05E-07			9.90E-08
	WP010	1.91E-08		1.48E-08	
2	WP011			5.39E-08	4.52E-08
	WP014	1.11E-08			1.14E-08
	WP018			1.21E-07	1.29E-08
	WP021	3.84E-08		4.03E-08	
	WP023		3.71E-08	1.54E-08	
	WP001		2.01E-08	1.57E-08	
	WP003	2.25E-08			1.22E-08
	WP005	1.69E-07			1.23E-07
	WP010	2.62E-08		1.64E-08	
3	WP011			8.37E-08	6.51E-08
	WP014	1.91E-08			1.05E-08
	WP018			8.85E-08	2.05E-08
	WP021	5.26E-08		8.08E-08	
	WP023		5.81E-08	4.43E-08	
	WP001		2.77E-08	2.23E-08	
	WP003	2.84E-08			1.66E-08
	WP005	1.04E-07			1.10E-07
	WP010	3.23E-08		2.27E-08	
4	WP011			8.48E-08	6.95E-08
	WP014	1.85E-08			1.24E-08
	WP018			9.73E-08	3.32E-08
	WP021	4.62E-08		7.19E-08	
	WP023		8.80E-08	3.45E-08	
	WP001		2.49E-08	2.09E-08	
	WP003	1.66E-08			1.20E-08
	WP005	9.55E-08			1.38E-07
	WP010	2.57E-08		2.31E-08	
5	WP011			8.94E-08	6.84E-08
	WP014	2.32E-08			1.21E-08
	WP018			8.69E-08	2.04E-08
	WP021	2.74E-07		1.01E-07	
	WP023		7.43E-08	3.28E-08	

Table C-25. Effective Diffusivity  $(D_{e,n}^{Na})$  for Na [cm²/s]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		1.99E-08	1.86E-08	
	WP003	2.38E-08			1.42E-08
	WP005	9.72E-08			7.97E-08
	WP010	3.28E-08		2.67E-08	
6	WP011			5.44E-08	5.57E-08
	WP014	2.28E-08			1.96E-08
	WP018			9.77E-08	2.33E-08
	WP021	6.97E-08		1.13E-07	
	WP023		9.29E-08	4.86E-08	
	WP001		3.01E-08	1.66E-08	
	WP003	2.38E-08			1.60E-08
	WP005	6.83E-08			8.48E-08
	WP010	2.09E-08		3.02E-08	
7	WP011			6.64E-08	2.05E-08
	WP014	2.30E-08			1.19E-08
	WP018			5.64E-08	2.49E-08
	WP021	4.40E-08		8.98E-08	
	WP023		5.13E-08	4.09E-08	

Table C-25. Effective Diffusivity  $(D_{e,n}^{Na})$  for Na  $[\text{cm}^2/\text{s}]$ 

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		4.67E-07	3.89E-07	
	WP003	1.09E-07			1.74E-07
	WP005	2.18E-07			2.34E-07
	WP010	5.67E-07		7.04E-07	
1	WP011			1.12E-07	1.65E-07
	WP014	3.39E-08			1.02E-08
	WP018			9.55E-08	1.43E-08
	WP021	8.14E-08		1.73E-07	
	WP023		5.48E-08	3.43E-08	
	WP001		8.27E-07	1.01E-06	
	WP003	1.20E-07			2.01E-07
	WP005	2.54E-07			2.77E-07
	WP010	1.46E-07		1.77E-07	
2	WP011			3.89E-08	5.26E-08
	WP014	2.99E-08			2.75E-08
	WP018			3.99E-07	3.63E-08
	WP021	1.11E-07		1.21E-07	
	WP023		1.31E-07	4.48E-08	
	WP001		6.91E-08	6.22E-08	
	WP003	7.62E-09			7.38E-09
	WP005	1.07E-08			1.55E-08
	WP010	< 2.71E-10		3.45E-10	
3	WP011			3.96E-08	3.62E-08
	WP014	5.44E-08			3.59E-08
	WP018			2.64E-07	5.85E-08
	WP021	1.82E-07	-	2.43E-07	
	WP023		1.93E-07	1.48E-07	
	WP001		1.10E-08	6.90E-08	
	WP003	4.96E-08			9.39E-09
	WP005	1.21E-08			2.05E-08
	WP010	1.00E-07		6.89E-08	
4	WP011			2.75E-07	2.15E-07
	WP014	5.50E-08			3.27E-08
	WP018			2.90E-07	9.26E-08
	WP021	1.87E-07		2.22E-07	
	WP023		2.75E-07	1.21E-07	
	WP001		7.27E-08	5.99E-08	
	WP003	4.82E-08			3.58E-08
	WP005	2.53E-07			3.83E-07
_	WP010	6.87E-08		6.49E-08	1.0.17
5	WP011			2.47E-07	1.94E-07
	WP014	5.95E-08			3.14E-08
	WP018			2.33E-07	6.54E-08
	WP021	1.67E-07		3.34E-07	
	WP023		2.26E-07	1.02E-07	

Table C-26. Effective Diffusivity  $(D_{e,n}^{Na})$  for NO₃⁻ [cm²/s]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001		6.14E-08	5.79E-08	
	WP003	7.30E-08			4.19E-08
	WP005	2.73E-07			2.24E-07
	WP010	9.66E-08		7.75E-08	
6	WP011			1.69E-07	1.71E-07
	WP014	6.31E-08			5.64E-08
	WP018			2.97E-07	6.53E-08
	WP021	2.04E-07		3.56E-07	
	WP023		2.89E-07	1.37E-07	
	WP001		7.38E-08	4.48E-08	
	WP003	7.66E-08			4.12E-08
	WP005	1.94E-07			1.96E-07
	WP010	5.24E-08		7.27E-08	
7	WP011			2.30E-07	2.04E-07
	WP014	6.07E-08			2.93E-08
	WP018			1.59E-07	6.61E-08
	WP021	1.26E-07		2.77E-07	
	WP023		1.36E-07	1.32E-07	

Table C-26. Effective Diffusivity  $(D_{e,n}^{Na})$  for NO₃⁻ [cm²/s]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
	WP001	_	3.35E-07	2.88E-07	
	WP003	1.02E-07			1.59E-07
	WP005	2.09E-07			2.18E-07
	WP010	5.21E-07		6.41E-07	
1	WP011			1.00E-07	1.49E-07
	WP014	3.23E-08			<1.81E-08
	WP018			9.12E-08	< 1.39E-08
	WP021	6.77E-08		1.58E-07	
	WP023		5.26E-08	3.46E-08	
	WP001		6.27E-07	7.58E-07	
	WP003	1.13E-07			1.91E-07
	WP005	2.44E-07			2.59E-07
	WP010	1.54E-07		1.77E-07	
2	WP011			3.83E-08	4.95E-08
	WP014	2.79E-08			2.87E-08
	WP018			3.30E-07	3.20E-08
	WP021	1.05E-07		1.19E-07	
	WP023		1.21E-07	4.45E-08	
	WP001		5.43E-08	4.64E-08	
	WP003	7.01E-09			6.59E-09
	WP005	< 1.10E-08			1.53E-08
	WP010	< 9.06E-09		< 8.88E-09	
3	WP011			3.56E-08	3.52E-08
	WP014	5.56E-08			3.43E-08
	WP018			2.19E-07	4.81E-08
	WP021	1.63E-07		2.16E-07	
	WP023		1.78E-07	1.46E-07	
	WP001		1.06E-08	5.68E-08	
	WP003	4.01E-08			< 7.81E-09
	WP005	2.18E-08			2.87E-08
	WP010	8.63E-08		6.10E-08	
4	WP011			2.36E-07	1.86E-07
	WP014	5.61E-08			3.32E-08
	WP018			2.37E-07	7.47E-08
	WP021	1.64E-07		1.94E-07	
	WP023		2.43E-07	1.11E-07	
	WP001		6.45E-08	5.36E-08	
	WP003	4.06E-08			3.11E-08
	WP005	1.77E-07			2.73E-07
_	WP010	5.61E-08		5.25E-08	1.507.00
5	WP011			1.97E-09	1.58E-09
	WP014	5.99E-08		4.045.05	3.14E-08
	WP018			1.91E-07	5.26E-08
	WP021	1.51E-07		2.77E-07	
	WP023		1.96E-07	9.99E-08	

Table C-27. Effective Diffusivity  $(D_{e,n}^{Na})$  for NO₂⁻ [cm²/s]

Time Interval n	Saltstone Sample ID	Vial 1	Vial 2	Vial 3	Vial 4
6	WP001		5.40E-08	5.25E-08	
	WP003	6.12E-08			3.93E-08
	WP005	1.91E-07			1.58E-07
	WP010	7.72E-08		6.45E-08	
	WP011			1.34E-07	1.38E-07
	WP014	6.01E-08			5.20E-08
	WP018			2.39E-07	5.19E-08
	WP021	1.71E-07		2.97E-07	
	WP023		2.56E-07	1.22E-07	
7	WP001		7.13E-08	4.41E-08	
	WP003	6.46E-08			3.88E-08
	WP005	1.33E-07			1.38E-07
	WP010	4.63E-08		6.42E-08	
	WP011	·		1.74E-07	1.59E-07
	WP014	5.51E-08			2.92E-08
	WP018			1.31E-07	5.34E-08
	WP021	1.14E-07		2.35E-07	
	WP023		1.21E-07	1.21E-07	

Table C-27. Effective Diffusivity  $(D_{e,n}^{Na})$  for NO₂⁻ [cm²/s]

Now that the effective diffusivities for the Cr, Na,  $NO_3^-$ , and  $NO_2^-$  species for the various leaching time intervals  $(D_{e,n}^i)$  for the Saltstone samples have been defined, the Leachability Index of each species  $(L_i)$  can be calculated using Equation (3-2).

## Distribution:

S. L. Marra	773-A		
S. D. Fink	773-A		
K. M. Fox	999-W		
B. J. Giddings	786-5A		
C. C. Herman	999-W		
F. M. Pennebaker	773-42A		
P. M. Almond	773-43A		
A. D. Cozzi	999-W		
R. E. Eibling	999-W		
E. K. Hansen	999-W		
C. A. Langton	773-43A		
D. H. Miller	999-W		
B. R. Pickenheim	999-W		
M. M. Reigel	999-W		
M. G. Serrato	773-42A		
D. B. Stefanko	773-43A		
P. R. Jackson	703-46A		
K. H. Subramanian	249-8H		
J. M. Bricker	704-27S		
K. D. Dixon	704-14Z		
J. N. Leita	704-Z		
K. R. Liner	704-S		
P. D. Mason	704-14Z		
P. W. Norris	704-29S		
J.E. Occhipinti	704-S		
J. W. Ray	704-S		
S. C. Shah	704-14Z		
D. C. Sherburne	704-S		
A. V. Staub	704-27S		
J. R. Tihey	704-Z		
H. H. Burns	773-41A		
T. W. Coffield	705-1C		
D. A. Crowley	773-43A		
R. D. Freeman	705-1C		
F. M. Smith	705-1C		
K. H. Rosenberger	705-1C		