

Supplemental Immobilization of Hanford Low-Activity Waste: Cast Stone Screening Tests

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September 2013

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This document was prepared in conjunction with work accomplished under two Contracts No. DE-AC09-08SR22470 and DE-AC05-76RL01830 both with the U.S. Department of Energy.

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Printed in the United States of America

**Prepared for
U.S. Department of Energy**

ACKNOWLEDGMENTS

The work reported here was funded by Washington River Protection Solutions (WRPS). Dave Swanberg with WRPS led the effort and provided both programmatic guidance and technical input to the project team.

These screening tests were very laboratory intensive. At Pacific Northwest National Laboratory (PNNL), Ben Williams was instrumental in preparing the Cast Stone specimens and conducting the leach tests. Don Rinehart prepared the simulants. Keith Geiszler, Steve Baum, Igor Kutnyakov, Christian Iovin, and Denness Smith analyzed the many samples. Stan Pitman, Mike Dahl, and Karl Mattlin conducted the compressive strength measurements. At Savannah River National Laboratory (SRNL), Vickie Williams was the cornerstone of the preparation and measurement of the Cast Stone fresh properties, Kim Wyszynski and Vickie Williams prepared the simulants, and David Best, Whitney Riley, and Beverly Wall performed the analyses.

John Harris with LaFarge graciously provided the dry blend ingredients sourced from the northwest.

Jeff Serne at PNNL and Kevin Fox at SRNL provided independent technical review of the document. Dave MacPherson from PNNL; Paul Cavanah, Dave Darling, QD Ho, Felix Miera, Gene Ramsey, Rose Russell, and Leo Thompson from WRPS; and Fred Mann also provided comments on the document. Ellen Baer and Paul Eslinger from PNNL and Kevin Fox from SRNL provided calculation and document reviews.

Susan Ennor had the challenge of editing the inputs from multiple authors into a cohesive document. Kathy Neiderhiser did an outstanding job in preparing the document for publication.

EXECUTIVE SUMMARY

The Hanford Site has approximately 56 million gallons of radioactive waste stored in 177 underground storage tanks. The Hanford Waste Treatment and Immobilization Plant (WTP) is being constructed to treat all of the high-level waste (HLW) in the tank farms but will not have sufficient capacity to treat all of the low-activity waste (LAW) within the anticipated time frame for completion of the waste treatment mission. The LAW vitrification facility will need to be supplemented with a second LAW vitrification facility or an alternate LAW immobilization technology. A cementitious waste form known as Cast Stone is being considered to provide the required additional LAW immobilization capacity.

Hanford Tri-Party Agreement (TPA) milestone M-062-40ZZ requires the U.S. Department of Energy Office of River Protection (DOE/ORP) to submit a One-Time Hanford Tank Waste Supplemental Treatment Technologies Report by October 2014 if a technology other than a second LAW vitrification facility is proposed. The main purpose of the one-time report will be to describe the additional treatment facilities and technologies needed to treat all of Hanford's LAW. The report must describe the technologies considered, quantity of waste to be processed, quantity of final waste forms, secondary wastes, waste form performance data, technical viability, and life-cycle cost and schedule estimates. The DOE/ORP and the Washington State Department of Ecology would then have six months (until April 2015) to complete negotiations to select a final technology, and to establish milestones to implement the technology.

To support completion of the one-time report, Washington River Protection Solutions (WRPS) has contracted with Pacific Northwest National Laboratory (PNNL) and Savannah River National Laboratory (SRNL) to conduct a testing program with the following objectives:

- Determine an acceptable formulation for the LAW Cast Stone waste form.
- Evaluate the impact of different sources of dry materials for preparing the LAW Cast Stone.
- Demonstrate the robustness of the waste form for a range of LAW compositions.
- Demonstrate the robustness of the formulation for variability in the Cast Stone process.
- Provide Cast Stone contaminant release data for performance assessment and risk assessment evaluations.

The first step in determining an acceptable formulation for the LAW Cast Stone waste form was to conduct screening tests to evaluate the impact of key parameters including expected ranges in waste composition, waste stream concentrations, sources of dry materials, and mix ratios of waste (free water) to dry blend. A statistically designed test matrix was used to evaluate the effects of these key parameters on the properties of the Cast Stone as it was initially prepared and after curing. This report documents the results of these screening tests.

The screening tests comprise 26 individual Cast Stone mixes selected through the statistical design plus 12 additional tests described subsequently. The statistical experimental design included two blocks. The first block of 12 mixes plus 2 replicates was selected to estimate the main effects among the study parameters. The second block of 10 mixes plus 2 replicates

was selected to estimate the effects of some two-parameter interactions. There was one mix in Block 1 that was replicated in Block 2. The specific parameters included the following:

- Waste simulant composition
 - A single-shell tank (SST) blend based on the analyses of saltcake from six SSTs
 - An overall average LAW feed composition based on Hanford Tank Waste Operations Simulator (HTWOS) flowsheet modeling
 - A high-aluminum simulant based on HTWOS flowsheet modeling
 - A high-sulfate simulant based on HTWOS flowsheet modeling.
- Waste concentration expressed in terms of the sodium concentration in the wastes (5.0M and 7.8M Na)
- Sources of dry materials for Cast Stone dry blend, also known as premix
 - Class F fly ash source
 - Fly ash from the Pacific Northwest – relatively high in calcium
 - Fly ash from the Southeast – relatively low in calcium
 - Blast furnace slag
 - Pacific Northwest source
 - Southeast source
 - Ordinary portland cement from the Pacific Northwest
- Mix ratio, which is the ratio of free water in the waste to the mass of dry blend in the mix. The mix ratios included 0.4 (based on previous Cast Stone secondary waste studies) and 0.6 (based on current processing experience with the Saltstone Processing Facility at the Savannah River Site).

The ratio of cement to fly ash to blast furnace slag was held constant at the nominal Cast Stone mix ratio of 8:45:47 for all 26 mixes. The different combinations of simulants, Na concentrations, and mix ratios yielded waste loadings ranging from 9.5 wt% to 20.3 wt% total waste solids in the Cast Stone waste form.

Each of the 26 mixes was characterized with respect to the processing properties of the wet Cast Stone slurry as it was mixed and cured, and the properties of the final Cast Stone cured waste form. Processing properties that were measured included gel and set times, slurry rheology, flow cone, fresh density, and heat generation. Processing properties were measured for 12 mixes in addition to the 26 of the screening matrix. Final waste form properties that were measured included compressive strength, porosity, cured density, Toxicity Characteristic Leaching Procedure (TCLP) leach test, and EPA Draft Method 1315 leach test.

For the processing properties, the water-to-dry-blend mix ratio was the most significant parameter in affecting the range of values observed for each property. Table ES-1 shows the properties and their ranges at the 0.4 and 0.6 mix ratios. The method to process and dispose of the Cast Stone waste form has not yet been determined, so the function of the processing properties is to provide a potential range of property values for design input.

Table ES-1. Cast Stone Properties and Ranges at Mix Ratios

Property	0.4 Mix Ratio	0.6 Mix Ratio
Flow Diameter	94–160 mm	193–298 mm
Gel Time	1–15 minutes	12–121 minutes
Plastic Viscosity	220–580 centipoise	54–165 centipoise
Bingham Plastic Yield Stress	10–92 Pascals	2–24 Pascals
Heat Generation	150–338 J/g	270–434 J/g
Time to Peak Heat Generation	26–164 hours	16–101 hours
Fresh Density	1.87–1.96 g/cm ³	1.73–1.83 g/cm ³

Compressive strengths were measured in triplicate on Cast Stone monoliths prepared at PNNL and SRNL. Compressive strengths ranged from 5.8 to 62.0 MPa (850 to 8990 psi), excluding one sample that broke at 1.3 MPa (190 psi). The target minimum compressive strength is 3.4 MPa (500 psi), which is more than sufficient to withstand the overburden in a near-surface disposal facility, e.g., the draft waste acceptance criteria for the Hanford Integrated Disposal Facility (IDF) requires a minimum comprehensive strength of 586 kPa, or 85 psi. The lower compressive strengths tended to be from Cast Stone mixes with the 7.8M Na simulants and the 0.6 water-to-dry-blend ratio. The higher compressive strengths tended to be from Cast Stone mixes with the 5M Na simulants and/or the 0.4 water-to-dry-blend mix ratio.

To be acceptable for disposal at the IDF on the Hanford Site, Cast Stone containing hazardous metals must pass the TCLP. Land disposal restrictions require that the concentration of the species in the leachate be below the Universal Treatment Standards (UTS) in Title 40 Code of Federal Regulations Part 268 “Land Disposal Restrictions” (40 CFR 268). The LAW is projected to include Resource Conservation and Recovery Act (RCRA) regulated metals including As, Ba, Cd, Cr, Pb, Hg, Se, and Ag; and underlying hazardous constituents (UHCs) including Sb, Be, Ni, and Tl. In addition, some of the dry materials used to make Cast Stone may include these same and other hazardous materials. The simulants in the screening tests were spiked with Cr, Pb, Ni, and Cd, which were the highest concentration RCRA metals projected in the HTWOS flowsheet modeling. The dry blend materials contributed As, Ba, and Se to the Cast Stone waste forms. All of the 26 Cast Stone mixes easily met the UTS limits based on the TCLP results and total constituent analyses calculations.

Leach tests on cured Cast Stone cylinders were conducted for 91 days using EPA Method 1315 to measure the effective diffusivity of key constituents of concern including Tc, I, U, Cr, Na, and nitrates and nitrites. Effective diffusivities are used in performance assessments and risk assessments to describe the release of contaminants from the waste form. Effective diffusivities for Na, I, nitrate, and nitrite averaged over the 28- to 91-day cumulative leach intervals were all in the same range of 1×10^{-8} to 2×10^{-9} cm²/s for all but one of the 26 Cast Stone mixes. The corresponding average leachability indices (LIs) were in the range of 8 to 8.7. Technetium effective diffusivities were in the range of 2×10^{-10} to 6×10^{-12} cm²/s (LI = 9.7 to 11.2) for the same 25 mixes and total leach interval. Chromium effective diffusivities were in the range of 9×10^{-13} to 8×10^{-15} cm²/s (LI = 12.0 to 14.1). Uranium was not detected in most of leachates from the 25 mixes, indicating that the U is retained in the Cast Stone and is being minimally released under the conditions of the EPA 1315 leach test.

Statistical analyses were performed for Cast Stone properties including plastic viscosity, heat generation, compressive strength, and EPA Method 1315 leach indices. The objectives of the statistical analyses were to determine whether individual test parameters and their two-parameter

interactions have statistically significant effects on a given Cast Stone property, and to assess the relative magnitudes of these effects. Two approaches to statistical analyses of the data were performed to address these objectives. The “Full Model” approach was used to evaluate the individual effects of the five test parameters plus two-parameter interactions as used in developing the screening test matrix. The “Stepwise Model” approach used stepwise regression to build a model to identify the statistically significant individual parameter and two-parameter interaction effects. Of the five parameters varied in the screening tests, the mix ratio, fly ash source, and blast furnace slag source had the most significant effects on the Cast Stone properties. Sodium molarity and the waste simulant type were less significant variables affecting the Cast Stone. All five of the test parameters were involved in one or more statistically significant two-parameter interactions over the Cast Stone properties that were statistically analyzed, indicating that interactive effects of the parameters must be considered in future work to select optimal Cast Stone formulations.

The screening tests demonstrated that, for the ranges of parameters studied, almost all of the formulations could be processed and would provide acceptable waste forms. Of the 26 original Cast Stone mixes in the screening test matrix, all but 2 mixes had acceptable processing and waste form properties. That is, the ranges of properties measured are similar enough that a processing/disposal facility could be designed to accommodate all but two of the test compositions evaluated in the screening tests.

All of the Cast Stone mixes had acceptable properties with respect to anticipated waste acceptance criteria for the IDF. Compressive strengths exceeded the 500 psi (3.45 MPa) target. The leachates from the TCLP test met treatment standards to address Land Disposal Restrictions for hazardous chemicals in 40 CFR 268.

The next step in the planned testing program is to optimize the Cast Stone formulation. The screening test results suggest that the waste loading in the Cast Stone can be increased beyond the levels evaluated in the screening tests. Waste loading can be increased by concentrating the wastes to higher sodium concentrations and by increasing the free water-to-dry blend mix ratio. It is reasonable to conduct additional formulation optimization testing with sodium molarities in the range of 7M to 10M and free water-to-dry blend mix ratios in the range of 0.5 to 0.7. Though there were statistically significant effects of the sources of BFS and fly ash, the impacts were not drivers to use sources other than those available in the Pacific Northwest for the Cast Stone optimization work.

Cast Stone formulation optimization provides the opportunity to investigate methods to improve the retention of Tc and I in the waste form. The use of getters, fillers to reduce porosity, and increased BFS content will be evaluated.

This work has provided a sound technical basis for the next phase of technology maturation. The primary objective for the next phase will be to develop a preferred formulation of a low-temperature waste form for Hanford LAW based on knowledge gained during this screening test phase. The preferred formulation is expected to include formulation enhancements to improve retention of constituents of concern. Other objectives for the next phase include conducting an engineering-scale demonstration of the Cast Stone preparation process and conducting confirmatory testing with samples of real waste.

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LIST OF ABBREVIATIONS AND ACRONYMS

Å	angstrom(s)
ANOVA	analysis of variance
ANS	American Nuclear Society
ANSI	American National Standards Institute
ASME	American Society of Mechanical Engineers
BET	Brunauer–Emmett–Teller
BFS	blast furnace slag
CAWSRP	Conducting Analytical Work in Support of Regulatory Programs
CBO	Carbon Burn Out
Ci	curie(s)
CLSM	controlled low strength material
cm	centimeter(s)
cm ²	square centimeter(s)
cm ³	cubic centimeter(s)
COC	constituent of concern
cP	centipoise
DIW	deionized water
DOE	U.S. Department of Energy
DOF	degree of freedom
DSC	differential scanning calorimetry
DST	double-shell tank
EDC	effective diffusion coefficient
EPA	U.S. Environmental Protection Agency
FA	fly ash
FY	fiscal year
µg	microgram(s)
g	gram(s)
hr	hour(s)
HASQARD	Hanford Analytical Services Quality Assurance Requirements Documents
HDI	How Do I...?
HG300	heat generation at 300 hours
HGP	heat generation at peak

HLW	high-level waste
HSD	(Tukey's) Honestly Significant Difference
HTWOS	Hanford Tank Waste Operations Simulator
IC	ion chromatography
ICDD	International Center for Diffraction Data
ICP	inductively coupled plasma
ICP-MS	inductively coupled plasma-mass spectroscopy or spectrometer
ICP-AES	inductively coupled plasma-atomic emission spectrometer
ICP-OES	inductively coupled plasma-optical emission spectroscopy
ID	inside diameter or identification
IDF	Integrated Disposal Facility
in.	inch(es)
J/g	joule(s) per gram
kips-force	kilo-pound force
kg	kilogram(s)
kN	kiloNewton(s)
kV	kilovolt(s)
L	liter(s)
LAW	low-activity waste
LI	leachability index
ln	natural logarithm
LOF	lack-of-fit
μm	micron(s)
m ²	square meter(s)
mA	milliampere(s)
MCP	multiple comparison procedure
mg	milligram(s)
min	minute(s)
ml	milliliter(s)
mm	millimeter(s)
mM	millimolar
M/M	moles per mole
MPa	megapascal(s)
MR	mix ratio
MS	mass spectroscopy

mW/g	milliwatt(s) per gram
NIST	National Institute of Standards and Technology
OD	outer diameter
OED	optimal experimental design
OLS	ordinary least squares
OPC	ordinary portland cement
ORP	Office of River Protection
Pa	pascal(s)
PA	performance assessment
PNNL	Pacific Northwest National Laboratory
ppm	parts per million
psi	pounds per square inch
QA	quality assurance
QAP	Quality Assurance Plan
%RSD	relative standard deviation
RCRA	Resource Conservation and Recovery Act
rph	rotations per hour
rpm	rotations per minute
RR	random run
s	second(s)
SD	standard deviation
SEM	scanning electron microscopy
SPF	Saltstone Production Facility
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
SST	single-shell tank
STA	simultaneous thermal analysis
SwRI	Southwest Research Institute
TCLP	Toxicity Characteristic Leaching Procedure
TGA	thermal gravimetric analysis
TPA	Tri-Party Agreement
TPHG	time to peak heat generation
TRU	transuranic
UHC	underlying hazardous constituent
UPV	ultrasonic pulse velocity

UTS	Universal Treatment Standards
V	volt(s)
WC	waste concentration
WRPS	Washington River Protection Solutions
wt%	weight percent
WWFTP	WRPS Waste Form Testing Program
WTP	Hanford Tank Waste Treatment and Immobilization Plant
XRD	X-ray diffraction
XRF	X-ray fluorescence

1.0 Introduction

More than 56 million gallons of radioactive and hazardous waste are stored in 177 underground storage tanks at the U.S. Department of Energy's (DOE's) Hanford Site in southeastern Washington State. The Hanford Tank Waste Treatment and Immobilization Plant (WTP) is being constructed to treat the wastes and immobilize them in a glass waste form. The WTP includes a pretreatment facility to separate the wastes into a small volume of high-level waste (HLW) containing most of the radioactivity and a larger volume of low-activity waste (LAW) containing most of the nonradioactive chemicals. The HLW will be converted to glass in the HLW vitrification facility for ultimate disposal at an offsite federal repository. At least a portion (~35%) of the LAW will be converted to glass in the LAW vitrification facility and will be disposed of onsite at the Integrated Disposal Facility (IDF). The pretreatment and HLW vitrification facilities will have the capacity to treat and immobilize the wastes destined for each facility. However, a second LAW immobilization facility will be needed for the expected volume of LAW requiring immobilization.

A cementitious waste form known as Cast Stone is being considered to provide the required additional LAW immobilization capacity. The Cast Stone waste form must be acceptable for disposal in the IDF. The Cast Stone waste form and immobilization process must be tested to demonstrate that the final Cast Stone waste form can comply with the waste acceptance criteria for the disposal facility and that the immobilization processes can be controlled to consistently provide an acceptable waste form product. Further, the waste form must be tested to provide the technical basis for understanding the long-term performance of the waste form in the disposal environment. These waste form performance data are needed to support risk assessment and performance assessment (PA) analyses of the long-term environmental impact of the waste disposal in the IDF. The PA is needed to satisfy both Washington State IDF Permit and DOE Order requirements.

Cast Stone has been selected for solidification of radioactive wastes including WTP aqueous secondary wastes treated at the Effluent Treatment Facility (ETF) at Hanford. A similar waste form called Saltstone is used at the Savannah River Site (SRS) to solidify its LAW tank wastes.

1.1 Cast Stone Testing Program

A testing program was developed in fiscal year (FY) 2012 describing in detail the work needed to develop and qualify Cast Stone as a waste form for the solidification of Hanford LAW (Westsik et al. 2013). Included in the testing plan by Westsik et al. (2013) is a section on the near-term needs to address Tri-Party Agreement (TPA) Milestone M-062-40ZZ. The objectives of the testing program to be conducted in FY 2013 and FY 2014 are as follows:

- Determine an acceptable formulation for the LAW Cast Stone waste form.
- Evaluate sources of dry materials for preparing the LAW Cast Stone.
- Demonstrate the robustness of the waste form for a range of LAW compositions.
- Demonstrate the robustness of the formulation for variability in the Cast Stone process.
- Provide Cast Stone contaminant release data for PA and risk assessment evaluations.

The first step in determining an acceptable formulation for the LAW Cast Stone waste form is to conduct screening tests to evaluate the impact of key parameters including ranges in waste composition, waste stream concentrations, sources of dry materials, and mix ratios of waste (free

water basis) to dry blend. A statistically designed test matrix was used to evaluate the effects of these key parameters on the properties of the Cast Stone as it is initially prepared and after curing.

The next phase of testing will focus on final selection of a nominal Cast Stone formulation and demonstration that Cast Stone can meet anticipated waste form requirements for disposal in the IDF. This testing is expected to use the results of the screening tests to specify a suite of tests to optimize the waste loading and waste form performance of the Cast Stone formulation.

Preparation and testing of selected LAW Cast Stone formulation(s) made with actual radioactive wastes will be conducted to provide confirmation that the results observed with LAW waste simulants in screening, waste loading, and waste form qualification testing are representative of what is expected with actual wastes.

After a preferred formulation is selected for immobilization of Hanford LAW, an engineering-scale demonstration of the Cast Stone process and containerized waste form may be conducted in FY 2014 using nonradioactive waste simulants.

1.2 Tank Waste Processing

The Cast Stone process is being evaluated to treat and immobilize LAW separated from the radioactive waste currently stored in underground storage tanks at Hanford. The LAW fraction of the waste is characterized as a large-volume, low-radioactivity liquid process stream stripped of most of the solids, transuranic (TRU) elements, and long-lived radionuclides. The LAW is derived from the aqueous solutions in the tanks and dissolved saltcake. The composition of the LAW will vary from tank to tank because of the variability in types and sources of wastes stored in the individual tanks and the processes used to separate the wastes into HLW and LAW fractions.

The baseline source of the LAW wastes to be treated and immobilized through the Cast Stone process is the separations processes in the WTP pretreatment facility. The liquid wastes from the tanks will pass through ultrafilters in the pretreatment facility to remove solids and insoluble radioisotopes. For some tank wastes, additional processing as part of the ultrafiltration process will remove aluminum and/or chromium from the solids destined for HLW, and the Al and Cr will be added to the LAW stream. Some tanks contain organic complexants that keep strontium and TRU elements in the aqueous phase. The treatment of these wastes will include a precipitation step to remove the Sr and TRU elements from solution prior to filtration and ion exchange. For LAW wastes with higher concentrations of complexants and other organics, it may be decided to send these wastes to the LAW vitrification facility for treatment, which would destroy the complexants and organics in the vitrification process.

The liquid filtrate from the ultrafilters will pass through ion-exchange columns to remove cesium. The effluent from the ion-exchange columns will be then concentrated in an evaporator to a specified sodium concentration depending on the waste chemistry. A recycle stream from the LAW melter off-gas submerged bed scrubbers and wet electrostatic precipitators will be blended with the ion-exchange effluent as it enters the evaporator. The concentrated solution from the evaporator will be the LAW feed for the immobilization process. The waste feed will be principally Na with nitrates, nitrites, carbonates and a spectrum of radioactive fission products. Minor components can include Al, K, and Si along with the chloride, fluoride, phosphate, and sulfate anions.

To supplement WTP capacity, in-tank and near-tank separations are being considered to provide LAW feed directly to the supplemental immobilization process without processing through the WTP pretreatment facility. These processes would include a filtration step via a cross-flow filter or a rotary microfilter to remove the solids and insoluble radionuclides. Cesium would be removed through ion exchange. The separated solids and cesium would be returned to the double-shell tank (DST) system for eventual treatment and immobilization as HLW. A process for removing Tc from the LAW to be immobilized in Cast Stone is also being evaluated. The liquid effluent from the solids, Cs and Tc separation processes would become the feed to the LAW Cast Stone immobilization process.

Some tank wastes may be acceptable for direct Cast Stone processing without additional separations. These low-curie salt solutions are generated during the later stages of washing saltcake from the tanks. Experience has shown that the cesium is removed in the earlier stages of washing the saltcake such that the saltcake dissolved later is relatively free of ¹³⁷Cs and could be immobilized without further processing. Solid separation would be required if >1 wt% solids were present. The decontamination factors have been observed in saltcake dissolution and fractional crystallization work done by PNNL and 222-S.

Therefore, the composition of LAW to be processed as Cast Stone will vary greatly, requiring testing to look at several compositions of feed. Four different chemical simulant compositions were investigated in the screening tests to assess a range of different waste compositions that may be sent to the LAW Cast Stone immobilization process.

1.3 Report Contents and Organization

This report summarizes the results of the screening tests conducted as the first step in determining an acceptable Cast Stone formulation for solidification of Hanford LAW. The report also summarizes the selection and development of the chemical simulants that were used in the screening tests.

The balance of the report is organized as follows. Section 2.0 describes the approach to the Cast Stone screening tests including the statistically designed test matrix and quality assurance. Section 3.0 describes the LAW simulants used to make the Cast Stone specimens, and Section 4.0 describes the sources of the blast furnace slag, fly ash, and cement used to make the Cast Stone specimens. Section 5.0 describes the preparation of the Cast Stone specimens for the screening tests. The next sections present the results of the characterization of the Cast Stone processing properties (Section 6.0) including gel and set times, slurry rheology, and heat of hydration; and final waste form properties (Section 7.0) including compressive strength, cured density, Toxicity Characteristic Leaching Procedure (TCLP) results, and leach test results. Statistical analyses of selected Cast Stone properties are presented in Section 8.0. Section 9.0 provides conclusions from the screening test work, and Section 10.0 provides recommendations for the next phase of development. Appendices contain additional information including the statistical design of the screening test matrix (Appendix A), dry blend materials characterization data (Appendix B), Cast Stone processing properties data (Appendix C), cured Cast Stone properties data (Appendix D), data-interaction plots for selected Cast Stone properties (Appendix E), and vendor material certification reports (Appendix F).

2.0 Screening Tests Approach

Screening tests were performed to evaluate the effects of key parameters on the properties of the Cast Stone as it is initially prepared and after curing. The test parameters and their ranges that were investigated in the screening tests included

- simulants representing a range of LAW compositions (Average, single-shell tank [SST] Blend, High Al, and High SO₄ [discussed in Section 3.0])
- waste concentration (5 M and 7.8 M Na)
- Class F fly ash source (NW = High Ca, SE = Low Ca)
- blast furnace slag source (NW, SE)
- free-water-to-dry-blend solids mix ratio (0.4, 0.6).

The four simulants are discussed in Section 3.0. The Na concentrations of 5 M and 7.8 M were selected to represent a range of possible waste concentrations for processing. The Class F fly ash included a relatively high-Ca content material available in the Pacific Northwest (designated NW) and a lower Ca content material available in the southeastern (designated SE) part of the country. Blast furnace slags (BFSs) from the northwest (designated NW) and southeast (designated SE) were also selected for the screening tests. The 0.4 and 0.6 values of the free-water-to-dry-blend solids mix ratio (henceforth referred to as “mix ratio” for convenience) were selected based on the range (0.35 to 0.41 grams of water per gram dry blend solids) used for the secondary waste Cast Stone formulation work (Mattigod et al. 2011) and 0.60 grams of water per gram of dry blend solids used at the Saltstone Production Facility at the SRS. The different combinations of simulants, Na concentrations, and mix ratios yielded waste loadings ranging from 9.5 wt% to 20.3 wt% total waste solids in the Cast Stone waste form.

The dry blend mix ratio was held constant at 8 wt% portland Type I/II cement, 45 wt% Class F fly ash, and 47 wt% Grade 100-120 BFS (Lockrem 2005). A single source of portland cement was used for all of the testing because the variability in the cement is not expected to be significant among the possible sources.

2.1 Screening Test Matrix

The original screening test matrix was developed in two “blocks” in case project funding might be limited and only the Block 1 tests could be performed. However, sufficient funding was provided, so both Block 1 and Block 2 of the experimental design were performed. Appendix A discusses the details of the how both blocks of the test matrix were developed.

The original test matrix for the screening tests is composed of 26 test conditions as shown in Table 2-1. The matrix was developed using statistical optimal experimental design (OED) methods and software (as described in detail in Appendix A). The test matrix includes two blocks of tests. The first block is composed of 14 tests including 12 distinct parameter combinations and 2 replicates chosen using OED to assess the effects of the individual test parameters. The second block of tests is composed of 12 tests including 10 distinct parameter combinations and 2 replicates chosen by OED to augment the first block of tests to assess the block effect and 9 selected two-parameter interactions, along with the individual parameter effects (see Appendix A). A fifth replicate pair includes one test in Block 1 and one test in

Table 2-1. Screening Test Matrix

Mix # ^(a)	Random Run Order ^(b)	Block	Simulant	Sodium Molarity	Portland Cement	Fly Ash Class F	Blast Furnace Slag	Dry Blend Mix ^(c)	Water-to-Dry Blend Solids Ratio
1	7	1	High SO ₄	5.0	Type I/II	SE Low Ca	NW	8:45:47	0.40
2	@	1	Average	5.0	Type I/II	SE Low Ca	SE	8:45:47	0.60
3	\$	1	Average	7.8	Type I/II	NW High Ca	SE	8:45:47	0.60
4		1	High Al	5.0	Type I/II	NW High Ca	SE	8:45:47	0.40
5		1	Average	7.8	Type I/II	NW High Ca	NW	8:45:47	0.40
6		1	Average	7.8	Type I/II	SE Low Ca	NW	8:45:47	0.40
7	#	1	High SO ₄	7.8	Type I/II	SE Low Ca	SE	8:45:47	0.40
8		1	SST Blend	5.0	Type I/II	SE Low Ca	NW	8:45:47	0.40
9		1	Hi Al	7.8	Type I/II	NW High Ca	SE	8:45:47	0.60
10		1	High SO ₄	5.0	Type I/II	NW High Ca	NW	8:45:47	0.60
11		1	High Al	7.8	Type I/II	SE Low Ca	NW	8:45:47	0.60
12		1	SST Blend	5.0	Type I/II	NW High Ca	SE	8:45:47	0.60
13	@	1	Average	5.0	Type I/II	SE Low Ca	SE	8:45:47	0.60
14	#	1	High SO ₄	7.8	Type I/II	SE Low Ca	SE	8:45:47	0.40
15	%	2	High SO ₄	7.8	Type I/II	NW High Ca	NW	8:45:47	0.40
16		2	SST Blend	7.8	Type I/II	NW High Ca	NW	8:45:47	0.40
17		2	High Al	5.0	Type I/II	SE Low Ca	SE	8:45:47	0.60
18		2	SST Blend	7.8	Type I/II	SE Low Ca	SE	8:45:47	0.40
19		2	High Al	7.8	Type I/II	SE Low Ca	NW	8:45:47	0.40
20		2	Average	5.0	Type I/II	NW High Ca	SE	8:45:47	0.40
21	&	2	High SO ₄	7.8	Type I/II	SE Low Ca	NW	8:45:47	0.60
22	\$	2	Average	7.8	Type I/II	NW High Ca	SE	8:45:47	0.60
23		2	SST Blend	7.8	Type I/II	SE Low Ca	NW	8:45:47	0.60
24		2	High Al	5.0	Type I/II	NW High Ca	NW	8:45:47	0.60
25	%	2	High SO ₄	7.8	Type I/II	NW High Ca	NW	8:45:47	0.40
26	&	2	High SO ₄	7.8	Type I/II	SE Low Ca	NW	8:45:47	0.60
(a) Mix numbers with the same symbols are replicates.									
(b) The run order was randomized separately within Block 1 and within Block 2.									
(c) 8 wt% cement: 45 wt% fly ash, 47 wt% blast furnace slag.									

Block 2. The second column of Table 2-1 identifies the five pairs of replicate tests. The replicate tests provide for quantifying the experimental and measurement uncertainties in performing tests and measuring properties. These estimates of uncertainties also provide for statistically assessing the significance of individual parameter and two-parameter interaction effects. The order in which the test specimens were prepared within each block was based on a random run order as shown in Table 2-1. An additional 12 mixes were prepared that were not included in the test matrix in Table 2-1. These are discussed further in Section 6.0.

2.2 Cast Stone Properties

Cast Stone specimens for the screening tests were prepared in the laboratory and were characterized for processing properties and cured waste form properties. Processing properties include gel time (a subjective indication of when the Cast Stone slurry no longer flows freely), hardening time, slurry rheology, heat generation during curing, and residual free liquids. These processing properties were measured by Savannah River National Laboratory (SRNL) under a separate contract with Washington River Protection Solutions (WRPS).

Other properties of the solidified Cast Stone waste forms were measured after the test specimens cured for at least 28 days at room temperature in near-saturated conditions. Compressive strength was measured on Cast Stone waste forms made at both SRNL and PNNL; density and porosity were measured on the cured test specimens prepared at SRNL. The TCLP was conducted on Cast Stone prepared at PNNL to examine the retention of Resource Conservation and Recovery Act (RCRA) metals (As, Ba, Cd, Cr, Pb, Hg, Se, and Ag) and underlying hazardous constituents (Sb, Be, Ni, and Tl) to address land disposal restrictions in Title 40 *Code of Federal Regulations* Part 268 “Land Disposal Restrictions” (40 CFR 268). Leach tests on cured Cast Stone cylinders prepared at PNNL were conducted for up to 91 days using U.S. Environmental Protection Agency (EPA) Draft Method 1315 (2012) to measure the effective diffusivity of key constituents including Tc and nitrates. Leach testing beyond 91 days is being continued on selected samples under a separate PNNL project that is providing technical data and support for preparation of a risk assessment and future, more detailed PAs. Chemical compositions and mineralogy were also determined for the cured Cast Stone specimens.

Statistical analysis of the results of the testing and characterization at PNNL and SRNL were used to assess the effects on selected Cast Stone properties of the individual test parameters and evaluate two-parameter interactions (see Section 8.0).

2.3 Quality Assurance

The PNNL Quality Assurance (QA) Program is based upon requirements defined in DOE Order 414.1D, *Quality Assurance* (DOE 2011), and Title 10 *Code of Federal Regulations*, Part 830, “Nuclear Safety Management”, Subpart A – Quality Assurance Requirements (a.k.a. the Quality Rule) (10 CFR 830). PNNL has chosen to implement the following American Society of Mechanical Engineers (ASME) consensus standards in a graded approach:

- ASME NQA-1-2000, *Quality Assurance Requirements for Nuclear Facility Applications*, Part I, “Requirements for Quality Assurance Programs for Nuclear Facilities.”
- ASME NQA-1-2000, Part II, Subpart 2.7, “Quality Assurance Requirements for Computer Software for Nuclear Facility Applications,” including problem reporting and corrective action.

- ASME NQA-1-2000, Part IV, Subpart 4.2, “Guidance on Graded Application of Quality Assurance (QA) for Nuclear-Related Research and Development.”

The processes necessary to implement the requirements are documented through PNNL’s “How Do I...? (HDI) standards-based management system.

PNNL implements the WRPS Waste Form Testing Program (WWFTP) quality requirements by performing work in accordance with the WWFTP Quality Assurance Plan (QA-WWFTP-001).¹ Work is performed to the quality requirements of the NQA-1-2000 Quality Assurance Program, graded on the approach presented in NQA-1-2000, Part IV, Subpart 4.2. Data analysis is performed in accordance with the *Hanford Analytical Services Quality Assurance Requirements Documents* (HASQARD; DOE/RL-96-68 2007). The PNNL document for implementing HASQARD is *Conducting Analytical Work in Support of Regulatory Programs* (CAWSRP).²

The WWFTP addresses internal verification and validation activities by conducting an independent technical review of the final data report in accordance with WWFTP procedure QA-NSLW-0603, *Independent Technical Review*. Following this procedure, a technical review verifies that the reported results are traceable, that inferences and conclusions are soundly based, and that the reported work satisfies the objectives.

The SRNL work scope was performed in accordance with a Quality Assurance Program (QAP) that meets the Quality Assurance criteria specified in DOE Order 414.1D, *Quality Assurance*; 10 CFR 830, “Nuclear Safety Management,” Subpart A, “Quality Assurance Requirements,” paragraph 830.122; and also meets the requirements of ASME NQA-1-2004, *Quality Assurance Requirements for Nuclear Facility Applications*, including NQA-1a-2005 and NQA-1b-2007 Addenda, or later version. The SRNL work scope was performed in accordance with Savannah River Site Manual 1Q, QAP 2-3 (Control of Research and Development Activities).

WRPS has conducted an evaluation of the PNNL and SRNL QA programs and both laboratories are on their evaluated supplier list.

Certain tables and figures in this report are marked “For Information Only” because the PNNL QA procedures for calculations were not applied. The tables and figures so marked do not contain key, “quality affecting” results. The software codes used to create such tables and figures were carefully checked by the researcher, but to save considerable time and expense, were not included in the formal QA package for statistical analyses. Only the software and calculations included in the formal QA package were checked by an independent reviewer, per the applicable PNNL QA procedure.

¹ MacPherson, DB. 2012 as amended. *WRPS Waste Form Testing Program Quality Assurance Plan*. Pacific Northwest National Laboratory, Richland, Washington.

² CAWSRP – *Conducting Analytical Work in Support of Regulatory Programs*. 2006, as amended. Pacific Northwest National Laboratory, Richland, Washington.

3.0 Selection of Simulants

For the screening tests of the Cast Stone formulations for LAW, four chemical simulants were selected to represent a range of possible LAW compositions to be solidified in the Cast Stone waste form. They included a saltcake simulant used in previous testing of LAW immobilization technologies and three chemical simulants based on the Hanford Tank Waste Operations Simulator (HTWOS) flowsheet modeling of the LAW feed that is anticipated to be sent to a supplemental immobilization facility. A detailed description of the simulant development work is provided by Russell et al. (2013).

3.1 Saltcake Simulant

Saltcake waste is a predominant form of the wastes in a large fraction of the SSTs at Hanford. Saltcake wastes can be dissolved, stripped of cesium, and converted to glass or an alternative waste form such as Cast Stone for disposal. To support an evaluation of supplemental treatment alternatives for immobilizing this type of LAW, a saltcake simulant waste was developed based on a blend of real waste samples from SSTs S-101, S-109, S-110, S-111, U-106, and U-109 (Rassat et al. 2003). Table 3-1 shows the nominal simulant composition, which is referred to in this report as the “SST Blend” simulant. A recipe for preparing the simulant is provided by Rassat et al. (2003). This saltcake simulant has been used in previous Cast Stone testing with Hanford LAW simulants. The composition matches the average composition of saltcake from 68 Hanford SSTs representing 85 percent of the total saltcake inventory in all Hanford SSTs and DSTs at that time (Gasper et al. 2002).

Table 3-1. Composition of the Nonradioactive Dissolved Saltcake Solution for the SST Blend Simulant (Rassat et al. 2003)

Waste Constituent	Concentration (M)
Al	0.0637
Cs	5.1×10^{-8}
Cr	0.0104
K	0.0124
Na	5.00
Cl	0.0438
CO ₃	0.475
F	0.0316
NO ₂	0.424
NO ₃	2.51
PO ₄	0.0492
SO ₄	0.0900
C ₂ O ₄ (oxalate)	0.0118
Other TOCs ^(a) (as carbon, from acetate)	0.263
TOC Total	0.287
OH Total	0.740
Free OH	0.485
(a) TOC = total organic carbon.	

3.2 Simulants Based on System Plan 6 HTWOS Modeling

Three chemical simulants were developed to represent the range of LAW tank wastes that could be immobilized in a Cast Stone waste form. The chemical simulants were developed based on

runs of the HTWOS model to support the *River Protection Project System Plan*, Revision 6 (Certa et al. 2011). The HTWOS model is used to track the tank waste as it moves from storage through retrieval, feed staging, and multiple treatment and immobilization processes over the life of the WTP mission. The HTWOS model can be used to forecast the quantities and compositions of primary and secondary waste streams as a result of various proposed operating scenarios (Certa et al. 2011). As one of the outputs, the HTWOS model provides the projected compositions of LAW feed to a supplemental immobilization facility over the course of the tank waste cleanup mission.

The first chemical simulant is an overall average of the 1046 weeks of modeled LAW immobilization waste feed over a 20-year mission. Henceforth in this report, it is referred to as the “Average” simulant.

The second chemical simulant is a high-Al, high-Cl simulant corresponding to week 235 in the HTWOS flowsheet simulation. Henceforth in this report it is referred to as the “High Al” simulant. It corresponds to the maximum Al concentration normalized to Na. Cl is near its maximum as well. Unlike SO_4^{2-} and F^- , Al does not have a single spike at a high concentration but a broad region between 0.08 and 0.1 mole/mole (M/M) Na. Figure 3-1 shows the concentrations of Al (as $\text{Al}(\text{OH})_4^-$) and Cl^- for the 1046 weeks modeled in the HTWOS simulations. The point at week 235 is circled in green.

The third chemical simulant, referred to as the “High SO_4 ” simulant, was selected because sulfate and fluoride were at relatively high concentrations relative to sodium and phosphate was near its maximum. This corresponds to week 672 in the HTWOS flowsheet simulation. Figure 3-2 shows the concentrations of SO_4^{2-} , PO_4^{3-} , and F^- for the selected point circled in red. This week was selected over the later times near the end of the mission that are predicted to have even higher spikes in sulfate and fluoride concentrations because the sources of these late spikes are not immediately clear, are due in part to low sodium concentrations, and are not representative of most of the WTP mission duration. Also, the HTWOS does not use thermodynamic models to predict solubilities. For example, it is unlikely that such high levels of phosphate and fluoride could be maintained in solution at the high pH and ionic strength of these wastes.

Before the simulant batches were prepared for the Cast Stone waste form screening tests, smaller 1-L batches were prepared to check for chemical interactions and solids formation (Russell et al. 2013). The 5 M Na concentration is the expected concentration for processing the LAW through ultrafiltration and ion exchange. The SST Blend saltcake and High Al simulant had only minimal solids at 5 M Na. The overall Average and High SO_4 simulant had significant solids formation at 5 M Na. These solids were determined to contain Na fluorophosphates similar to what had been observed earlier when the SST Blend saltcake simulant was initially developed. Therefore, the fluoride and phosphate levels were reduced in the overall Average and High SO_4 simulants to levels that would result in little to no solids formation at the 5 M Na concentration. All of the simulants had solids formation at the 7.8 M Na concentration. Based on supernate analyses, all four 7.8 M Na simulants appear to have precipitated Na fluorophosphate solids, estimated at 1 to 3 weight percent. These solids were left in the final simulant batches used to make the Cast Stone waste forms used in the screening tests assuming that during production, pretreated LAW would be evaporated without subsequent filtration prior to immobilization in Cast Stone.

Table 3-2 shows the compositions of three chemical simulants (Average, High Al, and High SO_4) as well as the SST Blend saltcake simulant described in Section 3.1. For comparison purposes,

the simulants have been normalized to one-molar Na and are expressed as moles per mole Na. The target concentrations have been charge balanced by adjusting the anion levels in proportion to their relative concentrations.

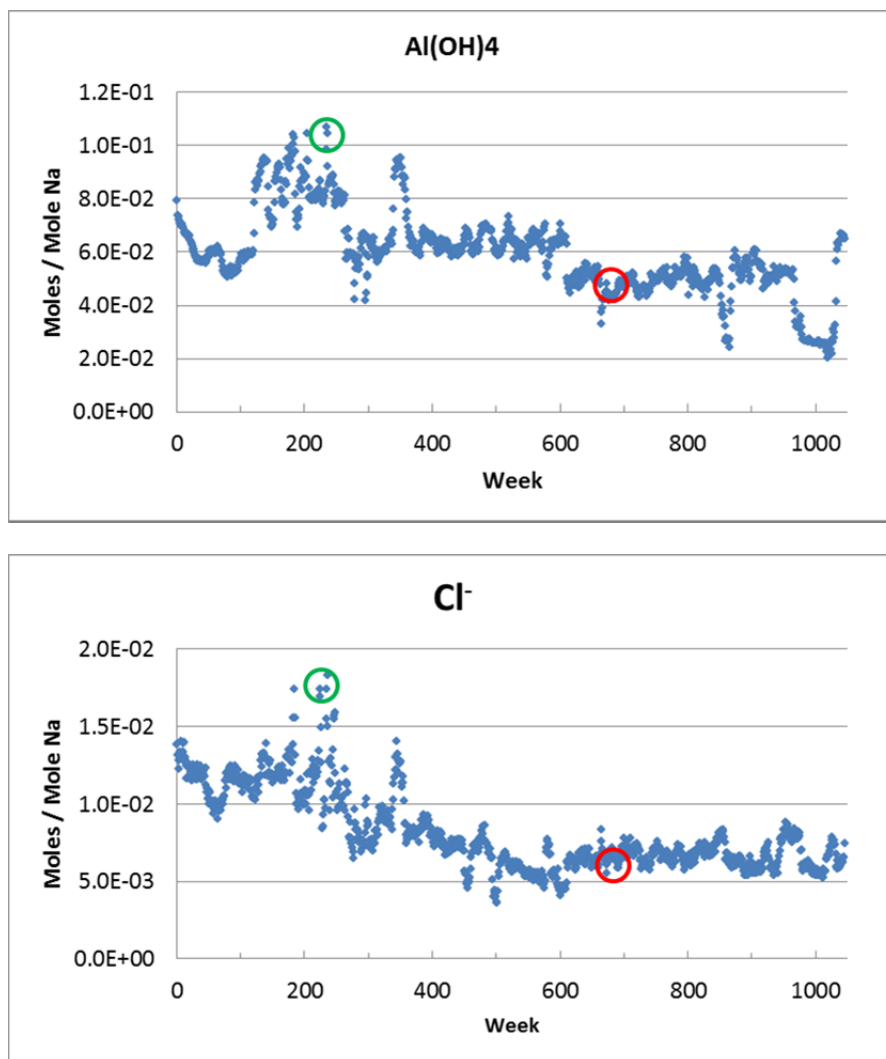


Figure 3-1. Aluminum and Chloride Concentrations from HTWOS Flowsheet Simulation.
Week 235 in green circle, week 672 in red circle.

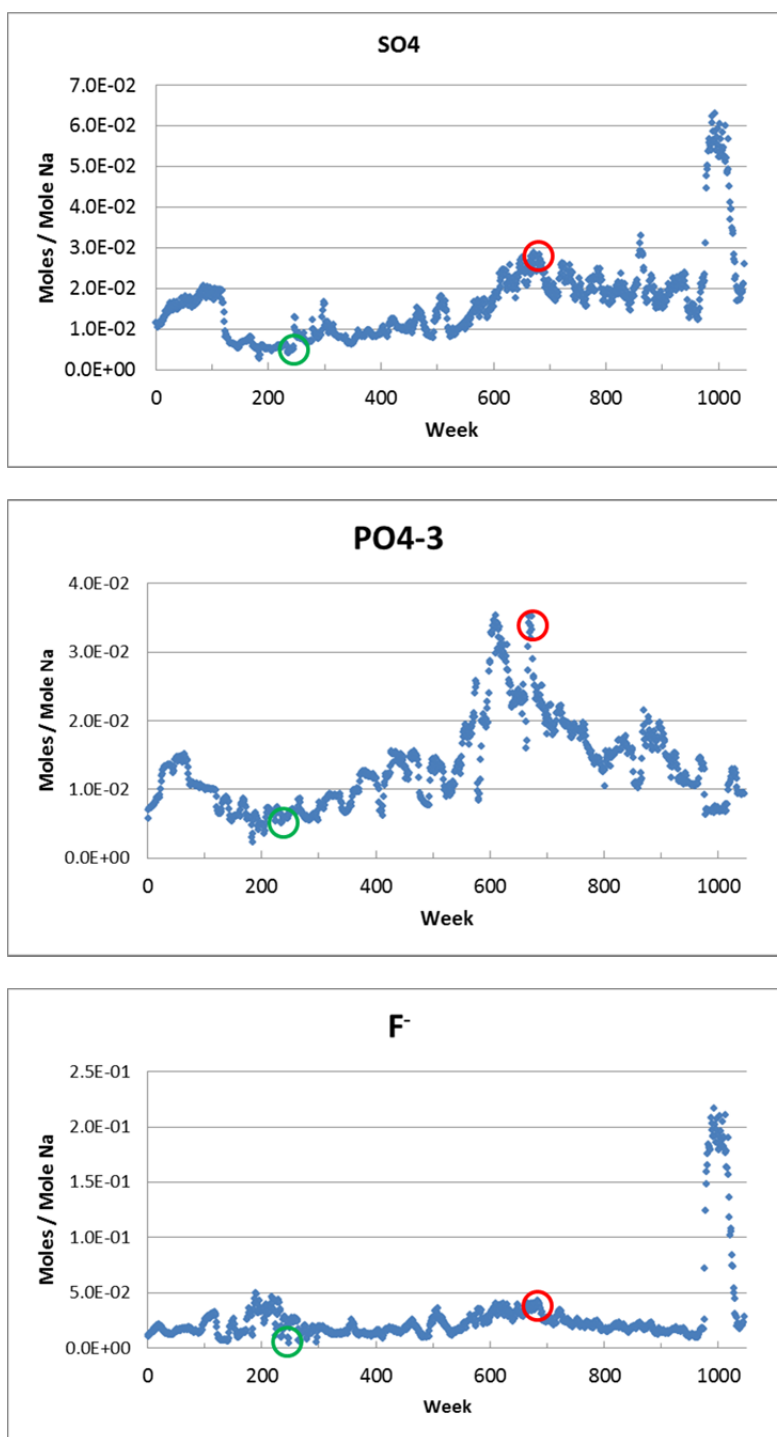


Figure 3-2. Sulfate, Phosphate, and Fluoride Concentrations from HTWOS Flowsheet Simulations. Week 672 in red circle, week 235 in green circle.

Table 3-2. Final LAW Simulants for Cast Stone Screening Tests

Waste Constituent	SST Blend Saltcake	HTWOS Overall Average	HTWOS High Al	HTWOS High SO ₄
Concentration (moles/mole Na) ^(a)				
Na	1.000	1.000	1.000	1.000
K	0.002	0.007	0.028	-
Al	0.013	0.061	0.112	0.047
Cl	0.009	0.008	0.018	0.007
F	0.006	0.006 ^(b)	0.010	0.012 ^(b)
SO ₄	0.018	0.017	0.004	0.030
PO ₄	0.010	0.010 ^(b)	0.005	0.010 ^(b)
NO ₂	0.085	0.113	0.194	0.098
NO ₃	0.502	0.324	0.287	0.367
CO ₃	0.095	0.055	0.040	0.035
TOC Total	0.057	0.015	0.021	0.007
Free OH	0.097	0.312	0.293	0.306

(a) After charge balancing.
(b) Concentration of F and PO₄³⁻ reduced from HTWOS values because of solids formation observed in preliminary simulants.
- = not included

3.3 Spike Levels for Constituents of Concern

In addition to the main components of the LAW, the simulants used to make Cast Stone waste forms prepared for contaminant release testing were spiked with hazardous chemicals and/or radionuclides of interest to determine how well the Cast Stone waste form controls the release of these constituents of concern (COCs). Results in the *Final Tank Closure and Waste Management Environmental Impact Statement for the Hanford Site, Richland Washington* (DOE/EIS 2012) showed that only ⁹⁹Tc, ¹²⁹I, Cr, and NO₃ had non-negligible projected groundwater concentrations from Cast Stone disposed at the IDF.

3.3.1 Hazardous Chemicals

Some testing required using spikes of COCs to address their retention within and release from the Cast Stone waste form. To address land disposal restrictions, including performance in the TCLP, spikes included RCRA metals and underlying hazardous constituents. The final spike levels are shown in Table 3-3 (Russell et al. 2013). For the screening tests, only Cr, Pb, Ni, and Cd were used to test the performance of RCRA metals in the Cast Stone waste form.

An initial RCRA spike vector was selected by taking the maximum weekly batch values from the HTWOS modeling for System Plan 6 (Certa et al. 2011) for the feed to Supplemental Low-Activity Waste Treatment. The list of RCRA metals to include in the simulants was reduced using total constituent analysis as allowed in Section 1.2 of EPA Method 1311 for the TCLP (EPA 1992; Russell et al. 2013). Conceptually, this analysis shows what the TCLP leachate concentrations would be if 100% of each COC were released from the waste form during the leaching procedure. This total constituent analysis screen showed As, Ba, Se, Ag, Sb, Be, and Ni levels derived from the waste feed compositions to be below Universal Treatment Standards (UTSs). None of the COCs that falls below UTS by total analysis was added to the simulants in the screening tests with the exception of Ni. Nickel was included because it contributes a significant mass to the RCRA metals vector.

Mercury was excluded due to anticipated interactions with iodine. Mercury and silver can form highly insoluble compounds with iodine which would decrease the ability of iodine to leach from the waste form. Because the initial vector used HTWOS maximum weekly values, it would be unrealistic to test iodine performance in the presence of these other compounds that would have the effect of improving iodine performance (i.e., reduce its leaching) in the EPA Draft Method 1315 leach testing.

Table 3-3. Final Spike Levels for Hazardous Constituents and Radionuclides in Simulants for Cast Stone Screening Tests

Waste Constituent	HTWOS Overall Average	HTWOS Maximum	Other Considerations
RCRA Metals and UHCs ^(d)	moles/mole Na	moles/mole Na	moles/mole Na
Cd	2.78E-06	3.19E-05	-
Cr	2.42E-03	9.99E-03	4.30E-03 ^(a)
Pb	1.16E-05	5.13E-05	-
Ni	6.41E-05	6.61E-04	-
Radionuclides	Ci/mole Na	Ci/mole Na	Ci/mole Na
⁹⁹ Tc	1.13E-05	4.13E-05	-
⁹⁹ Tc ^(c)	(6.65E+02 µg/mole Na)	(2.43E+03 µg/mole Na)	-
¹²⁹ I	1.44E-08	8.01E-08	3.54E-06 ^(b)
¹²⁷ I (stable) ^(c)	(8.14E+01 µg/mole Na)	(4.53E+02 µg/mole Na)	(2.00E+04 µg/mole Na)
²³² U+ ²³³ U+ ²³⁴ U+ ²³⁵ U+ ²³⁶ U+ ²³⁸ U	1.59E-08	5.63E-08	-
Natural or depleted U ^(c)	-	(3.56+04 µg/mole Na)	-
(a) Cr concentration adjusted based on review of best basis inventory and previous simulant work.			
(b) Iodine concentration increased to address possible detection limits issues in waste form leach tests. Iodine added as nonradioactive ¹²⁷ I.			
(c) These COCs were added to simulants based on mass (as shown).			
(d) UHCs = underlying hazardous constituents			

Thallium was not included in the screening tests because the secondary waste Cast Stone program showed satisfactory performance for thallium in TCLP leach testing (Mattigod et al. 2011). Thallium is also very close to the UTS value by total analysis (0.36 mg/L versus 0.2 mg/L) and thus would require minimal attenuation in TCLP leach testing to meet the UTS.

The HTWOS model for System Plan 6 (Certa et al. 2011) predicts a maximum Cr level in the LAW supplemental feed of 1.0×10^{-2} M/M Na. The average Cr concentration is 2.4×10^{-3} M/M Na and the 95th percentile for Cr is 4.3×10^{-3} M/M Na. The 2003 Cast Stone work used the SST Blend saltcake simulant with a Cr level of 2.0×10^{-3} M/M Na and the SST saltcake blend of real waste with a ratio of 3.7×10^{-3} M/M Na (Rapko et al. 2003). Simulants used in testing Cast Stone for the secondary waste program used a Cr ratio as high as 4.2×10^{-3} M/M Na (Sundaram et al. 2011). Based on the preceding information, a Cr spike level of 4.3×10^{-3} M/M Na corresponding to the HTWOS 95th percentile composition was selected for supplemental LAW Cast Stone waste form testing.

3.3.2 Radionuclides

To understand the retention and release of radionuclides of concern, spikes of ⁹⁹Tc, ¹²⁹I (substituted with nonradioactive ¹²⁷I), and U were added to Cast Stone batches prepared for EPA Draft Method 1315 leach testing studies. Table 3-3 shows the spike levels for these

radionuclides. The ^{99}Tc was spiked in all samples at the HTWOS maximum concentration. To increase the probability of being able to detect iodine in as many of the leachates as possible and at various leach intervals, the stable iodine (^{127}I) concentration in the simulants was increased to 100 mg/L for the 5 M Na simulants and 156 mg/L for the 7.8 M simulants. These concentrations of stable iodine are 245 times larger than the average mass concentration of ^{129}I and 44 times larger than the maximum mass concentration of ^{129}I projected by the HTWOS model to be present in LAW.

4.0 Dry Materials

The basic Cast Stone dry blend is composed of 47 wt% BFS, 45 wt% Class F fly ash, and 8 wt% ordinary portland cement (OPC, Type I/II) (Lockrem 2005). This basic dry blend composition was used for all the screening tests. Two sources of BFS and two sources of fly ash were used as part of the test matrix. One of the BFS and the high-Ca fly ash were from a supplier using sources available in the Pacific Northwest. The second BFS and the low-Ca fly ash are the same materials used in the Saltstone Processing Facility at the SRS in South Carolina. A single source of portland cement from the Pacific Northwest was used in the dry blend mixes. This section describes the chemical composition and X-ray diffraction characteristics of each of the dry blend components. Additional details are available in Appendix B (Dry Blend Materials Characterization) and Appendix F (Material Certification Reports).

4.1 Blast Furnace Slag

The product used for one source of BFS was Lafarge NewCem® slag cement. NewCem is a finely ground BFS produced from the iron-making process. The material was obtained from LaFarge North America Inc. in Pasco, Washington. This material is referred to as the NW slag in the discussions of the Cast Stone properties. The product used for the second source of BFS was granulated BFS obtained from Holcim (US) Inc. headquartered in Waltham, Maine. This product is referred to as the SE slag in the discussions of the Cast Stone properties because it is used at SRS. Test reports from the suppliers for the two BFSs are provided in Appendix F. Table 4-1 lists the compositions of the two slags as measured at PNNL using both inductively coupled plasma (ICP) and X-ray fluorescence (XRF) and at SRNL using ICP. The analytical methods are described in Appendix B.

The NW slag is characterized as having higher concentrations of Al, Ca, and S and lower concentrations of Mg and Si compared to the SE slag. Figure 4-1 shows the X-ray diffractograms for the two slags. The SE slag is amorphous, while the diffractogram of the NW slag indicates that gypsum has been added. In compositions containing an abundance of Al, such as the NW slag, suppliers add gypsum to provide a soluble sulfate source that reacts with the tricalcium aluminate to form ettringite, thus preventing flash setting of the concrete mix (Ramachandran and Feldman 1995).

Additional BFS characterization information including scanning electron microscopy (SEM) micrographs, heat generation, particle size, thermal gravimetric analyses, surface area, densities, and Hausner ratio are presented in Appendix B.

4.2 Fly Ash

The product used for the high-Ca fly ash was a Class C/Class F fly ash produced from the combustion of coal at the Centralia Power plant in western Washington. The material was obtained from LaFarge North America Inc. in Pasco, Washington. This material is referred to as the NW fly ash in the discussions of the Cast Stone properties. The product used for the low-Ca fly ash was a thermally beneficiated Class F fly ash from the Wateree Station Carbon Burn Out (CBO) facility and was obtained from South Carolina Electric & Gas through the SEFA Group in Lexington, South Carolina. This product is referred to as the SE fly ash in the discussions of the Cast Stone properties. Test reports from the suppliers for the two fly ashes are provided in Appendix F.

Table 4-2 lists the chemical compositions of the two sources of fly ash as measured at PNNL and SRNL. Analytical methods are described in Appendix B.

Table 4-1. Measured Chemical Composition of Blast Furnace Slags

Analyte	NW Slag (µg/g)		SE Slag (µg/g)		Oxide	NW Slag (wt%)			SE Slag (wt%)		
	PNNL	SRNL	PNNL	SRNL		PNNL		SRNL	PNNL		SRNL
	ICP	ICP	ICP	ICP		ICP	XRF	ICP	ICP	XRF	ICP
Aluminum	77,700	67,300	43,300	45,100	Al ₂ O ₃	14.68	11	12.7	8.18	7.2	8.51
Antimony	<2340	-	<2320	-		-	-	-	-	-	-
Arsenic	<28.6	-	<28.3	-		-	-	-	-	-	-
Barium	523	358	364	358	BaO	0.06	0.057	0.04	0.04	0.035	0.03
Cadmium	<4.72	<100	<4.67	<100		-	-	-	-	-	-
Calcium	356,000	304,000	228,000	240,000	CaO	49.81	46	42.6	31.9	36	33.6
Chromium	<65.6	<100	69.6	130	Cr ₂ O ₃	-	0.01	-	0.01	0.009	0.02
Copper	<113	<100	<112	<100		-	-	-	-	-	-
Iron	6,200	4,100	2,300	1,970	Fe ₂ O ₃	0.89	0.78	0.59	0.33	0.33	0.28
Lead	<17.9	<100	<17.7	<100		-	-	-	-	-	-
Magnesium	27,700	25,200	69,300	78,700	MgO	4.59	4.9	4.2	11.49	13	13.1
Manganese	2,130	1,530	3,330	2,780	MnO ₂	0.34	0.28	0.24	0.53	0.55	0.44
Mercury	<15.9	-	<8.63	-		-	-	-	-	-	-
Molybdenum	<31.4	<100	<31.1	<100		-	-	-	-	-	-
Nickel	<488	<1,000	<484	<1,000		-	-	-	-	-	-
Phosphorus	<3110	<1,000	<3080	<1000	P ₂ O ₅	-	0.021	-	-	0.032	-
Potassium	<9010	2,800	<8930	3,590	K ₂ O	-	0.37	0.34	-	0.43	0.43
Selenium	<5980	-	<5920	-		-	-	-	-	-	-
Silicon	181,000	154,000	190,000	190,000	SiO ₂	38.72	33	33	40.65	39	40.6
Silver	<1.79	-	<1.77	-		-	-	-	-	-	-
Sodium	<6630	1,530	<6570	1,860	Na ₂ O	-	0.52	0.21	-	0.5	0.25
Strontium	670	-	352	-	SrO	0.08	0.074	0.07	0.07	0.043	0.05
Sulfur	23,800	16,400	<11100	7,810	SO ₄	7.13	5	4.9	(<3.33)	1.69	2.3
Titanium	-	2,300	-	1,580	TiO ₂	-	0.56	0.36	-	0.35	0.25
Zinc	-	<100	-	<100	ZnO	-	-	0.03	-	-	-
Zirconium	-	183	-	293	ZrO ₂	-	0.035	-	-	0.023	0.04
Total Carbon	751	-	632	-	C	-	-	-	-	-	-
					Mass %	116.3%	102.6%	99.2%	93.2%	99.2%	99.9%

- = Not measured or not detected.

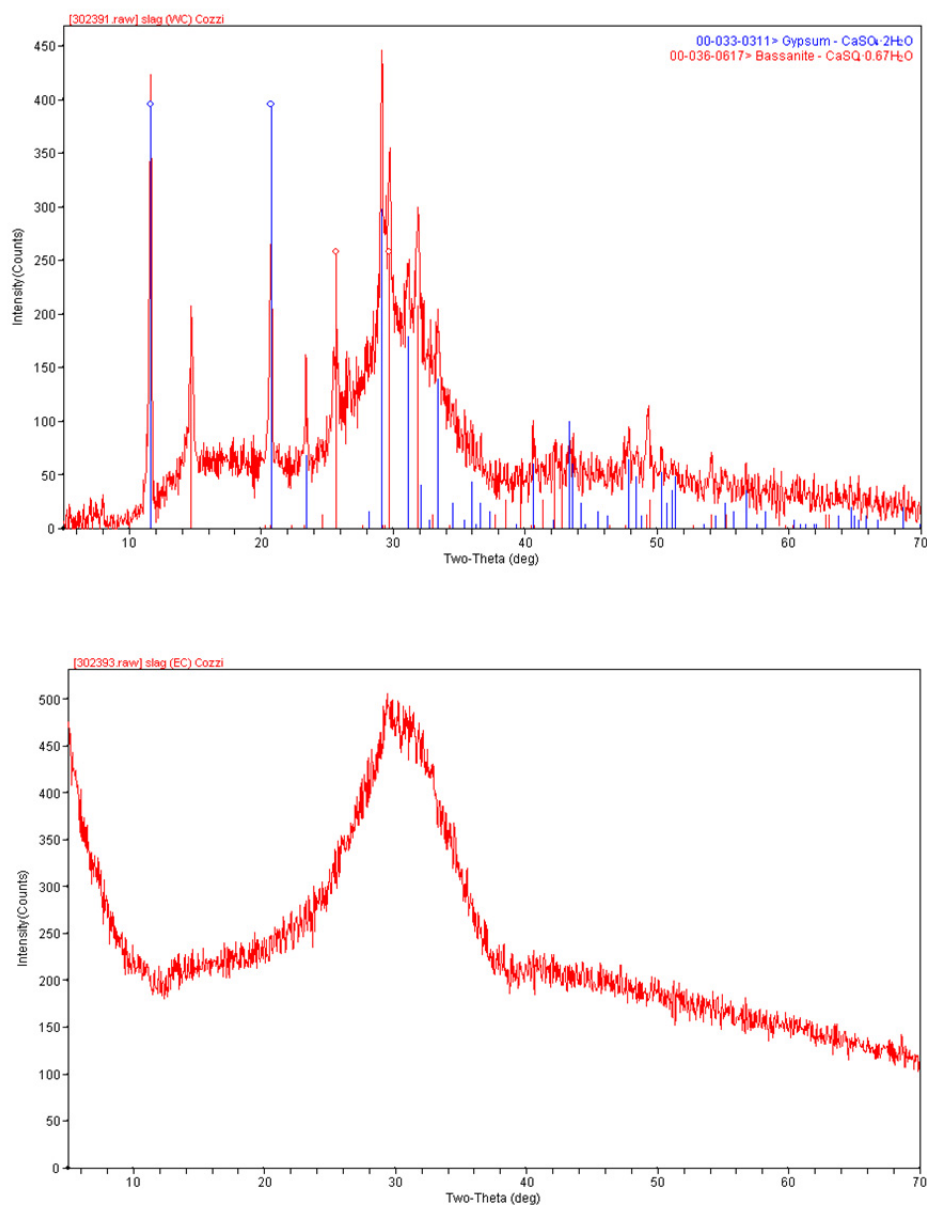


Figure 4-1. X-Ray Diffraction Patterns Northwest (top) and Southeast (bottom) Blast Furnace Slags

The NW fly ash has higher concentrations of Ca and Mg and lower concentrations of Al compared to the SE fly ash. Figure 4-2 shows the X-ray diffractograms for the fly ashes. The diffractograms for the NW fly ash indicated the presence of quartz, anhydrite,¹ and periclase,²

¹ Anhydrite is anhydrous calcium sulfate (CaSO_4).

² Periclase is a cubic form of magnesium oxide (MgO). Periclase is usually found in [marble](#) produced by [metamorphism](#) of [dolomitic limestones](#).

while the diffractograms for the SE fly ash identify the quartz and mullite¹ crystalline phases. These results are consistent with Class F fly ashes from their respective regions (McCarthy et al. 1989).

Appendix B contains additional information about the properties of the fly ashes including SEM micrographs, pozzolanic activity, particle size distribution, thermal gravimetric analysis, surface areas, and densities.

Table 4-2. Measured Chemical Composition of Fly Ashes

Analyte	NW FA (µg/g)		SE FA (µg/g)	Oxide	NW FA (wt%)			SE FA (wt%)	
	PNNL	SRNL	SRNL		PNNL		SRNL	PNNL	SRNL
	ICP	ICP	ICP		ICP	XRF	ICP	XRF	ICP
Aluminum	103,000	86,100	142,000	Al ₂ O ₃	19.46	16	16.3	25	26.8
Antimony	<2,320	-	-	-	-	-	-	-	-
Arsenic	<28.3	-	-	-	-	-	-	-	-
Barium	6,960	4,840	1,240	BaO	0.78	0.71	0.54	0.18	0.14
Cadmium	<4.68	<100	<100	-	-	-	-	-	-
Calcium	114,000	92,800	17,200	CaO	15.95	15	13	2.8	2.4
Chromium	<65.0	233	275	Cr ₂ O ₃	-	0.015	0.03	0.034	0.02
Copper	<112	<100	<100	-	-	0.017	0.02	0.02	0.04
Iron	52,700	41,500	54,400	Fe ₂ O ₃	7.53	6.8	5.93	8.0	7.8
Lead	31.3	<100	<100	-	-	-	-	-	-
Magnesium	30,000	26,500	5,280	MgO	4.97	5.6	4.4	2.1	0.88
Manganese	557	473	177	MnO ₂	0.09	0.088	0.08	0.03	0.03
Mercury	<15.8	-	-	-	-	-	-	-	-
Molybdenum	<31.1	<100	<100	-	-	-	-	-	-
Nickel	<484	<1000	<1000	-	-	-	-	-	-
Phosphorus	<3,080	<1000	<1000	P ₂ O ₅	-	0.41	-	0.31	-
Potassium	16,800	12,400	18,600	K ₂ O	2.02	1.7	1.49	2.8	2.2
Selenium	<5,920	-	-	-	-	-	-	-	-
Silicon	270,000	242,000	260,000	SiO ₂	57.76	48	51.8	56	55.6
Silver	2.18	-	-	-	-	-	-	-	-
Sodium	34,100	24,700	3,720	Na ₂ O	4.6	3.8	3.3	0.71	0.5
Strontium	3,730	-	-	SrO	0.44	0.38	0.3	0.159	0.12
Sulfur	<11,100	3,390	709	SO ₄	-	1.1	1	0.2	0.21
Titanium	-	4,760	7,340	TiO ₂	-	1.2	0.75	1.7	1.16
Zinc	-	111	139	ZnO	-	0.02	0.01	0.024	0.02
Zirconium	-	336	338	ZrO ₂	-	0.034	0.05	0.055	0.05
Total Carbon	530	-	-	C	-	-	-	-	-
				Mass %	107.0%	100.9%	99.0%	100.1%	98.1%

- = Not measured or not detected.

4.3 Ordinary Portland Cement

The product used for the OPC was a portland cement Type I/II obtained from Lafarge North America Inc. in Pasco, Washington. The cement is from the LaFarge plant in Richmond, British Columbia. The vendor test report for this portland cement is provided in Appendix F.

¹ Mullite is a silicate mineral of post-clay genesis. It can form two stoichiometric forms 3Al₂O₃•2SiO₂ or 2Al₂O₃•SiO₂. It is produced during various melting and firing processes.

Table 4-3 lists the chemical composition based on chemical analyses at PNNL and SRNL. Figure 4-3 shows the X-ray diffractogram of the cement used in this study. The phases present are consistent with OPC (Walenta 2004). The analytical methods use to characterize the OPC are described in Appendix B. This cement was used for testing at both PNNL and SRNL.

Appendix B contains additional information about the cement including SEM micrographs, heat flow and total heat, particle size distribution, thermal gravimetric analysis, surface area, and density.

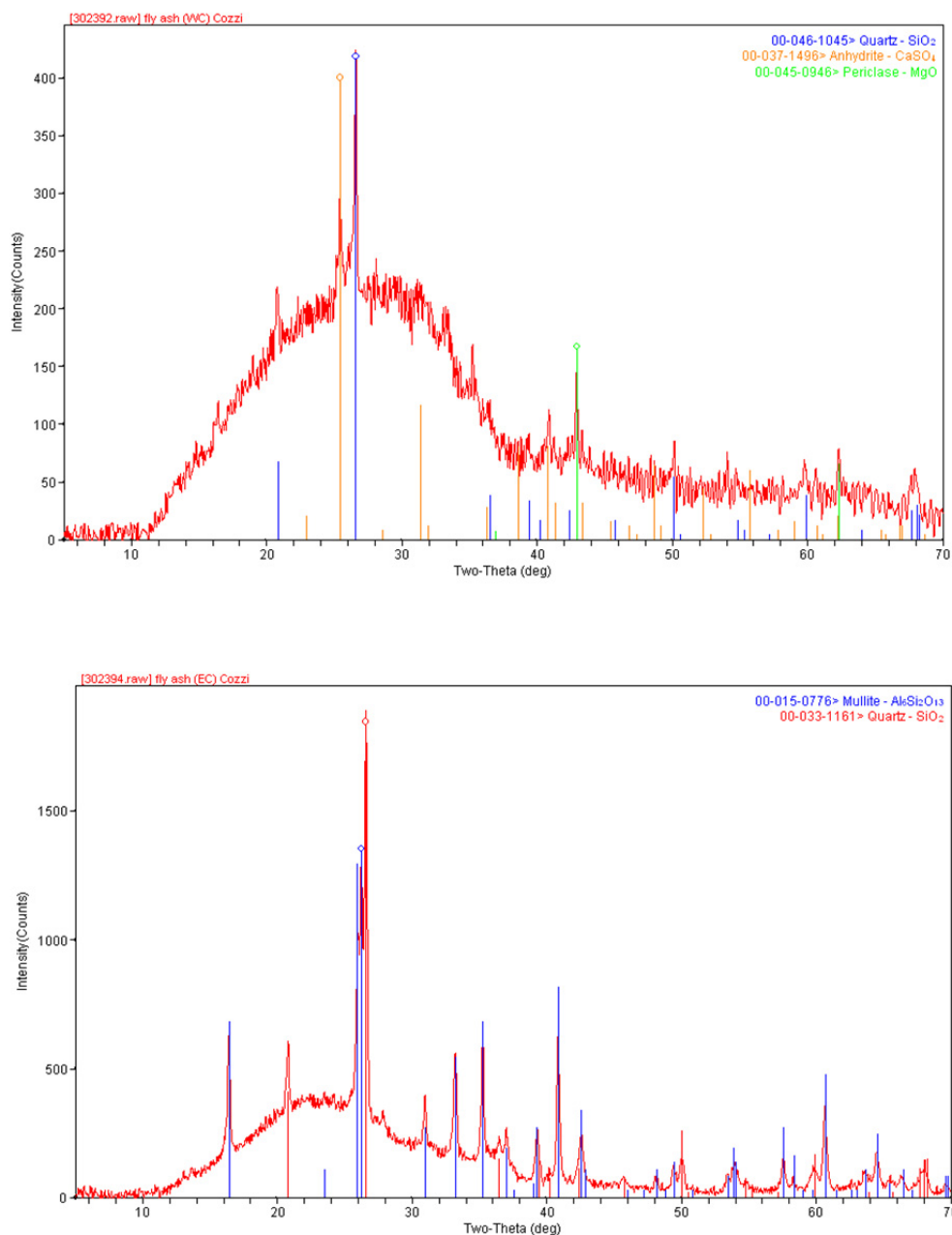


Figure 4-2. X-Ray Diffraction Patterns Northwest (top) and Southeast (bottom) Fly Ashes

Table 4-3. Measured Chemical Composition of Ordinary Portland Cement

Analyte	NW OPC (µg/g)	NW OPC (µg/g)	Oxide	NW OPC (wt%)		
	PNNL	SRNL		PNNL		SRNL
	ICP	ICP		ICP	XRF	ICP
Aluminum	27,600	25,600	Al ₂ O ₃	5.21	3.7	4.83
Antimony	<2,320	-	-	-	-	-
Arsenic	<28.3	-	-	-	-	-
Barium	492	394	BaO	0.05	0.06	0.04
Cadmium	<4.67	<100	-	-	-	-
Calcium	486,000	451,000	CaO	68	66	63.1
Chromium	165	203	Cr ₂ O ₃	0.02	0.03	0.03
Copper	242	315	-	0.03	0.035	0.04
Iron	27,800	24,900	Fe ₂ O ₃	3.97	3.4	3.55
Lead	37.9	<100	-	0.004	-	-
Magnesium	5,010	4,050	MgO	0.83	1.7	0.69
Manganese	614	579	MnO ₂	0.1	0.1	0.09
Mercury	<15.8	-	-	-	-	-
Molybdenum	<31.1	<100	-	-	-	-
Nickel	<484	<1000	-	-	-	-
Phosphorus	<3,080	<1000	P ₂ O ₅	-	-	-
Potassium	<8,930	2710	K ₂ O	-	0.34	0.33
Selenium	<5,920	-	-	-	-	-
Silicon	110,000	96,100	SiO ₂	23.53	19	20.6
Silver	1.79	-	-	-	-	-
Sodium	<6,570	2,400	Na ₂ O	-	0.54	0.32
Strontium	1,480	-	SrO	0.18	0.16	0.16
Sulfur	14,000	16,200	SO ₄	4.19	3.63	4.86
Titanium	-	1,480	TiO ₂	-	0.31	0.23
Zinc	-	1,390	ZnO	-	0.18	0.17
Zirconium	-	233	ZrO ₂	-	0.019	0.03
Total Carbon	4,050	-	C	-	-	-
			Mass %	106.1%	99.2%	99.1%

- = Not measured or not detected.

4.4 Final Dry Blend Mix Chemical Compositions

Prior to making the Cast Stone monoliths, the dry materials were pre-mixed in the four combinations needed to perform the screening tests. Table 4-4 lists the measured chemical compositions of the four dry blend mixes as measured at PNNL. Additional characterization data are presented in Appendix B.

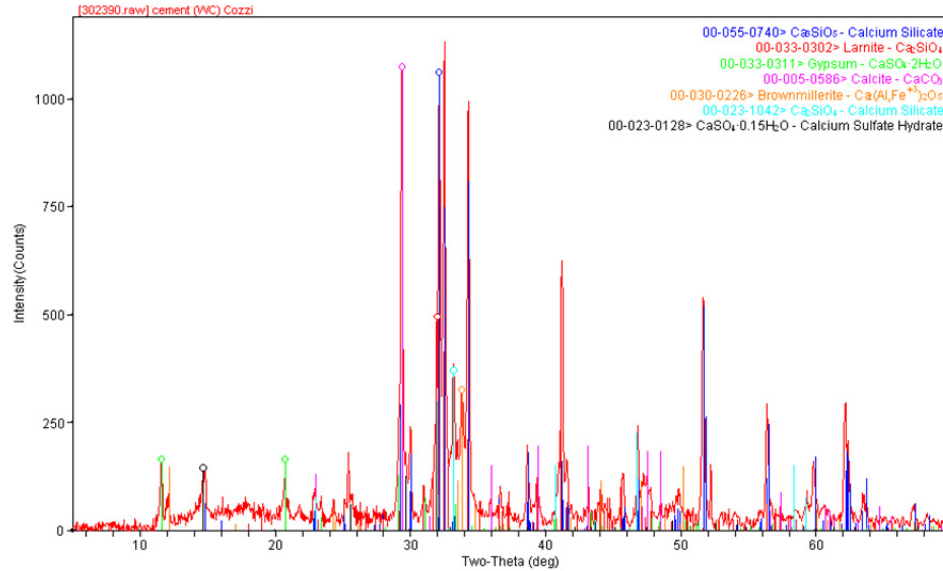


Figure 4-3. X-Ray Diffraction Pattern of Portland Cement Used for All Cast Stone Mixes

Table 4-4. Measured Chemical Compositions of Final Dry Blend Mixes

Analyte	NW High Ca FA NW BFS (µg/g)		NW High Ca FA SE BFS (µg/g)		SE Low-Ca FA NW BFS (µg/g)		SE Low-Ca FA SE BFS (µg/g)	
	ICP	XRF	ICP	XRF	ICP	XRF	ICP	XRF
Aluminum	-	59,000	69,200	51,100	93,100	83,700	84,100	74,400
Arsenic	-	-	< 28.4	-	43.9	-	37.1	-
Barium	-	3,260	2,740	3,010	895	1,060	871	953
Calcium	-	232,000	206,000	206,000	171,000	191,000	153,000	168,000
Chromium	-	86	84.7	94	122	145	124	155
Copper	-	105	< 112	93	114	126	< 110	114
Iron	-	23,200	23,500	20,400	28,300	28,100	26,400	25,300
Lead	-	-	< 17.8	-	52.2	-	42.4	-
Magnesium	-	27,200	21,800	54,500	13,500	19,000	36,300	45,900
Manganese	-	1,290	1,030	2,110	808	1,030	1,630	1,850
Molybdenum	-	30.7	< 31.2	22.6	< 31.6	24	< 30.6	18.7
Nickel	-	52.9	< 486	45.2	< 492	92.6	< 477	85.3
Phosphorus	-	651	< 3,100	573	< 3,130	545	< 3,040	477
Potassium	-	8,720	9,370	8,200	12,300	12,100	12,000	11,400
Silicon	-	165,000	185,000	187,000	189,000	182,000	206,000	201,000
Silver	-	-	< 1.78	-	2.35	-	1.95	-
Sodium	-	12,500	13,200	12,000	< 6,680	4,080	< 6,470	4,160
Strontium	-	1,710	1,620	1,580	891	944	824	853
Sulfur	-	10,600	11,300	5,310	< 11,300	9,180	< 11,000	4,270
Titanium	-	4,570	4,000	4,030	5,330	6,370	4,930	5,710
Total Carbon	-	-	946	-	4,050	-	3,480	-
% Dry Solids (% by weight)	-	-	99.8	-	99.5	-	99.8	-
- = Not measured.								

5.0 Cast Stone Preparation

The Cast Stone monoliths were prepared by mixing aliquots of the simulant batches and the dry blend mixes and then casting the slurry into plastic molds to cure. Batches of each of the four simulants were prepared ahead of time as described in Section 3.0. Similarly, 26 individual 1.75-kg batches of the dry blend mixes were prepared ahead of time by weighing the cement, fly ash, and BFS at a ratio of 8:45:47, and mixing the materials by hand in a large plastic bag. The Cast Stone monoliths were prepared in the sequence as indicated by the random run order shown in Table 2-1 or Table C-1, depending on the property. To reduce the impact of inter-laboratory sample preparation, the mixing method developed at SRNL for fresh properties was used at PNNL for sample preparation.

At PNNL, the monoliths were prepared as follows. Simulants were aliquoted for each respective batch into a 2-L plastic beaker. The beaker containing the simulant was placed under the overhead mixer. A Caframo model BDC1850 overhead mixer with a 3.5-in. outer diameter (OD) elliptical impeller (Figure 5-1) was used to mix the simulants and blend the Cast Stone mixture. The beaker was positioned such that the impeller was 0.5 in. from the front of the beaker and 1 in. off the bottom, and the impeller rotation was as shown in Figure 5-1. The appropriate mass of simulant was placed in the 2-L plastic beaker and the mixer was started with an agitator speed of 200 rpm. When agitation was started, the ^{99}Tc and U radionuclide spikes were added to the simulant. After approximately 1 minute of agitation, the pre-mixed dry material was added to the beaker over a time period of 4.5 to 7 minutes in the location indicated in Figure 5-2. The agitation speed was adjusted to maintain a slight vortex (Figure 5-2). The wet slurry was stirred for 15 minutes from the start of addition of the dry blend. At the completion of mixing, the Cast Stone material was poured into 2-in. \times 4-in. forms. The forms were agitated to release any potential entrained air and capped with a perforated lid as shown in Figure 5-3. The capped monoliths were placed in racks within a 5-gal bucket (Figure 5-3) containing \sim 1 in. of water at the bottom to provide a humidified curing environment. The racks with the Cast Stone monoliths were above the 1-in. water layer. The bucket was closed and the monoliths were allowed to cure at room temperature for 28 days before testing.

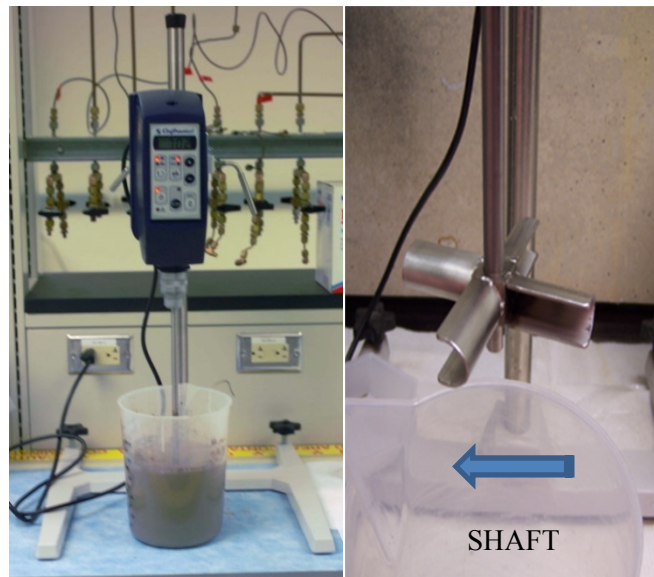


Figure 5-1. Overhead Mixer and Impeller for Cast Stone Preparation

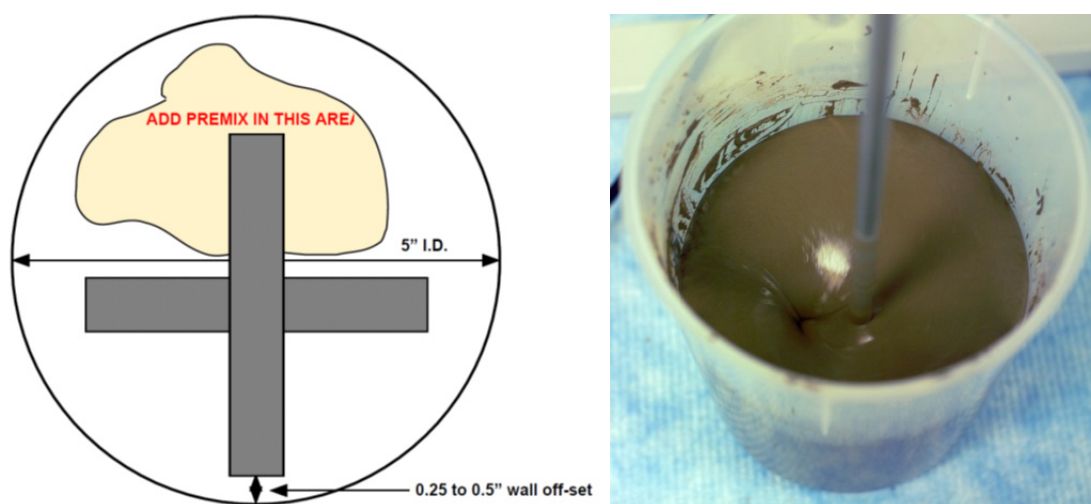


Figure 5-2. Position of Dry Blend Addition and Vortex Formed During Mixing



Figure 5-3. Cast Stone Monolith in Plastic Mold and Monoliths Curing in Bucket

The setup used at SRNL was the same as shown in Figure 5-1. A Caframo model BDC3030 overhead mixer was used with the same impellers that SRNL fabricated for the PNNL mixing step. The volume of the batch varied depending on the properties to be measured. The size of the plastic beaker was chosen to be approximately twice the volume of the mix being prepared. The initial agitator speed was 200 rpm and the initial temperature of the salt solution was recorded. As the dry blend was added, the agitator speed was increased to incorporate the dry blends. All of the dry blend materials were added within 2 minutes. After the dry blend was incorporated, the temperature of the slurry was recorded. Mixing continued for a total of 15 minutes. The agitator speed was adjusted to maintain a vortex as shown in Figure 5-2. At the conclusion of mixing, the temperature of the slurry was recorded.

6.0 Cast Stone Processing and Curing Properties

The freshly prepared Cast Stone was characterized for properties relevant to the mixing, pouring, flow, and heat generation during curing. These properties were measured at SRNL using Cast Stone slurries prepared at SRNL. The properties are discussed in the order in which the samples were cast. Additional information is available in Appendix C.

After the processing properties of the initial 26 Cast Stone mixes had been characterized, SRNL discovered that the sources of slag in two of the four possible fly ash and slag combinations had been transposed on the laboratory preparation batch sheets. This occurred at about the same time the project changed terminology for describing these materials from “Source 1” and “Source 2” for the slag to “NW” and “SE” and from “High Ca” and “Low Ca” for the fly ash to “NW” and “SE.” SRNL researchers investigated and confirmed, by comparing analyzed chemical compositions to compositions calculated from the dry blend and simulant compositions, that 12 of the 26 mixes were affected. They also confirmed that the remaining 14 mixes had been properly batched. As a corrective action, SRNL repeated the 12 mixes with the correct dry blend combinations and measured all of the properties that had previously been measured with the exception of compressive strength, cured density, and porosity. The mis-batched samples were assigned mix numbers 27 through 38 and, where appropriate, the data were included in the statistical analysis and presented in this report. Table 6-1 shows the test conditions for the additional 12 mixes. Table C-1 and Table C-2 in Appendix C shows the entire test matrix with the 38 mixes.

6.1 Flow Consistency (Modified ASTM D6103)

The flow consistency test is a demonstrative method of expressing the measured rheological properties (yield stress and plastic viscosity) available in field conditions. The ASTM International Standard D6103, *Standard Test Method for Flow Consistency of Controlled Low Strength Material (CLSM)* (ASTM D6103 2004), was developed to provide an accepted, consensus method of measuring the flow characteristics of CLSM. CLSM is much more fluid than concrete so that it readily fills voids and spaces. This test method provides a procedure to quantify the flow characteristics.

6.1.1 Flow Consistency Approach

An open-ended cylinder was placed on a flat, level surface and filled with fresh Cast Stone slurry. The cylinder was raised quickly so that the slurry flowed into a patty. The average diameter of the patty was determined and compared to mixes of the same or similar mix compositions. A cylinder of smaller proportions than those specified by the ASTM method was used (77 mm height × 43 mm inside diameter [ID] rather than the 150 mm × 76 mm specified in the method). This was done based on historical data indicating that some of the mixes tested would flow a distance that could not be easily measured or contained when the larger cylinders were used.

6.1.2 Flow Consistency Results

Flow diameters for the 38 Cast Stone mixes (Table C-1) ranged from 93.5 mm (Mix 1) to 298.3 mm (Mix 12). Figure 6-1 presents photographs of samples with the smallest and largest flow diameters. Figure 6-2 shows the flow diameters for the 38 mixes. Note that the colored circles in this and future figures at zero value represent test conditions that were not included in the screening test matrix. The results are tabulated in Appendix C. All of the mixes with a water-to-dry-blend solids ratio of 0.60 resulted in a flow diameter greater than the mixes with a water-to-dry-blend solids ratio of 0.40 (see Table C-3 in Appendix C).

Table 6-1. Twelve Mixes with BFS Sources Inadvertently Switched for Cast Stone Properties Tested by SRNL

Mix Number ^(a)	Random Run Order ^(b)	Block ^(c)	Simulant	Sodium Molarity	Portland Cement	Fly Ash Class F	Blast Furnace Slag	Dry Blend Mix ^(d)	Water-to-Dry Blend Solids Ratio
27 (3a) ^	3	1	Average	7.8	Type I/II	NW High Ca	NW	8:45:47	0.60
28 (4a)	5	1	High Al	5.0	Type I/II	NW High Ca	NW	8:45:47	0.40
29 (5a)	9	1	Average	7.8	Type I/II	NW High Ca	SE	8:45:47	0.40
30 (9a)	10	1	Hi Al	7.8	Type I/II	NW High Ca	NW	8:45:47	0.60
31 (10a)	13	1	High SO ₄	5.0	Type I/II	NW High Ca	SE	8:45:47	0.60
32 (12a)	8	1	SST Blend	5.0	Type I/II	NW High Ca	NW	8:45:47	0.60
33 (15a) *	17	2	High SO ₄	7.8	Type I/II	NW High Ca	SE	8:45:47	0.40
34 (16a)	22	2	SST Blend	7.8	Type I/II	NW High Ca	SE	8:45:47	0.40
35 (20a)	24	2	Average	5.0	Type I/II	NW High Ca	NW	8:45:47	0.40
36 (22a) ^	19	2	Average	7.8	Type I/II	NW High Ca	NW	8:45:47	0.60
37 (24a)	26	2	High Al	5.0	Type I/II	NW High Ca	SE	8:45:47	0.60
38 (25a) *	21	2	High SO ₄	7.8	Type I/II	NW High Ca	SE	8:45:47	0.40
<p>(a) The mis-batched mixes were numbered 27–38. Following those numbers in parentheses are the original intended mix numbers followed by an “a”, so that the relationship to the intended mix numbers is clear. Mix numbers with the same symbols are replicates.</p> <p>(b) The random run order used for Mixes 27–38 (3a, 4a, ..., 25a) tested by SRNL were the same as for Mixes 3, 4, ..., 25 tested by PNNL.</p> <p>(c) The mis-batched mixes were run in the original Block 1 and Block 2 sets of tests.</p> <p>(d) 8 wt% cement: 45 wt% fly ash, 47 wt% blast furnace slag.</p>									

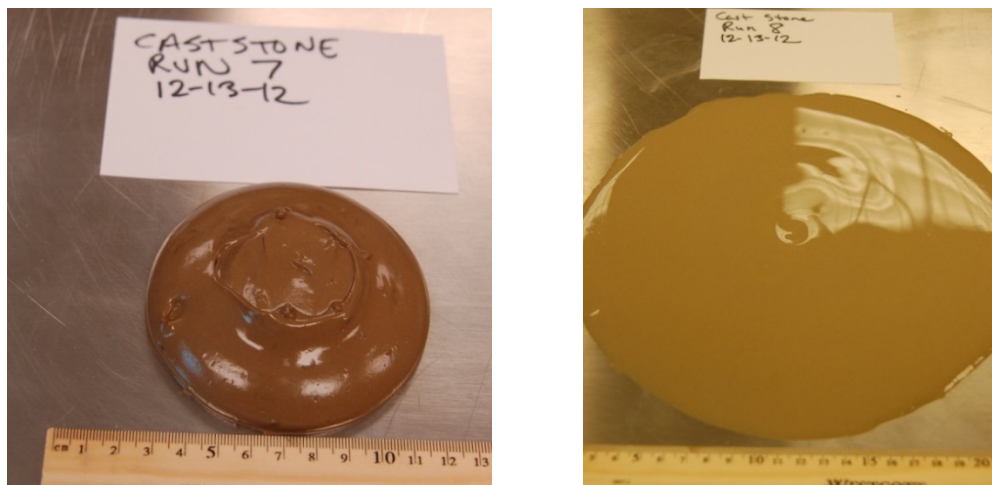


Figure 6-1. Flow Consistency for Cast Stone Mixes with the Smallest (Mix 1) and Largest (Mix 12) Flow Diameters

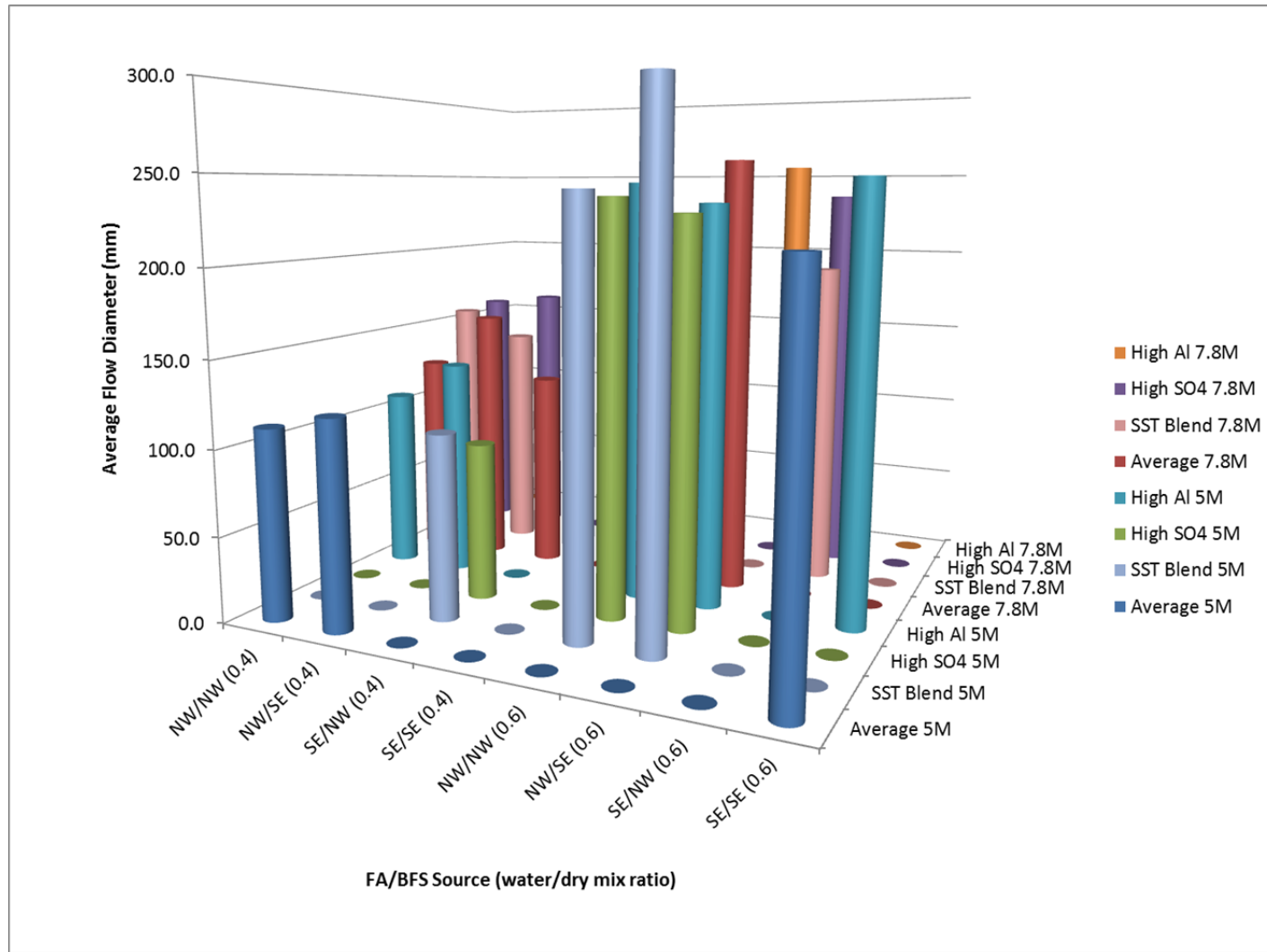


Figure 6-2. Average Flow Diameters for 38 Cast Stone Mixes

6.2 Gel Time

Gel time is a subjective method of determining the duration of slurry flowability. In a continuous process, the gel time is an indication of the time after an interruption in Cast Stone production that is available to restart the process before it becomes necessary to perform a cleanup/shutdown sequence. Gel time is also an indication of how long the placed Cast Stone (in a waste container) can maintain flowability to reach the boundaries of the waste container. However, formulations with gel times greater than the time necessary to reach the boundaries of the waste container have the potential for settling of the denser components—cement and slag—resulting in a layered waste form. Cozzi and Pickenheim (2012) evaluated saltstone grout mixes prepared with greater than normal water content and cast into standing water. Densities of cast cylinders were cut into sections and were measured individually to determine the effect of excess water on settling. Samples prepared with excess water and cast into standing water resulted in density gradients of approximately 10% through the 4-in. samples with a trend of decreasing density from the bottom section of the cylinder to the top. Cylinders prepared with typical water content of 0.6–0.64 water/dry mix ratio showed density variation of 1% or less.

6.2.1 *Gel Time Approach*

Gel time was measured by partially filling a series of ~100 ml containers (#VL25H, LAContainer, Yorba Linda, CA) with fresh Cast Stone slurry. After 5 to 10 minutes, a container was deliberately poured out and the flowability was evaluated. This process continued, with the time interval determined by the flowability of the prior pour, until the Cast Stone would no longer flow from the container when tilted to the pouring position. For each of the mixes prepared, five vials were approximately 75% filled with fresh Cast Stone slurry, and a timer was started. The initial pour time was determined by the operator based on the flow consistency measurement as described in Section 6.1. Subsequent pour intervals were determined by the operator based on the observed flowability in the previous pour.

6.2.2 *Gel Time Results*

Measured gel times for the 38 Cast Stone mixes (Table C-1) ranged from 1 minute to more than 2 hours (see Figure 6-3). For Mix 22, the gel test could not be completed with the number of samples available. The pours gave the operator the sense of gelation, but the mix continued to pour, leaving insufficient material to attain gel. With the exception of some overlap at the gel time of 15 minutes, mixes prepared with a water-to-dry-blend solids ratio of 0.60 exhibited longer gel times than mixes prepared with a water-to-dry-blend solids ratio of 0.40. Gel times tended to be highest for the 0.6 mix ratio and the 7.8 M simulant concentrations. The results are tabulated in Table C-4 of Appendix C.

6.3 Set or Hardening Time

Set time is measured by ASTM C191 *Standard Test Methods for Time of Setting of Hydraulic Cement by Vicat Needle* (ASTM C191 2008). A modified version of the final set described in the ASTM procedure was used to allow for up to 2 mm of penetration; the initial set is not currently used for Saltstone waste forms. The measurement time unit for the Cast Stone mixes was in days rather than minutes as in the ASTM procedure due to the retardation of the hydration reactions caused by the high concentrations of salts in the solutions being solidified. The set time of Saltstone was initially used to determine the time required between pours to prevent excessive hydraulic head on the vault walls at the SRS Saltstone Disposal Facility. The set time has since been used for estimating material at risk for deflagration calculations. Set time corresponds to the development of structure from hydration reactions and may be used as a process control point for

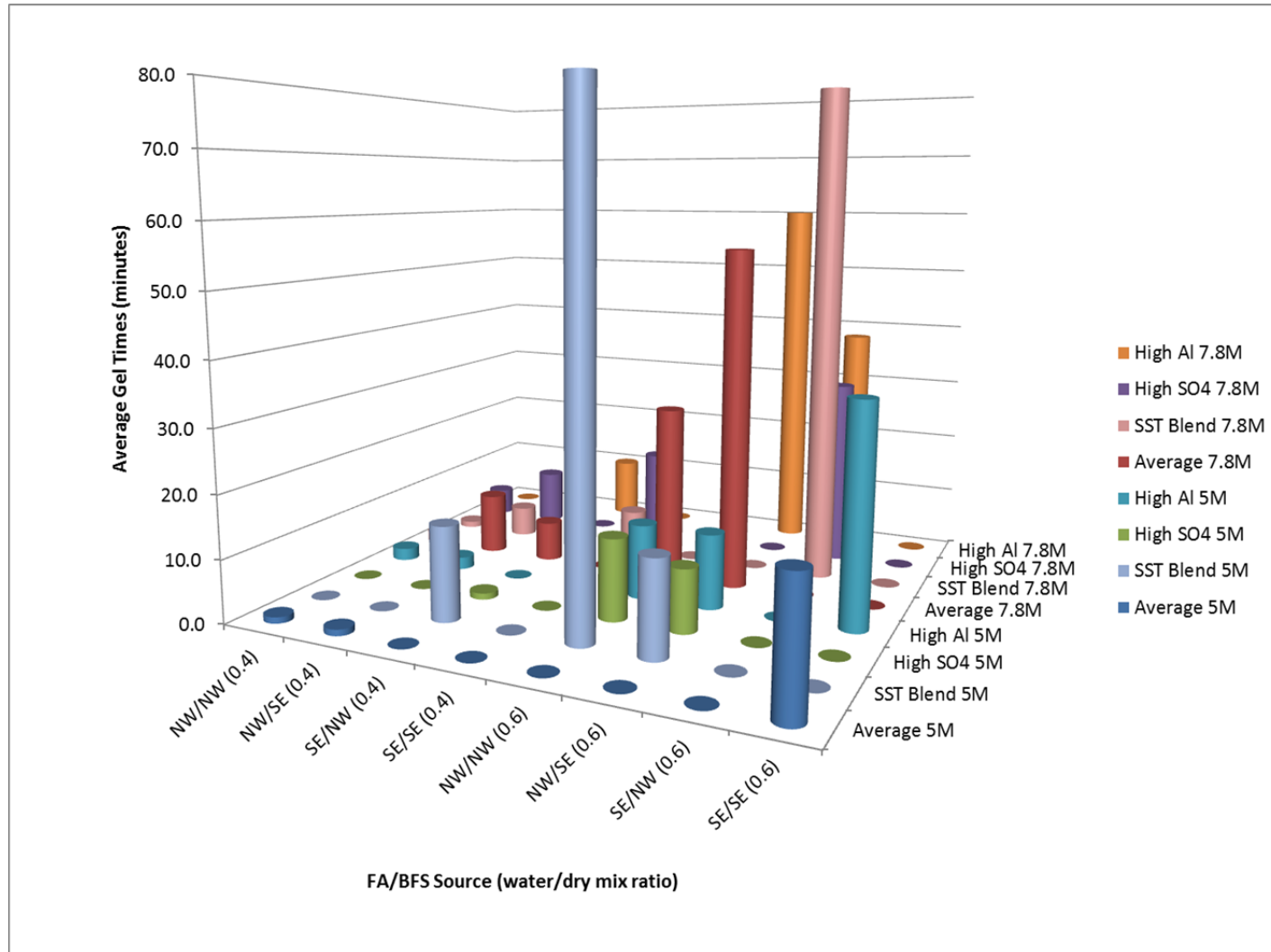


Figure 6-3. Gel Times for 38 Cast Stone Mixes

allowing the movement of Cast Stone waste packages from the production facility to interim storage or the disposal facility. The gel time and set time are milestones that occur during the curing of cementitious materials. Both provide an indication of the extent to which the curing process has progressed at that point in time. Because curing continues as long as there is humidity and unreacted material available, the term “curing” or “cured” only have meaning when defined for the specific use.

6.3.1 Set or Hardening Time Approach

To measure the set time of a Cast Stone waste form, fresh slurry is cast into a container to a depth of approximately 35 mm and the cast time is noted. The container is sealed and placed in a zip-top bag with a moistened towel to provide a humid environment. Each day, the sample is removed from the bag and the needle penetration is measured with a Vicat tester (#H-3050, Humboldt Mfg. Co., Schiller Park, IL). When the needle penetration is <2 mm, the sample is defined as set, and the time is noted. The set time is the elapsed time between the cast and set time.

In an effort to correlate the set time to other Cast Stone properties, slurry samples were cast into a mold for monitoring ultrasonic pulse velocity (UPV). A Pundit Lab meter (Proceq USA, Aliquippa, PA) (see Figure 6-4) was used to monitor the pulse velocity of an ultrasonic sound wave passed through a freshly cast sample. Two 150-kHz transducers (a transmitter and a receiver) were butted up against polycarbonate sheets separated by a rubber mold (see Figure 6-5). A standard polycarbonate cylinder with a known time of flight was used to calibrate and check the meter and transducer system (see Figure 6-6). The sample distance was corrected for the thickness and attenuation of the polycarbonate sheets. An ultrasonic couplant was used on the transducer ends to ensure a continuous path through the sample. When the sample for Vicat testing was cast, the UPV mold was filled with fresh slurry. The Proceq Pundit Lab meter was set with a pulse amplitude of 500 V. After casting, a sampling frequency of 1 second was used for the initial 10 velocity measurements. The sampling frequency was then reduced to 10 minutes for the remainder of the test. With each Vicat penetration test, the UPV through the sample was logged. The Cast Stone was declared set via the Vicat test when the penetration was 2 mm or less, then the velocity sampling frequency was reduced to 1 second for 10 iterations and the UPV run was terminated. The ultrasound velocity system was then checked with a calibration polycarbonate cylinder with a known travel time.



Figure 6-4. Proceq Pundit Lab Meter

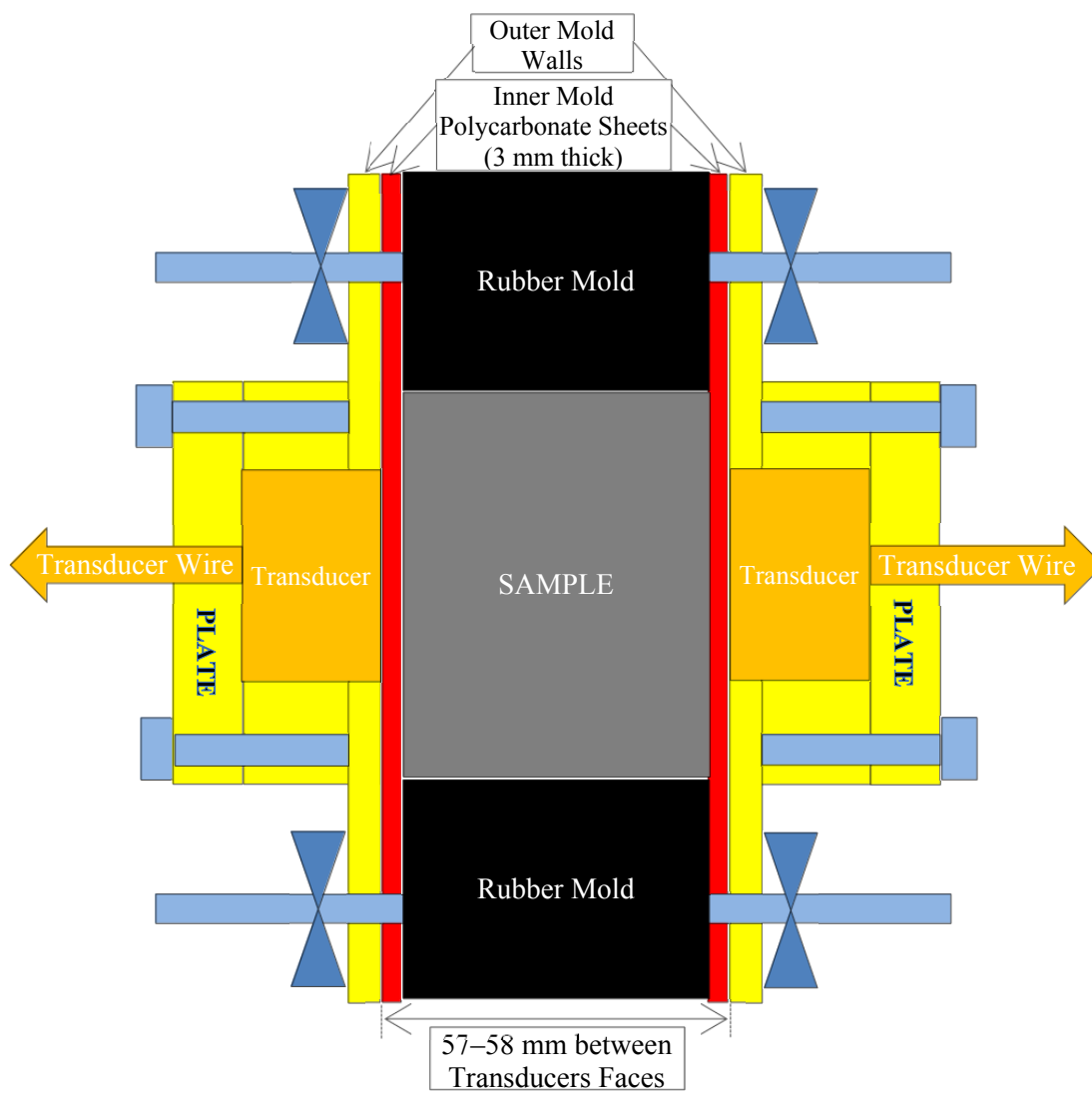


Figure 6-5. UPV Mold



Figure 6-6. UPV Calibration Rod

6.3.2 Set or Hardening Time Results

A representative UPV measurement is shown in Figure 6-7. Also on the plot are the Vicat results when it was noted that the sonic velocity was increasing. The two points plotted show the penetration of the needle fully extending to 35 mm at 24.7 hours and not penetrating at 28.8 hours, indicating “set.” The UPV generally rises, levels out, and then may rise again. It is believed the UPV measurements can be related to the Vicat measurements by performing special runs where the Vicat measurements are taken every 30 minutes. Either the onset of increased velocity, the inflection in the curve, or the return to a level signal corresponds to setting characteristics and also isothermal calorimeter events. The qualitative evidence implies that there is a set velocity corresponding to the Vicat set indication. This hypothesis could be validated with more planned experiments where Vicat measurements are taken frequently to capture its curve. The remaining UPV traces with the associated Vicat data are in Section C.3 of Appendix C.

The pulse velocity of the fresh Cast Stone mixes was significantly lower than the pulse velocity of either the simulant salt solution or the set Cast Stone. Robeyst (2008) calculates the impact of dissolved or entrained air on the measurement of the UPV. The impact of entrained air on the UPV or P-wave velocity measurement for cement paste is shown in Figure 6-8. This figure illustrates that a small change in the entrained air results in a significant change in the P-wave velocity or UPV. Similar calculations were performed for the SRS saltstone mixes using a bulk modulus of the salt solution calculated from the equation reported by de Korte and Brouwers (2011) using the measured density and sound velocity of the salt solution (Cozzi et al. 2013):

$$K = c^2 \cdot \rho \quad (6-1)$$

where K is the bulk modulus, Pa; c is the velocity of sound, m/s; ρ is the density of mix, kg/m³; and an estimate of the bulk modulus for the premix of 30 GPa. Figure 6-9 is the calculated effect of entrained air on the clean cap and saltstone mixes using the equations from de Korte (2011):

$$c_e^2 = \left[\left(\phi_t \frac{1}{K_f} + (1 - \phi_t) \frac{1}{K_s} \right) \times \left(\frac{\rho_f(\rho_s(\phi_t + (1 - \phi_t)S) + \rho_f S \phi_t)}{\rho_s \phi_t^2 + \rho_f(S + \phi_t(1 - \phi_t))} \right) \right]^{-1} \quad (6-2)$$

where c_e is the effective wave velocity and ρ is the density with the subscript “f” referring to the fluid, “s” to the solid, and ϕ_t to the fluid volume fractions, respectively. The parameter, S , is a shape factor approximated for spherical particles by the equation from de Korte and Brouwers (2011) by

$$S = \frac{1}{2} \left(\frac{1 + 2(1 - \phi_t)}{\phi_t} \right). \quad (6-3)$$

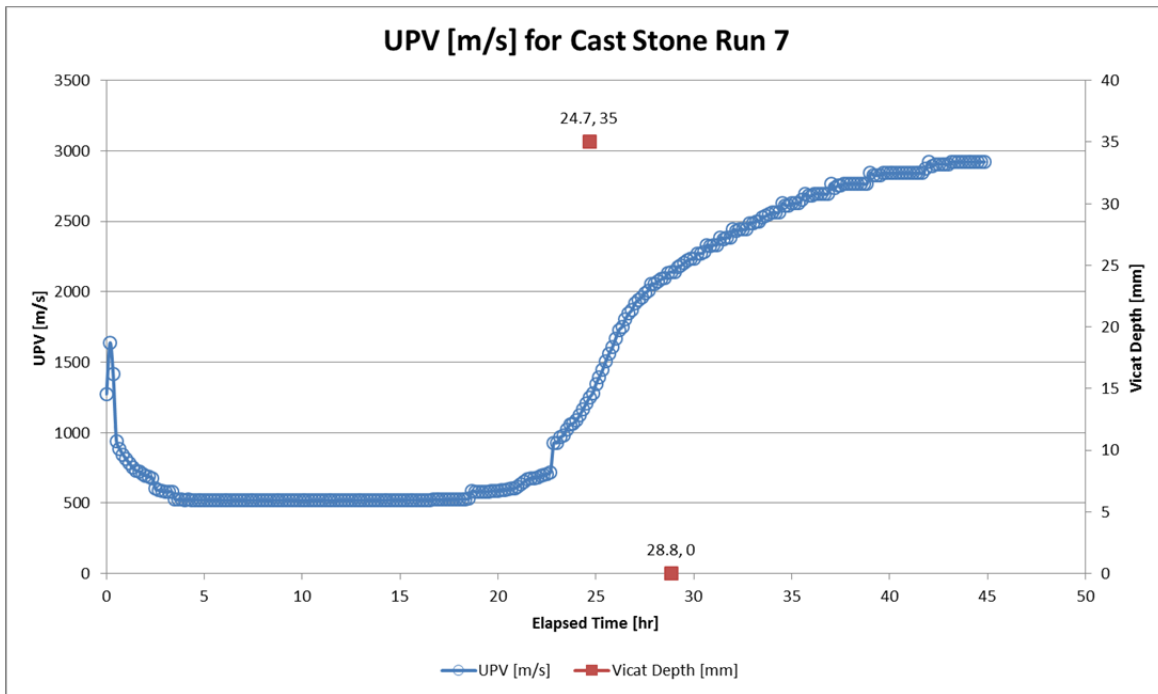


Figure 6-7. Representative UPV Data for Mix 1, RR 7

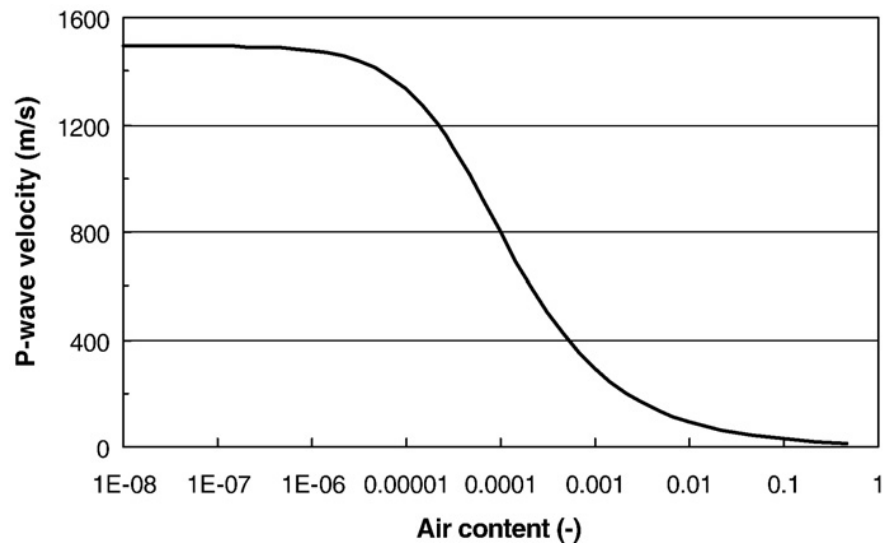


Figure 6-8. Change in UPV or P-Wave Velocity in Cement Paste (W/C=0.5) (Robeyst 2008)

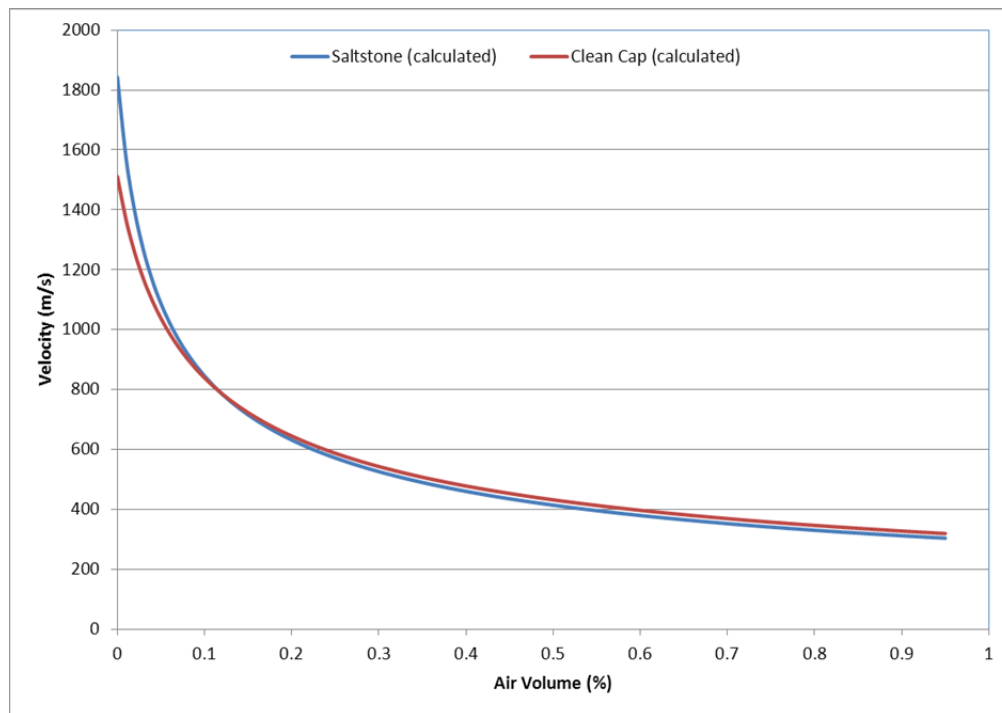


Figure 6-9. Calculated Effect of Volume of Entrained Air on Sound Velocity

6.4 Slurry Rheology

Rheological properties of freshly prepared Cast Stone specimens were measured using two different methods, shear stress as a function of shear rate and yield stress. For the first method, a Haake VT550 rotoviscometer was used to measure the flow properties of the Cast Stone slurry. The VT550 was used to obtain a flow curve (shear stress versus shear rate data) using a concentric geometry bob and cup. The data were analyzed using a Bingham Plastic rheological model (Macosko 1994), providing yield stress and plastic viscosity values. The flow curve profile used for this activity was the same one used to assess fresh Saltstone slurries. The second measurement method used a vane and cup configuration (model Haake RS6000, ThermoFisher, Waltham, MA). The vane has been extensively used by SRNL to measure the yield stress of sludges and grouts (Hansen et al. 2012). For these screening tests, the vane/cup configuration was used to determine whether it could assess the changes in the developing stress in the Cast Stone slurry as it cures. This second method is experimental and is still not fully developed.

6.4.1 Slurry Rheology Approach

All the flow curves were obtained with a rotoviscometer (Haake VT550, ThermoFisher, Waltham, MA), using the MV2 cylindrical rotor and cup configuration. The MV2 bob was selected given its range of measurement and design (e.g., the only shearing surface is the cylinder itself). The functionality of the VT550 was checked using a National Institute of Standards and Technology (NIST)-traceable N35 Newtonian viscosity oil standard at 25°C at the beginning of each day when flow curve measurements were required. The VT550 was considered functional if the resulting flow curve, analyzed as a Newtonian fluid, was within $\pm 10\%$ of the NIST viscosity value. The flow curve used to quantify the rheological properties of the slurry is shown in Table 6-2. These measurements were obtained at the temperature of the slurry (i.e., the temperature as measured at the end of the mixing activities). This flow curve was developed for

the Saltstone slurries analyzed by SRNL given gel time issues, hence the short time to obtain the flow curve profile. Potential issues are over-estimating the properties on the up curve and under-estimating the properties on the down curve due to the MV2 inertia effects, if the fluid is very thin. However, these issues were not observed for this data set. Thixotropic response was expected, given that some of the slurries will start developing structure when shearing (mixing) stops.

Table 6-2. Flow Curve Profile Using MV2 Geometry

Shear Rate and Time of Measurement		
Up Curve	Hold	Down Curve
0 to 300 s ⁻¹ linearly in 2 minutes	300 s ⁻¹ for 30 seconds	300 to 0 s ⁻¹ linearly in 2 minutes

The Haake software converts the rotational rate of the rotating surface of the bob into a shear rate assuming the material being measured is a Newtonian fluid. This shear rate and range are when modeling the flow curve as a Bingham Plastic fluid. Both the up curve and down curve are fitted to the Bingham Plastic model

$$\tau = \tau_o + \eta_{\infty}\dot{\gamma} \quad (6-4)$$

where

- τ = measured shear stress (Pa)
- τ_o = Bingham Plastic Yield Stress (Pa)
- η_{∞} = plastic viscosity (Pa · s)
- $\dot{\gamma}$ = shear rate (1/s).

The Haake RS6000 was used to perform the vane measurements. The functionality of the RS6000 was checked using a NIST-traceable N35 Newtonian viscosity oil standard at 25°C, using the Z38 bob and cup configuration, at the beginning of each day when vane measurements were required. The RS6000 was considered functional if the resulting flow curve, analyzed as a Newtonian fluid, was within ±10% of the NIST viscosity value. The vane used in this task was the Haake FL22 vane (Figure 6-10), and the vane was positioned such that all the geometric requirements specified on the left side of Figure 6-10 were satisfied. Unlike normal vane measurements, where the rotational speed can range from 0.01 to 1 revolution per minute (rpm), the vane was rotated at one revolution per hour (rph) for up to two hours. This very slow rotational rate allows for the Cast Stone structure to develop. Measurements were obtained every second. The measurement was stopped when the shear stress reached 200 Pa or the program reached completion at two hours. The reported yield stress data were related to the gel data, when the Cast Stone was considered gelled.

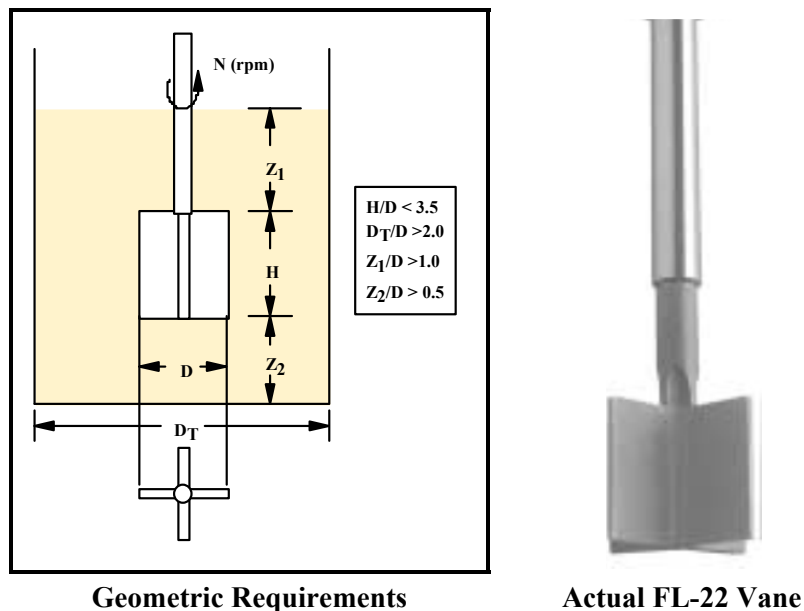


Figure 6-10. Vane Requirements and Actual Vane

6.4.2 Slurry Rheology Results

Figure 6-11 shows the plastic viscosity results for the 38 Cast Stone mixes in Table C-1. As expected, the mixes with the 0.4 mix ratio are more viscous than the mixes with the higher water content at a mix ratio of 0.6. Plastic viscosities were in the range of 220 to 580 centipoise (cP) for the 0.4 mixes and 54 to 165 for the 0.6 mixes. There appears to be a smaller trend with the mixes prepared with the SE fly ash being more viscous than those prepared with the NW fly ash. No data were obtained for Mix 1 because it had set before the test could be completed. Results of the plastic viscosity and yield stress measurements are tabulated in Table C-5 of Appendix C.

Figure 6-12 shows the Bingham Plastic yield stress for the 38 Cast Stone mixes. The mixes with the 0.4 mix ratio generally have higher yield stresses when compared to the mixes with the 0.6 mix ratio. Yield stresses were in the range of 10 to 92 pascals for the 0.4 mixes and 2 to 24 pascals for the 0.6 mixes. The highest yield stresses were associated with the mixes with the 0.4 mix ratio and 5M sodium simulants.

6.5 Heat of Hydration

The isothermal heat of hydration for the Cast Stone mixes was measured in accordance with ASTM C1679, *Standard Method for Measuring Hydration Kinetics of Hydraulic Cementitious Mixtures Using Isothermal Calorimetry* (ASTM C1679 2009). This measurement is used to compare the hydration kinetics of salt solutions and dry mix blends. Salt solution components can affect either the energy produced during hydration, or to a greater extent, the time frame over which the energy is released. The composition of dry blend components and composition and amount of additives can also affect the magnitude and timing of hydration heat development. In large pours, the energy (heat) produced can alter the mineralogy and microstructure developed in the waste form and influence cured properties.

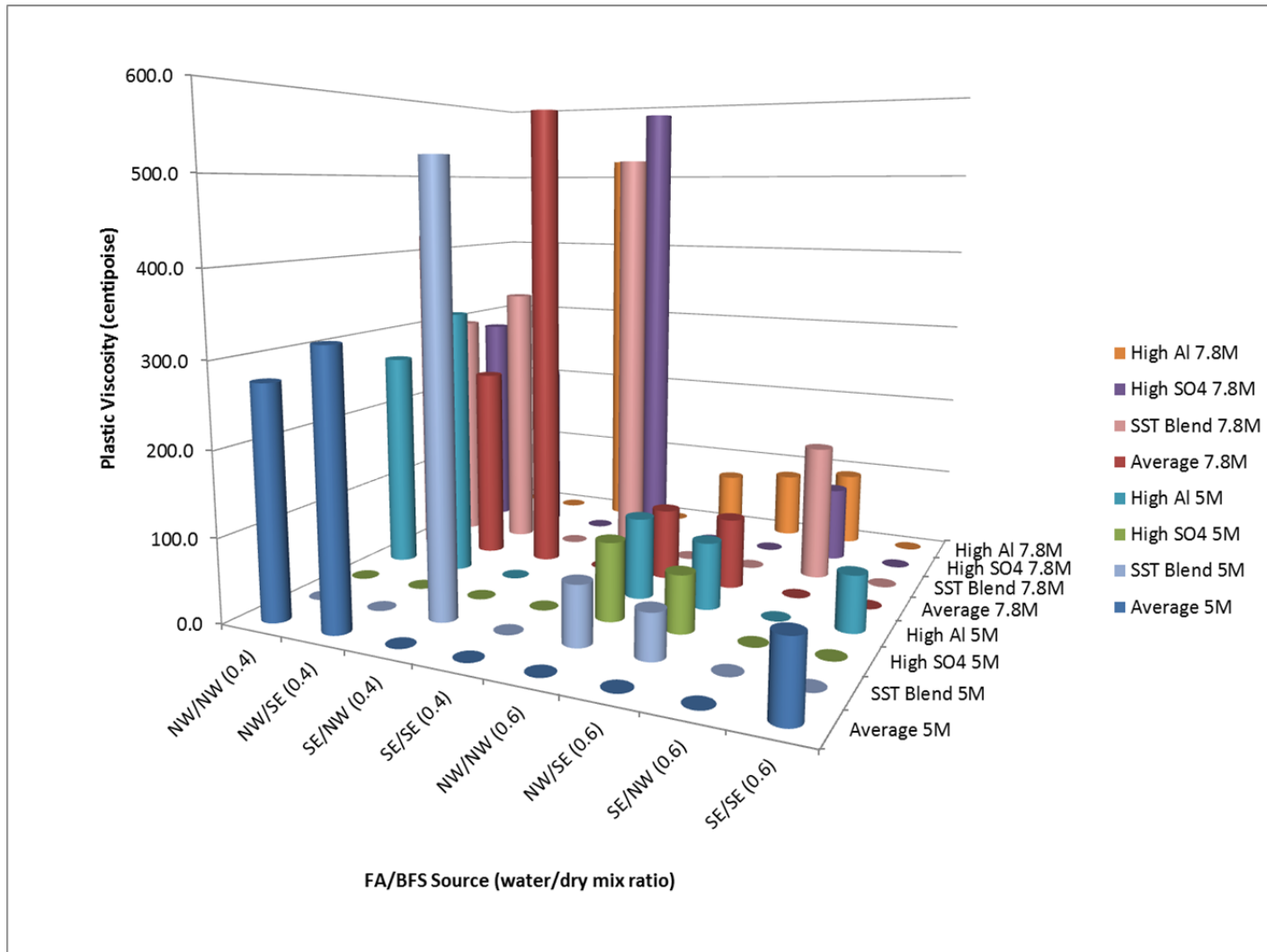


Figure 6-11. Results of Plastic Viscosity Measurements for the 38 Cast Stone Slurries

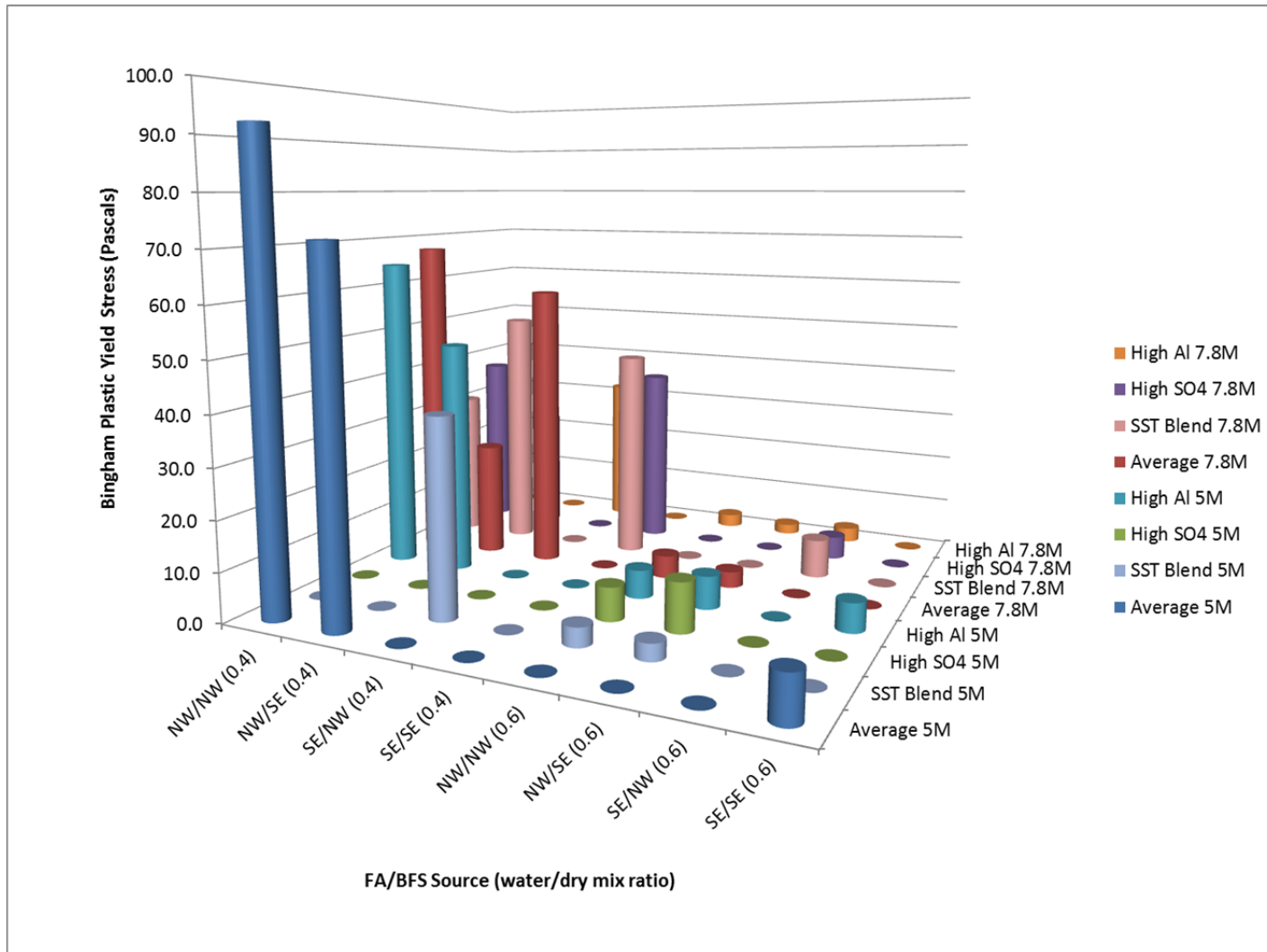


Figure 6-12. Bingham Plastic Yield Stress for the 38 Cast Stone Slurries

6.5.1 Heat-of-Hydration Approach

An eight-channel isothermal calorimeter (TAM Air, TA Instruments, Newcastle, DE) was used to collect the heat generation rate and total energy generated for each of the mixes. Each channel consists of a twin configuration with one side for the sample and the other side for the reference material. Figure 6-13 depicts the configuration of one of the calorimeter channels. The reference ampoule was balanced with 20 g of quartz sand to approximate the heat capacity of the Cast Stone mixes. The isothermal calorimeter was maintained at 25°C for all of the testing.

An 18-g sample (dry blend + salt solution) of each of the Cast Stone screening test compositions was mixed using resonant acoustic mixer (LabRAM, Resodyne™ Acoustic Mixers, Inc., Butte, MT). After the dry blend was incorporated into the waste simulant solution, the materials were mixed for an additional minute. The mix was transferred to the calorimeter and the test initiated. After 300 hours, the test was terminated. The total energy produced, normalized per gram of dry blend material, was determined at 300 hours. The maximum heat generation rate (heat flow) and the elapsed time to attain this rate were also determined. Figure 6-14 shows examples of total energy and heat flow curves.

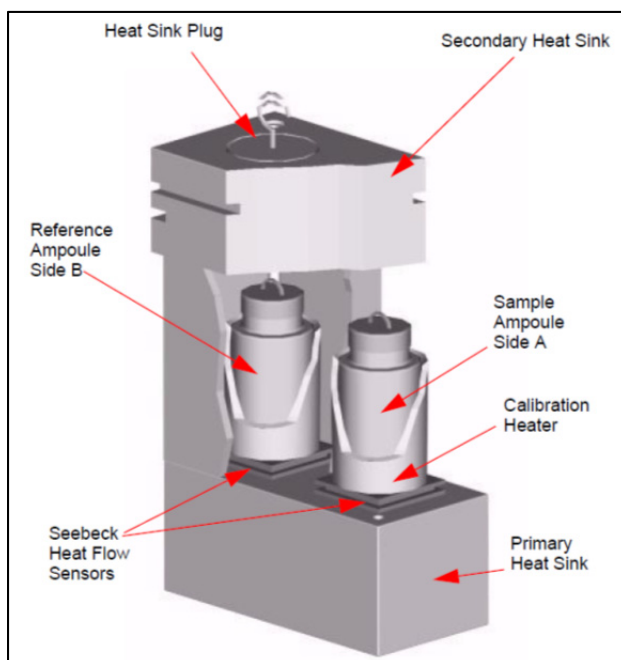


Figure 6-13. Cutaway View of a Single Channel in the Isothermal Calorimeter from TAM Air Manual, TA Instruments.

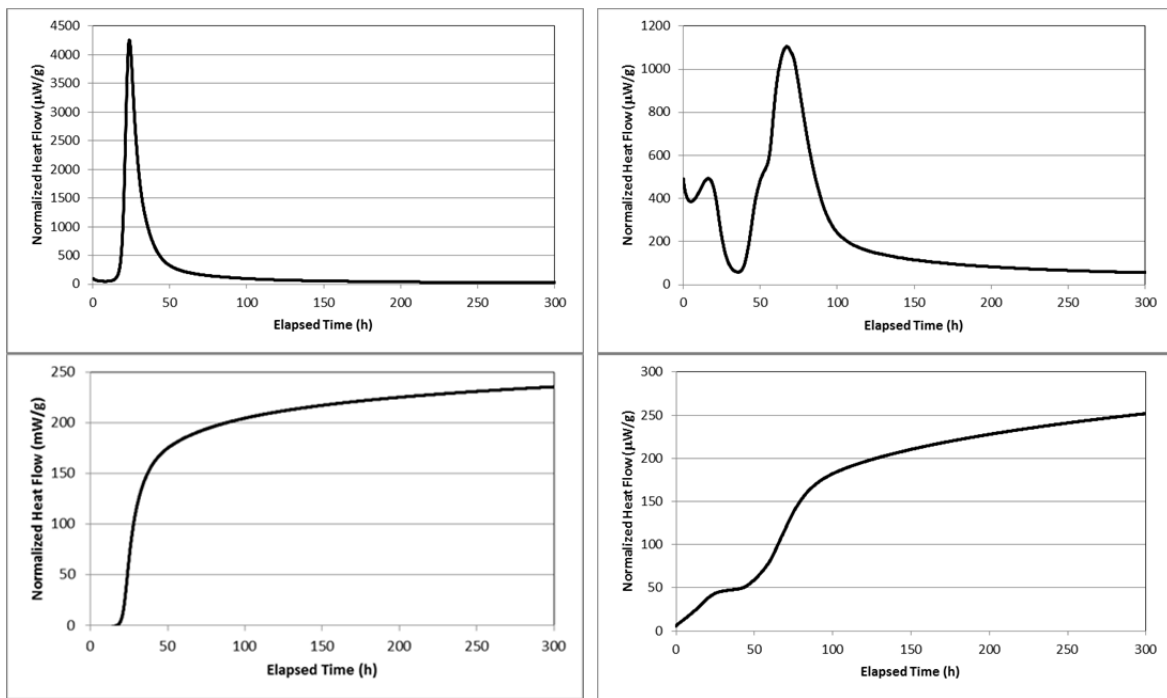


Figure 6-14. Normalized Heat Flow for Two Mixes Analyzed Using Isothermal Calorimetry. Left: Mix 1, RR7; Right: Mix 19, RR15.

6.5.2 Heat-of-Hydration Results

The heat-of-hydration results include the total energy generated over 300 hours of testing, the maximum heat flow, and the elapsed time to reach the maximum heat flow. These properties are referred to as “Heat Generation at 300 Hours”, “Heat Generation at Peak”, and “Time to Peak Heat Generation” in the balance of the report. The heat generation results for the 38 Cast Stone mixes are tabulated in Table C-8 of Appendix C. Figure 6-15 shows the heat generated over the 300 hours for the 38 Cast Stone mixes. The heat generated was in the range of 150 to 434 joules/gram (J/g) of dry blend material. There appears to be a trend of higher heat generation for the 0.6 mix ratio mixes compared with the 0.4 mix ratio mixes. Figure 6-16 shows the time to reach the peak heat generation rate. The time to peak ranged from 16 to 164 hours.

6.6 Fresh Density

The density of freshly prepared Cast Stone was measured with mini weight per gallon sample cups (model #WG-SS-8.32, Gardco, Pompano Beach, FL) using a simplified ASTM D1475, *Standard Test Method For Density of Liquid Coatings, Inks, and Related Products* (ASTM D1475 1998). The density of fresh grout can also be calculated from density measurements of the individual dry materials and the salt solution and the known percentages of each component in the final mix. However, the fresh density can be affected by air entrainment; thus, the actual measurement is made using the ASTM D1475 method.

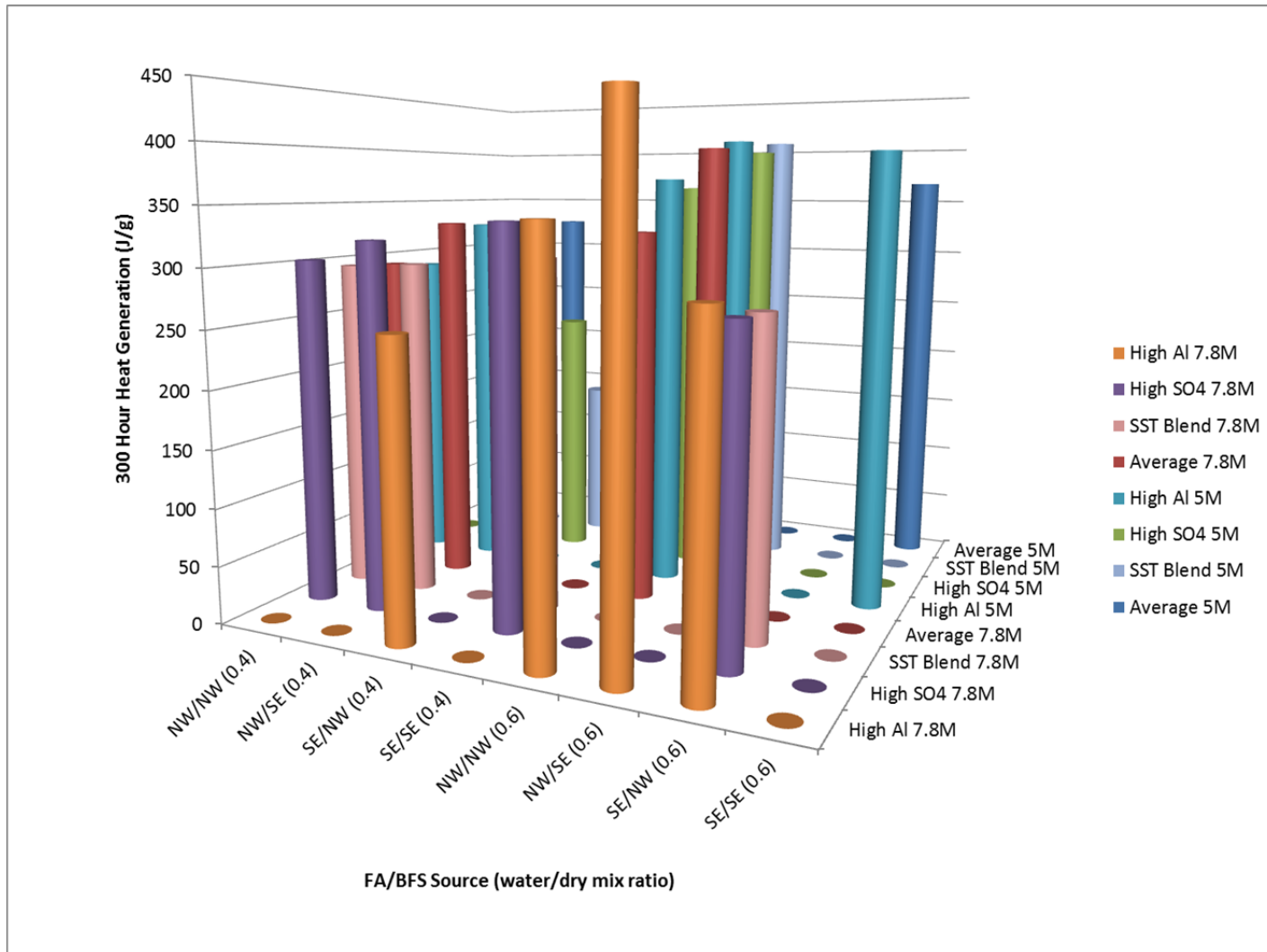


Figure 6-15. Heat Generation at 300 Hours (J/g) for 38 Cast Stone Mixes

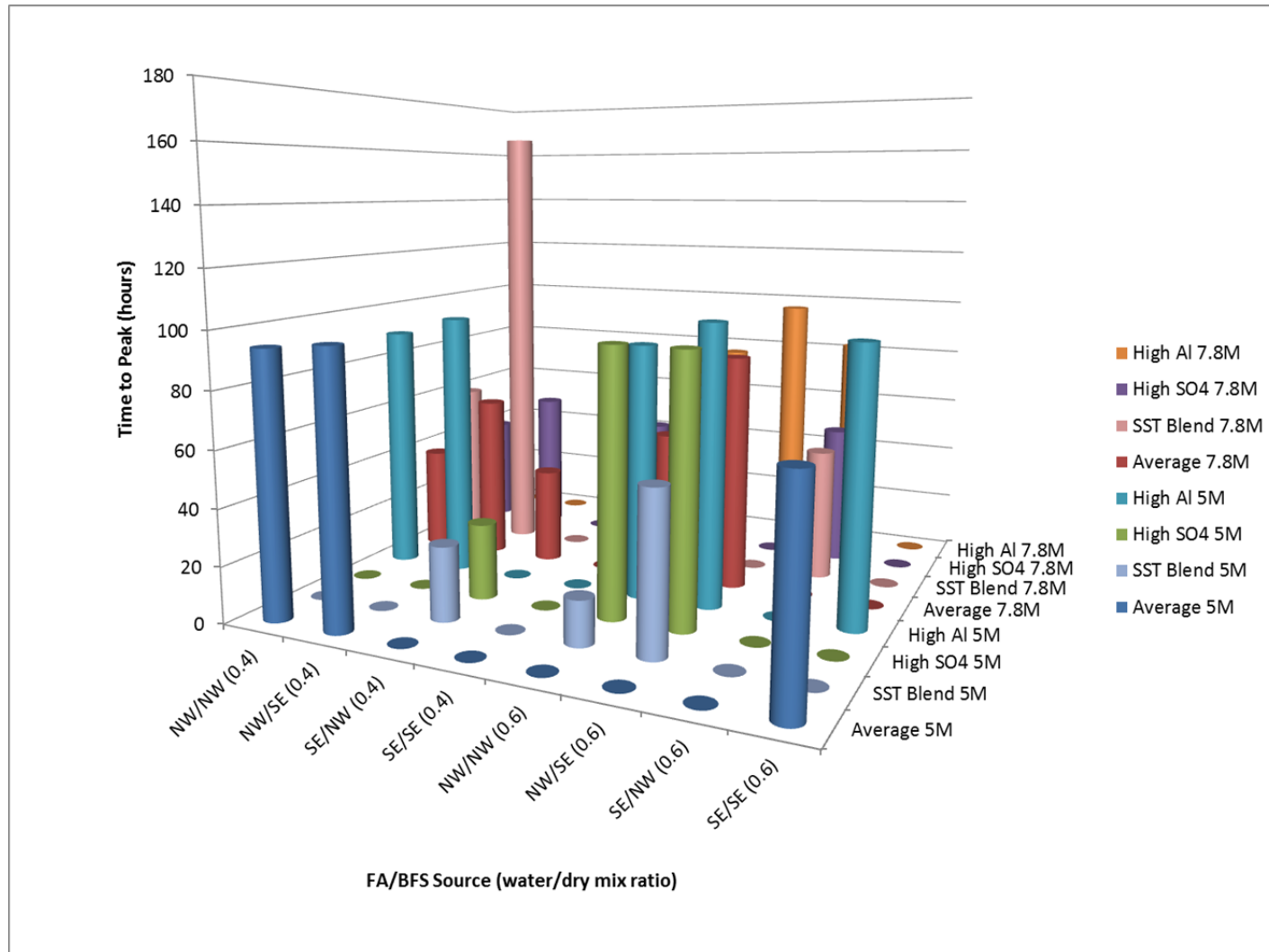


Figure 6-16. Time to Peak Heat Generation (hours) for 38 Cast Stone Mixes

6.6.1 Fresh Density Approach

Prior to testing, the volume of the sample cup was verified with ASTM Type I water at room temperature following the calibration steps in the ASTM D1475 method. After the initial calibration check, only the tare weight of the cup was recorded assuming that the volume of the stainless steel cup remained constant throughout the testing period.

To measure the fresh density, the sample cup was filled with fresh slurry to form a meniscus. The container was capped and the excess material expressed from the overflow was wiped away. The sample cup was wiped to remove any material from the outer surfaces and then was placed on a balance to obtain the mass of the sample. The fresh density is calculated from the mass of the sample divided by the known volume of the sample cup using the equation

$$\rho = \frac{W-w}{V} \quad (6-5)$$

where

ρ	=	density of the fresh slurry, g/ml
W	=	mass of the filled sample cup, g
w	=	mass of the empty sample cup, g
V	=	volume of the sample cup, ml.

6.6.2 Fresh Density Results

Figure 6-17 shows the measured fresh densities for the 38 Cast Stone mixes in Table C-1. The results are tabulated in Table C-9 of Appendix C. The fresh densities are highest for the 0.4 mix ratio mixes and are in the range of 1.87 to 1.96 g/cm³. Fresh densities for the 0.6 mix ratio mixes are in the range of 1.73 to 1.83 g/cm³.

6.7 Free Liquids

Free liquids (standing water) were determined by measuring the residual liquid remaining after predetermined curing time periods, typically 24 hours and 3 days. The volume of the liquid was calculated from the mass of the recoverable liquid. The density of the liquid was either measured or assumed to be the same as the waste simulant salt solution used to prepare the Cast Stone mix. The density of standing water remaining after 3 days was previously determined to be similar to that of the initial salt solution (Hansen et al. 2006). The standing water calculation is reported as the volume of fluid collected over the volume of hardened Cast Stone. The presence of standing water is a preliminary indication that settling may have occurred within the mix. This may or may not be an indication of preferential settling (segregation). The standing water generally is reabsorbed into the Cast Stone waste form with time (e.g., within a few days).

6.7.1 Free Liquids Approach

The method ASTM C232 *Standard Test Methods for Bleeding of Concrete* (ASTM C232 2004) was modified to accommodate the smaller test samples available and the longer time associated with gel and set. To measure the residual free liquids for the prepared Cast Stone slurries, fresh slurry was poured into two ~100-ml snap-top vials (#VL25H, LA Container, Yorba Linda, CA) to approximately three-quarters full. If the slurry did not self-level, the vial was tapped to level the surface. The vials were capped and placed in a zip-top bag with a moist cloth to mitigate evaporation. The bagged vials were set upright to maintain a level surface. After approximately 24 hours of setting, the vials were removed from the bag and inspected for free liquids. If liquid

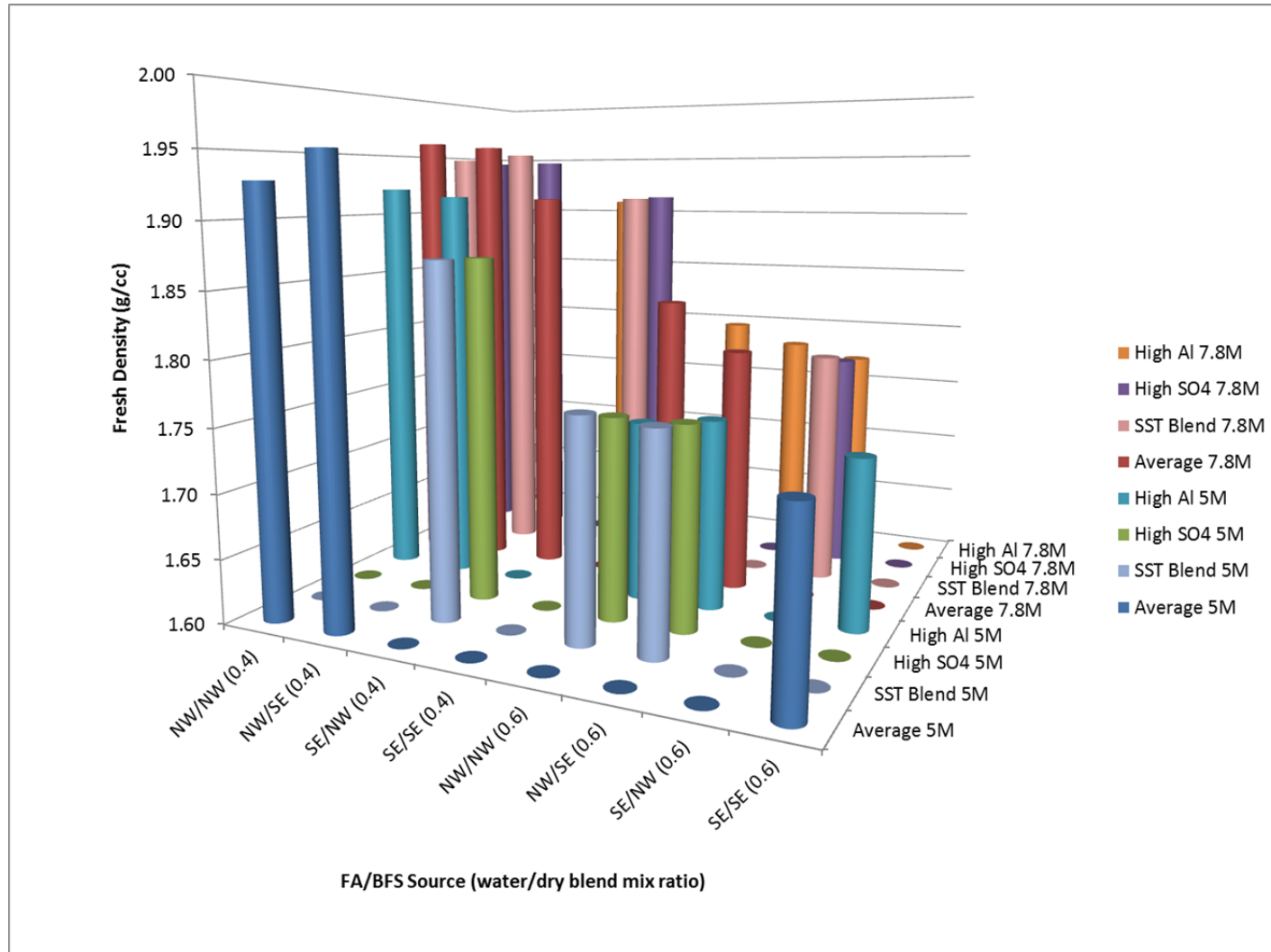


Figure 6-17. Fresh Densities for 38 Cast Stone Mixes

was present, the vial was wiped dry and weighed. A tared pipette was used to draw off the free liquid. The pipette was weighed with the liquid. The liquid was returned to the vial and the vial was reweighed, capped, and returned to the bag. The process was repeated after an additional 2 days of setting (3 days from casting).

6.7.2 Free Liquids Results

Five of the 38 screening test mixes prepared resulted in samples with free liquids after 24 hours. Three of the mixes (Mixes 17, 23, and 32) all produced free liquid that persisted to 3 days. Free liquids generated by the other two mixes were reabsorbed between the 1-day and 3-day measurement. Table 6-3 shows the free liquid results for the five mixes generated from duplicate samples cast from the same mix after 1 and 3 days of setting. The full set of data for all 38 mixes is listed in Table C-10 of Appendix C.

Table 6-3. Cast Stone Mixes Generating Free Liquids

Mix Number	Random Run Order	Free Liquid (Vol%)			
		Day 1		Day 3	
		Duplicate 1	Duplicate 2	Duplicate 1	Duplicate 2
12	29	1.09	1.11	0.00	0.00
9	31	0.45	0.40	0.00	0.00
23	20	2.33	2.39	2.43	2.20
16	36	0.84	0.68	0.00	0.00
17	25	0.52	0.45	0.28	0.21
32(12a)	8	5.84	5.91	4.47	4.46

7.0 Cast Stone Cured Waste Form Characterization

After curing for 28 days, the Cast Stone waste form monoliths were characterized with respect to chemical composition, compressive strength, density, TCLP, and contaminant leachability. The characterization work was conducted at both PNNL and SRNL. Porosity data are in Appendix D.

7.1 Chemical Composition

This section provides the chemical compositions of the Cast Stone monoliths as measured at PNNL for the 26 radioactive Cast Stone mixes used in the EPA Draft Method 1315 leach tests. The chemical composition of each of the 26 Cast Stone mixes was determined by dissolving a piece from a hardened (cured ~28 days) cylinder of each of the 26 radioactive batches using microwave digestion. The resulting solutions were characterized using inductively coupled plasma-mass spectroscopy (ICP-MS) and inductively coupled plasma-optical emission spectroscopy (ICP-OES) (see Appendix B for a description of the analytical methods). The first three columns of Table 7-1 show the overall averages and ranges for the compositions of the 26 Cast Stone mixes. Appendix D contains the analyzed composition of each of the mixes.

Table 7-1 also shows the average compositions for subsets of the 26 mixes segregated by different parameters in the screening matrix, including the Na molarity of the simulants, dry blend mix, and the High Al simulant. Within the analytical uncertainties, the Na concentrations are higher for the monoliths prepared with the 7.8 M Na simulants than for the 5 M Na simulants, but the ratio is only 1.15 compared to the value of 1.56 that one can calculate from the ratio of the 7.8 M Na simulants and 5.0 M Na simulants. The Na in the cured Cast Stone is tempered by the Na present in the NW fly ash, which contains about an order of magnitude more sodium than the SE fly ash (see Table 4-2). The average Al concentrations in the three samples prepared with the High Al simulants at 5 M Na are lower than the average for the three samples prepared with the Average simulants at 5 M Na. This is because the Al variability is driven more by the variability in the Al concentrations among the dry blend ingredients, as seen in the columns in Table 7-1 showing the average compositions for the monoliths prepared with the four different dry blend mixes, than by the Al variability in the simulants.

7.2 Compressive Strength

Compressive strength was measured for the Cast Stone on as-cured monoliths after 28 days at both SRNL and PNNL. All compressive strength tests were conducted on nonradioactive monoliths prepared at both laboratories. After curing for a minimum of 28 days, three 2-in.-diameter × 4-in.-high cylinders of each Cast Stone mix were subjected to the ASTM C39 *Standard Test Method for Compressive Strength of Cylindrical Concrete Specimens* (ASTM C39 2012).

7.2.1 Compressive Strength Approach

According to the ASTM C39 test method, a sample is loaded into the testing apparatus so that the axis of the monolith is aligned with the center of thrust of the testing apparatus. Before testing the monolith, the load indicator is set to zero. The load is applied continuously without any shock at a stress rate of 0.25 ± 0.05 MPa/s (35 ± 7 psi/s). The designated rate of loading should be maintained at least during the latter half of the anticipated loading phase. The loading is maintained until the load indicator starts to decrease steadily, and the monolith displays a well-defined fracture pattern as illustrated in the C39/C39M test method. The compressive strength is calculated by dividing the maximum load imposed on the monolith during the test by the average cross-sectional area of the monolith. The result is typically expressed to the nearest 0.1 MPa or 10 psi.

Table 7-1. Summary Table of Chemical Compositions of the 26 Mixes Spiked with Radionuclides and Subsets of the 26 Mixes

Analyte	Units	Average	Max	Min	Average 5M Na	Average 7.8M Na	Hi Al 5M Na	Hi Al 7.8M Na	NW FA, NW BFS	NW FA SE BFS	SE FA NW BFS	SE FA SE BFS
		Mixes										
		All	All	All	2,13,20	3,5,6,22	4,17,24	9,11,19	5,15,16, 25,10,24	4,20 3,9,12, 22	1,6,8,19 11,21,23, 26	7,14,18 2,13,17
Aluminum	µg/g	62,400	83,900	42,500	63,800	61,300	59,400	66,500	56,400	48,700	72,300	68,800
Barium	µg/g	1,360	2,280	575	1,340	1,370	1,540	1,020	2,100	2,010	828	682
Cadmium	µg/g	10	13	6	10	10	-	12	10	13	11	6
Calcium	µg/g	139,000	167,000	115,000	142,000	136,000	135,000	129,000	157,000	137,000	137,000	125,000
Cesium	µg/g	4	5	2	4	3	3	4	3	2	4	5
Chromium	µg/g	574	976	246	496	631	503	701	560	633	574	528
Iron	µg/g	19,300	25,600	13,900	20,100	18,700	18,300	18,300	18,200	16,200	21,100	21,100
Lead	µg/g	57	80	29	52	60	46	67	48	42	70	60
Magnesium	µg/g	21,000	36,600	8,860	22,700	19,800	24,800	15,600	16,500	32,000	10,900	28,100
Manganese	µg/g	990	1,570	465	1,070	931	1,080	795	765	1,360	640	1,310
Mercury	µg/g	12	12	12	12	-	-	-	-	-	12	-
Nickel	µg/g	-	-	-	-	-	-	-	-	-	-	-
Phosphorus	µg/g	-	-	-	-	-	-	-	-	-	-	-
Potassium	µg/g	12,600	13,200	11,500	12,300	13,200	13,000	13,200	-	-	12,400	13,000
Silicon	µg/g	149,000	172,000	127,000	155,000	144,000	149,000	136,000	142,000	148,000	145,000	163,000
Silver	µg/g	6	8	4	6	-	-	-	-	-	8	4
Sodium	µg/g	72,200	101,000	43,100	66,400	76,500	75,700	86,800	72,300	76,900	74,400	64,700
Strontium	µg/g	944	1,330	577	952	937	985	802	1,247	1,162	757	671
Titanium	µg/g	3,430	4,740	2,300	3,570	3,320	3,180	3,340	3,070	2,690	3,900	3,890
Technetium-99	µg/g	7	10	4	6	8	6	8	7	7	8	7
Iodine-127	µg/g	68	149	25	62	73	61	65	65	70	79	56
Uranium-238	µg/g	144	1,050	46	87	185	89	123	107	244	125	104
% Dry Solids	% by Weight	77	83	72	75	78	75	77	78	77	77	77
- = Not detected.												

At PNNL, the compressive strength tests were conducted using the servo-hydraulic universal test machine (MTS servo-hydraulic system with Instron® data acquisition and control system) shown in Figure 7-1. This apparatus has a maximum load capacity of 20 kilo-pound force (kips-force), which is equivalent to 88.96 kN (kilo-newtons). The compressive load was applied until the complete fracture of the specimens was observed. The loading rate was set at 0.25 MPa/s (29.4375 kN/min) as specified by ASTM C39 (ASTM C39 2012). Three cylinders (2-in. diameter by 4-in. long) of nonradioactive Cast Stone specimens were tested for each of the 26 test mixes. Before testing, some of the cylinders were capped with an epoxy to provide a smooth surface. In all tests, unbonded neoprene caps were used as described in ASTM C1231, *Standard Practice for Use of Unbonded Caps in Determination of Compressive Strength of Hardened Concrete Cylinders* (ASTM C1231 2012). Commercially available neoprene pads with a Shore A Durometer Hardness of 70 were used in the tests. The pads were inspected prior to each test and replaced more frequently than the maximum 50 reuses allowed in the ASTM method.



Figure 7-1. The Compressive Strength Test Apparatus

At SRNL, the compressive strength tests were conducted using a hydraulic compression test machine (Humboldt Manufacturing, Schiller Park, IL, model #HCM-0300 with Test Mark Industries, LXI data acquisition system). This apparatus has a maximum load capacity of 300 kilo-pound force (kips-force), which is equivalent to 1334 kN. The compressive load was applied until the load indicated by the equipment was reduced to 75% of the maximum load applied to the specimen. The loading rate was set at approximately 0.25 MPa/s (29.4 kN/min) as specified by ASTM C39 (ASTM C39 2012). Three cylinders (2-in. diameter by 4-in. long) of

nonradioactive Cast Stone specimens were tested for 14 of the 26 screening test mixes in Table 2-1. Compressive strength was also measured for the 12 additional mixes shown in Table 6-1. In all tests, unbonded neoprene caps were used as described in ASTM C1231 (ASTM C1231 2012). Commercially available neoprene pads with a Shore A Durometer Hardness of 70 were used in the tests. The pads were inspected prior to each test and replaced more frequently than the maximum 50 reuses allowed in the ASTM method.

7.2.2 Compressive Strength Results

The results of the compressive strength measurements are shown in Figure 7-2 and Figure 7-3 for the PNNL and SRNL tests, respectively. These are the means of triplicate measurements. Note that the colored circles in the figures at zero compressive strength represent conditions that were not tested. Compressive strengths of the triplicate samples ranged from 5.8 to 62.0 MPa (850 to 8990 psi), excluding one sample that broke at 1.3 MPa (190 psi). The other two monoliths from that mix (#23) broke at 11.6 and 9.4 MPa (1683 and 1360 psi). For the 14 mixes tested by both PNNL and SRNL, the PNNL compressive strength measurements tended to be slightly higher than the SRNL results. The target minimum compressive strength is 3.4 MPa (500 psi). The results are tabulated in Table D-2 and Table D-3 of Appendix D. Those tables also present the means and SDs of the values over the triplicate samples. The lower compressive strengths tended to be from mixes with the 7.8M Na simulants and the 0.6 water-to-dry-blend mix ratio. The higher compressive strengths tended to be from mixes with the 5 M Na simulants and/or those with the 0.4 water-to-dry-blend ratio. Mix 26 at PNNL was prepared with 14% less dry blend than planned (0.7 water-to-dry-blend ratio). At an average of 6.5 MPa, the Mix 26 specimens still passed the minimum compressive strength requirement, though the compressive strength was lower than that of its replicate, Mix 21 (9.4 MPa).

7.3 Cured Density

The densities of the cured Tc and U-spiked Cast Stone samples produced at PNNL were determined using the measured weight and physical dimensions of each of the 28-day cured specimens. At SRNL, the cured density was measured on fractured pieces from compression specimens using helium pycnometry.

7.3.1 Cured Density Approach

To determine the cured density of each monolith, the diameter was measured at the top, middle, and bottom of each monolith. Then the overall length was measured at three locations. The average value of the diameter measurements and the average value of the overall length measurements were used to determine the volume of the monolith. Each monolith was weighed to determine its mass. The mass of the monolith was divided by the determined volume resulting in the cured density. At SRNL, a gas pycnometer (model # MVP-5DC, Quantachrome Corp., Boyton Beach, FL), using helium as the fill gas was used to measure the volume of the specimen using procedure Manual L29, ITS-0168. A piece from each of the compression samples was weighed and the volume measured in triplicate. The density of the sample was calculated from the measured mass and the average of the triplicate volume measurements. The reported measured density for each mix was the average of the calculated density of the triplicate compression samples.

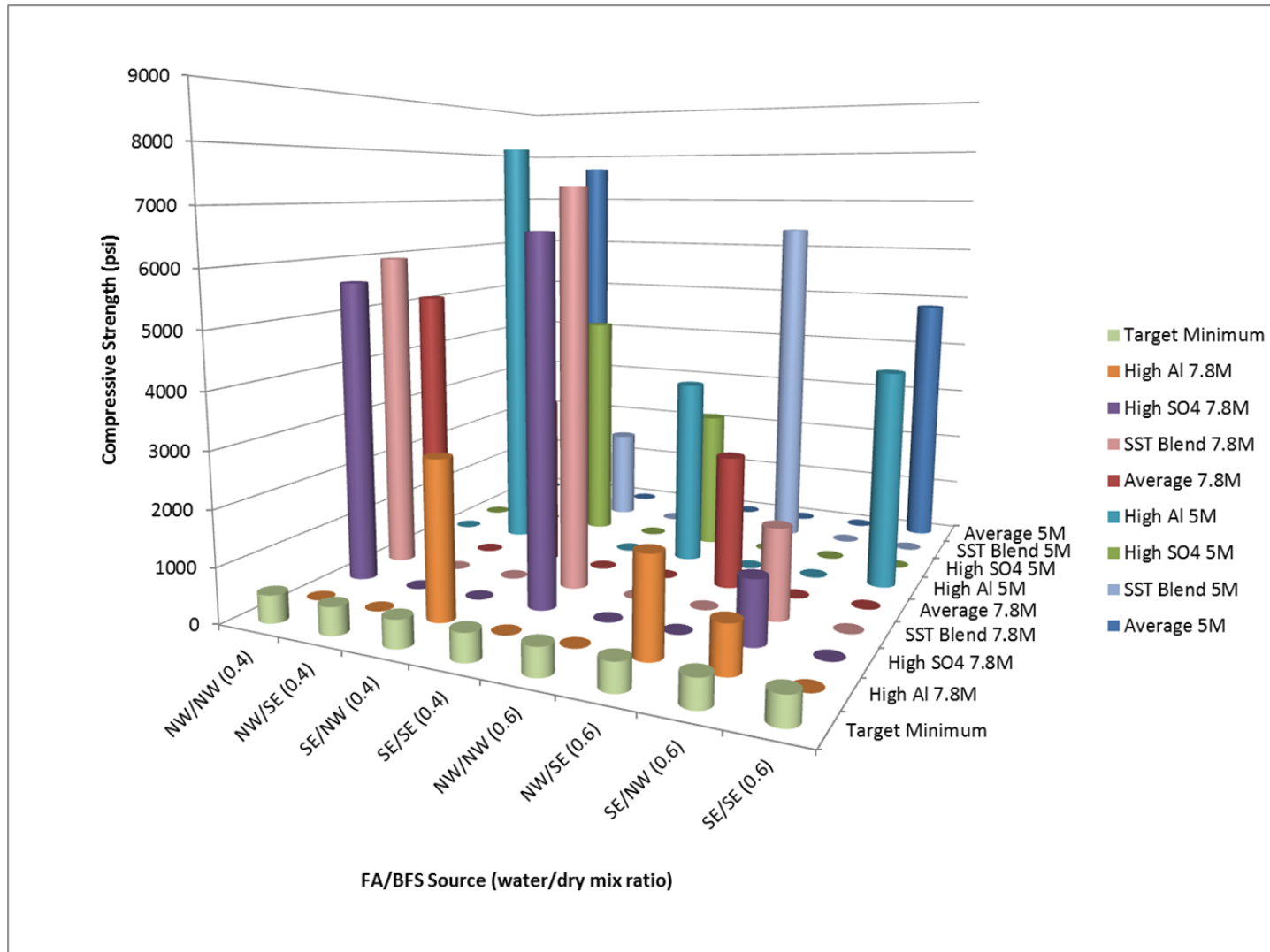


Figure 7-2. Results of PNNL Compressive Strength Measurements for 26 Cast Stone Mixes

7.3.2 Cured Density Results

Densities were measured by PNNL on each of six specimens from each of the 26 Cast Stone mixes in Table 2-1. Densities ranged from 1.68 to 2.04 g/cm³. Figure 7-4 shows the averages of the six measurements and Appendix D has a tabulation of all of the values (see Table D-4). Note that the “flat” colored circles in the figure at densities <1.7 g/cm³ represent conditions that were not included in the screening test matrix. Table D-5 in Appendix D also lists the densities measured using a pycnometer at SRNL. Generally, densities were higher for the 0.4 water-to-dry-solids mix ratio mixes than for the 0.6 water-to-dry-solids mix ratio mixes. There is a less pronounced trend for higher densities with the 7.8 M Na simulants than for the 5 M Na simulants.

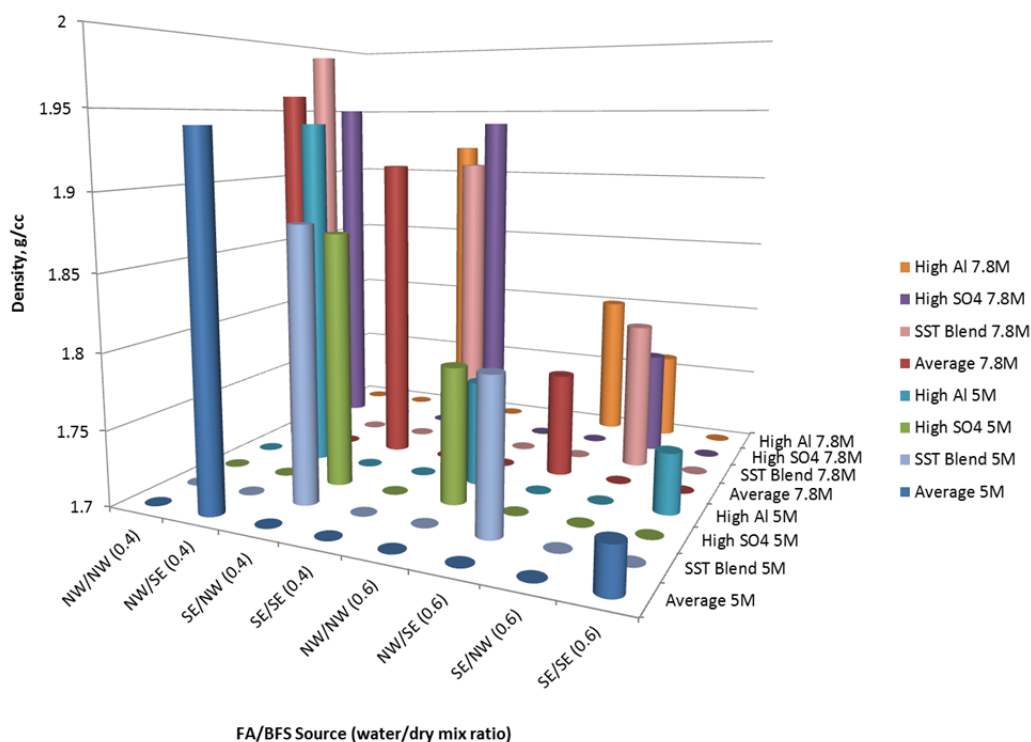


Figure 7-4. Average Densities of PNNL Cast Stone Monoliths

7.4 Toxicity Leaching Characteristic Procedure

The TCLP, EPA Method 1311 (EPA 1992), was conducted to demonstrate that the Cast Stone screening test formulations will meet RCRA land disposal restrictions for hazardous wastes. The LAW is projected to include RCRA metals including As, Ba, Cd, Cr, Pb, Hg, Se, and Ag; and underlying hazardous constituents (UHCs) including Sb, Be, Ni, and Tl. In addition, some of the dry materials may include these same and other hazardous materials. The results are compared with the UTS limits in 40 CFR 268, *Land Disposal Restrictions*, as shown in Table 7-2.

7.4.1 TCLP Methodology

TCLP testing was conducted at the Southwest Research Institute (SwRI) in San Antonio, Texas. SwRI is one of PNNL's evaluated suppliers and the work was conducted in accordance with the Hanford HASQARD requirements. A 2-in.-diameter by 4-in.-long cylinder of each of the 26 Cast Stone test mixes prepared at PNNL was sent to SwRI with the appropriate chain of custody.

Table 7-2. Universal Treatment Standards from 40 CFR 268

Constituent Category	Regulated Constituent	Allowable Concentration in TCLP Leachate (mg/L)
RCRA Metals	Arsenic	5.0
	Barium	21
	Cadmium	0.11
	Chromium	0.60
	Lead	0.75
	Mercury	0.025
	Selenium	5.7
	Silver	0.14
	Antimony	1.15
Underlying Hazardous Constituents	Beryllium	1.22
	Nickel	11
	Thallium	0.20

In the EPA 1311 method, for wastes containing $\geq 0.5\%$ solids, the liquid, if any, is separated from the solid phase and stored for later analysis. The Cast Stone samples were 100% solid. The particle size of the solid phase is reduced by crushing to a particle size less than 1 cm. The solid phase is extracted with an amount of extraction fluid equal to 20 times the weight of the solid phase. The extraction fluid used is a function of the alkalinity of the solid phase of the waste. For the Cast Stone samples, extraction fluid #2 was used. This is a mixture of 5.7 ml of glacial acetic acid ($\text{CH}_3\text{CH}_2\text{OOH}$) per liter of water. The pH of this extraction fluid is 2.88. After extraction, the liquid extract is separated from the solid phase by filtration through a 0.6- μm to 0.8- μm glass fiber filter. The resulting solution is chemically analyzed for the RCRA metals and UHCs.

7.4.2 TCLP Results

The results of the TCLP testing are shown in Table 7-3 (note that the table is populated with only values above the reporting [detection] limit). As described in Section 3.3, the simulants used in the Cast Stone specimens were spiked with only Cr, Pb, Ni, and Cd. Neither Pb nor Cd was detected in any of the TCLP extracts. Chromium and Ni were measured in most but not all of the extracts. The As, Ba, and Se were not included in the simulants. They are present in the dry materials used to make the Cast Stone. Even with a 100% contribution from the dry materials, the TCLP results for As and Se would be below the UTS limits. For the values in Table 7-3, all of the 26 Cast Stone mixes easily met the UTS limits.

7.5 Effective Diffusion Coefficients

The Cast Stone monoliths were subjected to the draft EPA Draft Method 1315, *Mass Transfer Rates of Constituents in Monolithic or Compacted Granular Materials Using as Semi-Dynamic Tank Leaching Test* (EPA 2012).¹ Two monoliths for each of the 26 screening test formulations

¹ These screening tests used the EPA Draft Method 1315 cited. As this report was being prepared, the EPA 1315 method became a formally accepted EPA method. It is available on the EPA website at http://www.epa.gov/epawaste/hazard/testmethods/sw846/new_meth.htm#1315.

Table 7-3. Toxicity Characteristic Leaching Procedure Screening Test Results

Mix #	Element	Sb	As	Ba	Be	Cd (spike)	Cr (spike)	Pb (spike)	Hg	Ni (spike)	Se	Ag	Tl
	UTS Limits, µg/L	1150	5000	21000	1220	110	600	750	25	11000	5700	140	200
	Reporting Limits, µg/L	<20	<10	<5	<5	<5	<5	<10	<0.2	<5	<10	<5	<25
1		-	16.4	312	-	-	11.4	-	-	43.8	40.7	-	-
2		-	30.6	1120	-	-	-	-	-	7.3	30.0	-	-
3		-	-	311	-	-	8.0	-	-	-	34.7	-	-
4		-	-	735	-	-	-	-	-	-	27.4	-	-
5		-	-	136	-	-	8.2	-	-	-	38.0	-	-
6		-	30.0	421	-	-	11.4	-	-	19.1	68.1	-	-
7		-	35.6	958	-	-	-	-	-	22.9	26.7	-	-
8		-	21.0	288	-	-	-	-	-	68.8	57.2	-	-
9		-	-	471	-	-	10.6	-	-	-	31.4	-	-
10		-	-	163	-	-	28.3	-	-	5.7	27.3	-	-
11		-	-	346	-	-	14.8	-	-	7.0	37.4	-	-
12		-	-	360	-	-	-	-	-	27.6	19.9	-	-
13		-	20.3	691	-	-	-	-	-	14.1	15.7	-	-
14		-	26.9	492	-	-	-	-	-	33.0	15.8	-	-
15		-	-	135	-	-	23.9	-	-	16.2	29.2	-	-
16		-	-	225	-	-	-	-	-	155	25.4	-	-
17		-	13.1	780	-	-	-	-	-	6.6	24.8	-	-
18		-	45.1	942	-	-	-	-	-	119	39.7	-	-
19		-	-	382	-	-	10.0	-	-	26.8	50.7	-	-
20		-	-	542	-	-	-	-	-	7.6	26.5	-	-
21		-	12.1	397	-	-	47.5	-	-	-	67	-	-
22		-	-	337	-	-	7.6	-	-	-	32.2	-	-
23		-	27.7	392	-	-	-	-	-	71.6	42.9	-	-
24		-	-	174	-	-	13.2	-	-	-	29.9	-	-
25		-	-	151	-	-	20	-	-	23.6	30.4	-	-
26		-	-	101	-	-	106	-	-	-	33.8	-	-
- = Values were below the reporting (detection) limits.													

were tested. The monoliths were prepared with simulants spiked with ^{99}Tc , stable I as iodide, U (U(VI)), and selected RCRA metals and UHCs, as shown in Table 3-3. Deionized water (DIW) was used as the leachant. The tests were conducted at ambient room temperature. The leach tests were conducted on 2-in.-diameter \times 4-in.-high cylinders. Surface areas of the Cast Stone monoliths are based on the geometric surface area of the monoliths.

7.5.1 EPA Draft Method 1315

The EPA Draft Method 1315 (EPA 2012) is a semi-dynamic leach test that consists of submerging a monolithic sample in DIW at a fixed liquid-volume to solid-geometric-surface-area ratio. The sampling was done at fixed periods of time at cumulative leaching times of 0.08, 1, 2, 7, 14, 28, 42, 49, and 63 days (EPA 2012). For these screening tests, two additional samplings were conducted at cumulative leaching times of 77 and 91 days. At each sampling interval, all the leaching fluid was removed and replaced with fresh fluid. A schematic of this process is shown in Figure 7-5.

At each of the predetermined leaching intervals, the monolith mass is recorded, and the leaching solution is changed. This method is similar to ANSI/ANS 16.1, *Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure* (ANSI 2003), but the leaching intervals are modified, and the developers of this method claim that the process of mass transfer can be interpreted by more complex release models that account for physical retention of the porous medium and chemical retention at the pore wall through geochemical speciation modeling.

In the screening tests described in this report, cylindrical monolith samples (\sim 2-in. diameter by \sim 4-in. in height) were placed into the centers of leaching vessels containing sufficient DIW to maintain a solution-to-solid surface area ratio of 9 ± 1 ml of leachant per square centimeter of sample geometric surface area. The geometric surface area is used in this test method and is calculated from the dimensions of the cylindrical monolith. Sample stands and holders were used to maximize the contact area of the monolith sample with the leaching solution. In between the sampling/replacement intervals, the leach vessels were covered with lids. Solution pH and electrical conductivities were measured within the leach vessel at each leaching interval. Then unfiltered leachate aliquots were removed and submitted for chemical analyses.

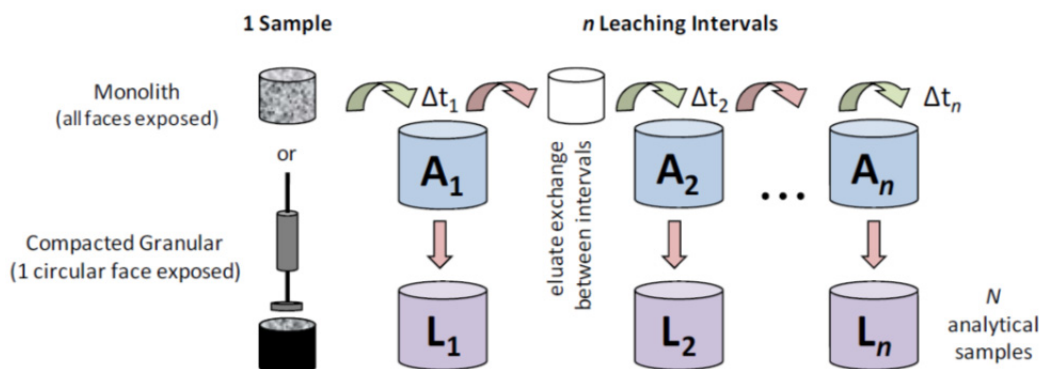


Figure 7-5. EPA Draft Method 1315 Testing Scheme (EPA 2012)

The observed diffusivity for each constituent was calculated using the analytical solution for simple radial diffusion from a cylinder into an infinite bath as presented by Crank (1975)

$$D_i = \pi \left[\frac{M_{ti}}{2\rho C_0(\sqrt{t_i} - \sqrt{t_{i-1}})} \right]^2 \quad (7-1)$$

where

- D_i = mean observed diffusivity of a specific constituent for leaching interval, i (m^2/s)
- M_{ti} = mass released per unit area of the monolith during leaching interval i (mg/m^2)
- t_i = cumulative contact time after leaching interval, i (s)
- t_{i-1} = cumulative contact time after leaching interval, $i-1$ (s)
- C_0 = initial leachable content (mg/kg)
- ρ = sample density (kg/m^3).

The leachability index (LI), a parameter derived directly from immersion test results, evaluates diffusion-controlled contaminant release with respect to time. The LI was calculated using

$$LI_n = -\log[10,000D_i], \quad (7-2)$$

where LI_n is the leachability index for each leach interval and D_i is the effective diffusivity for elements of interest (m^2/s) during the leach interval n . Note that the units of D_i in Equation (7-1) are square meter(s) per second, and the 10,000 multiplier in Equation (7-2) converts the units to square centimeter(s) per second (cm^2/s) prior to calculating the LI. In general, the LIs for each time interval are averaged for each COC to calculate an average LI. The average LI is used as a criterion to assess whether solidified/stabilized waste is likely to be acceptable for subsurface disposal in waste repositories.

For purposes of the statistical analyses of the screening test results, the effective diffusivities and LIs for the 28-, 42-, 49-, and 63-day intervals were averaged for each COC to calculate an average effective diffusivity and an average LI for each of the two duplicate samples of a given Cast Stone mix. Then, the average effective diffusivities and LI values for the two duplicates were used to calculate means and standard deviations.

7.5.2 Effective Diffusivity Results

The EPA Draft Method 1315 leach tests were conducted for a total of 91 days with additional leachant changes at 77 and 91 cumulative days, which were 14 and 28 days beyond the standard 63 days of the EPA Draft Method 1315 but similar in duration to the ANSI/ANS 16.1 method. Figure 7-6 shows the resulting effective diffusivities for Na, nitrate, nitrite, I (added as iodide), Tc (added as pertechnetate), Cr (added as chromate), and U (added as uranyl) for Cast Stone Mix 5 (Average 7.8 M Na simulant mixed with NW/NW dry blend and a free-water-to-dry-blend ratio of 0.4). The figure shows the relative leaching performance of the different constituents with Na, nitrate, nitrite, and I having comparable effective diffusivities and with Tc, Cr, and U having increasingly lower effective diffusivities. As is discussed further below, many of the U leachate concentrations were below detection limits so the U diffusivity values shown represent

maximums. As will be more evident in the figures below showing the leach results for the individual species, the effective diffusivities for Na, I, nitrate, and nitrite appear to continue to decrease over time, while the effective diffusivities for Tc and Cr are relatively constant for the duration of the test. This is due in part to the Na, I, nitrate, and nitrite being more leachable such that the semi-infinite solid boundary conditions associated with Equation (7-1) are not strictly met when the cumulative fraction leached of the species exceeds approximately 20 percent of the initial inventory (ANSI/ANS 16.1 2003). This can occur within the first 7 to 28 days for these more leachable species.

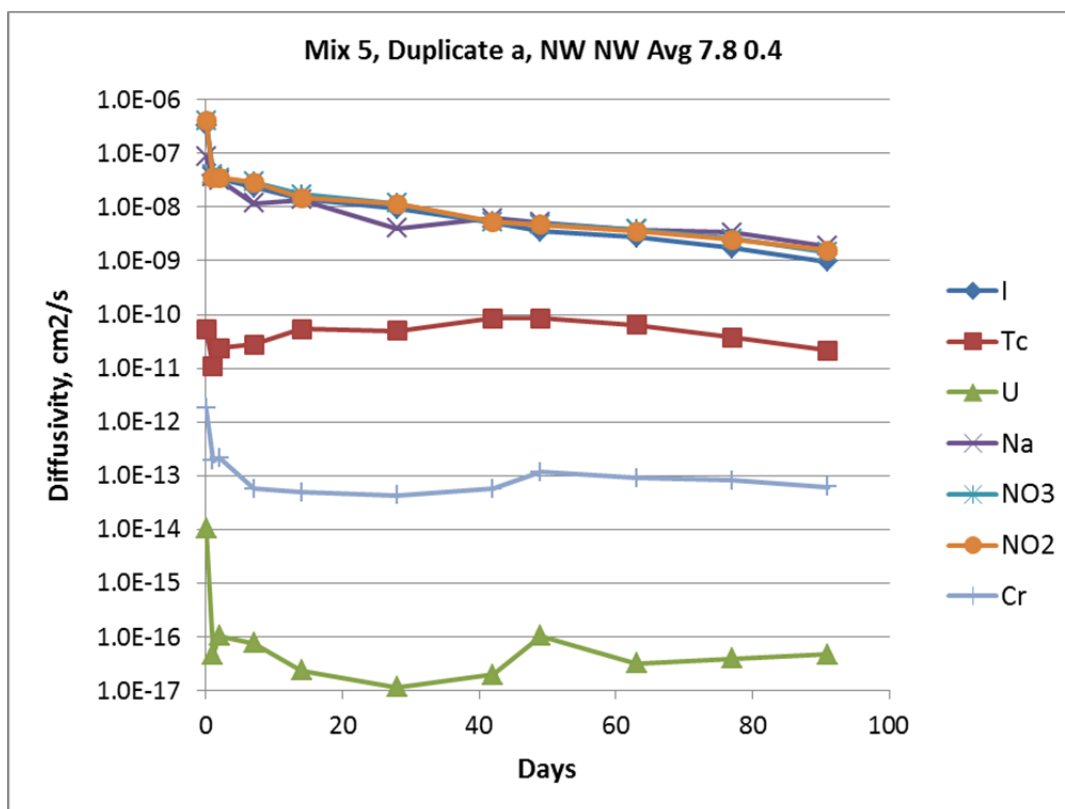


Figure 7-6. Range of Effective Diffusivities for Selected Waste Components

Each of the waste constituents' leach properties is discussed further in the sections that follow. Figures show the range of effective diffusivities across the 26 Cast Stone mixes. As discussed in the processing properties discussions of Section 6.0, some of these 26 formulations are not viable from a processing perspective and would not be used in production operations. The figures also show some of the impacts of waste composition and the dry blend mix. Detailed discussion of the statistical analyses of the leach test results follows in Section 8.0. Calculated effective diffusivities for Na, nitrate, nitrite, iodine, Tc, Cr, and U are provided in Tables D-7 through D-13 in Appendix D.

7.5.3 Sodium Effective Diffusivity Results

Figure 7-7 shows the range of Na effective diffusivities observed among the 26 Cast Stone mixes in the screening test matrix (Table 2-1). The mix designations "a" and "b" refer to the duplicate

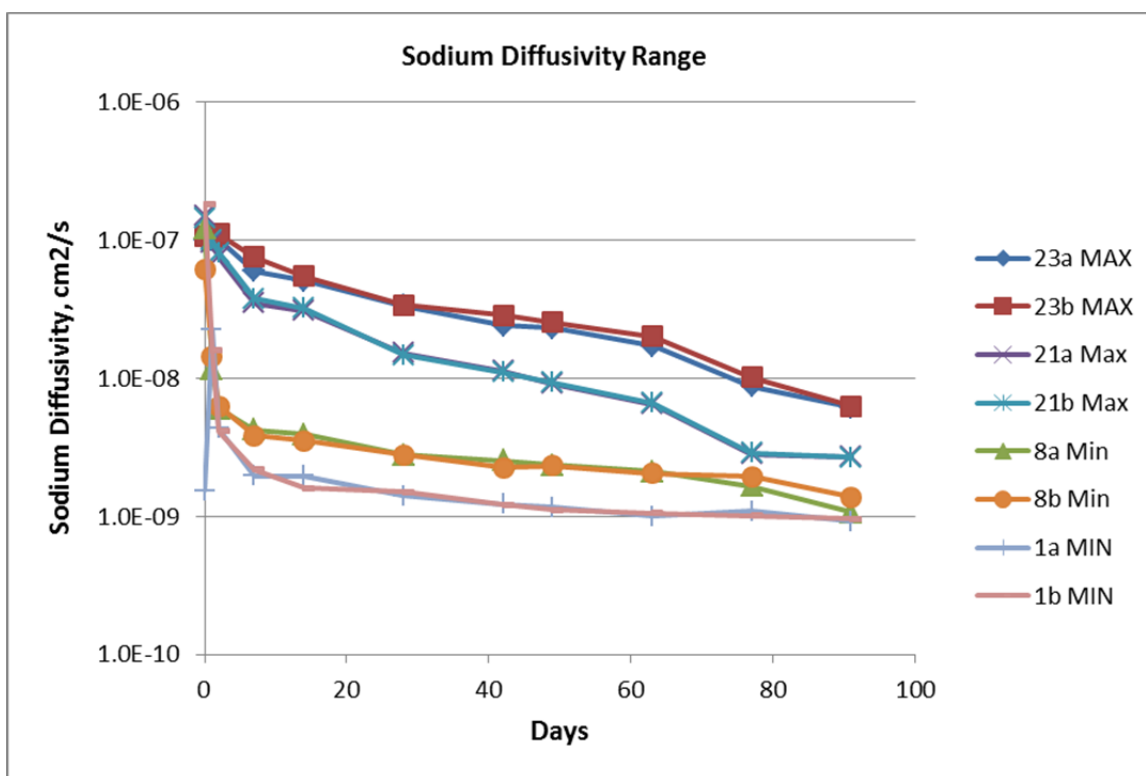


Figure 7-7. Range of Sodium Effective Diffusivities

samples of each Cast Stone mix. At the extremes, the Na diffusivities are bounded by Mix 23 on the high end and Mix 1 on the low end. Because of processing properties (i.e., Mix 1 set up immediately, Mix 23 set too slowly), neither of these mixes would be considered for actual production. Excluding these two mixes, the Na diffusivities are approximately bounded by Mixes 21 and 8. At 63 days, the range of Na effective diffusivities among the remaining 24 mixes is between 1.7×10^{-9} and 8.0×10^{-9} cm²/s (Mixes 19 and 24, respectively).

Figure 7-8 compares the effective diffusivities for mixes with the NW dry blend components and mixes with the SE dry blend components for the High SO₄, High Al, and SST Blend simulants at the same Na molarities and water-to-dry-blend solids ratios. There appears to be some impact of the source of the dry blend components; the NW dry blend components yield Cast Stone mixes with slightly higher Na effective diffusivities than the SE dry blend components.

Figure 7-9 shows the Na effective diffusivities for the NW dry blend components and the four simulants used in the screening test matrix. The different simulant compositions appear to have no impact on the Na diffusivity. All three Na diffusivity figures show that the effective diffusion coefficients for the duplicate samples (a and b) for each mix yield very similar values.

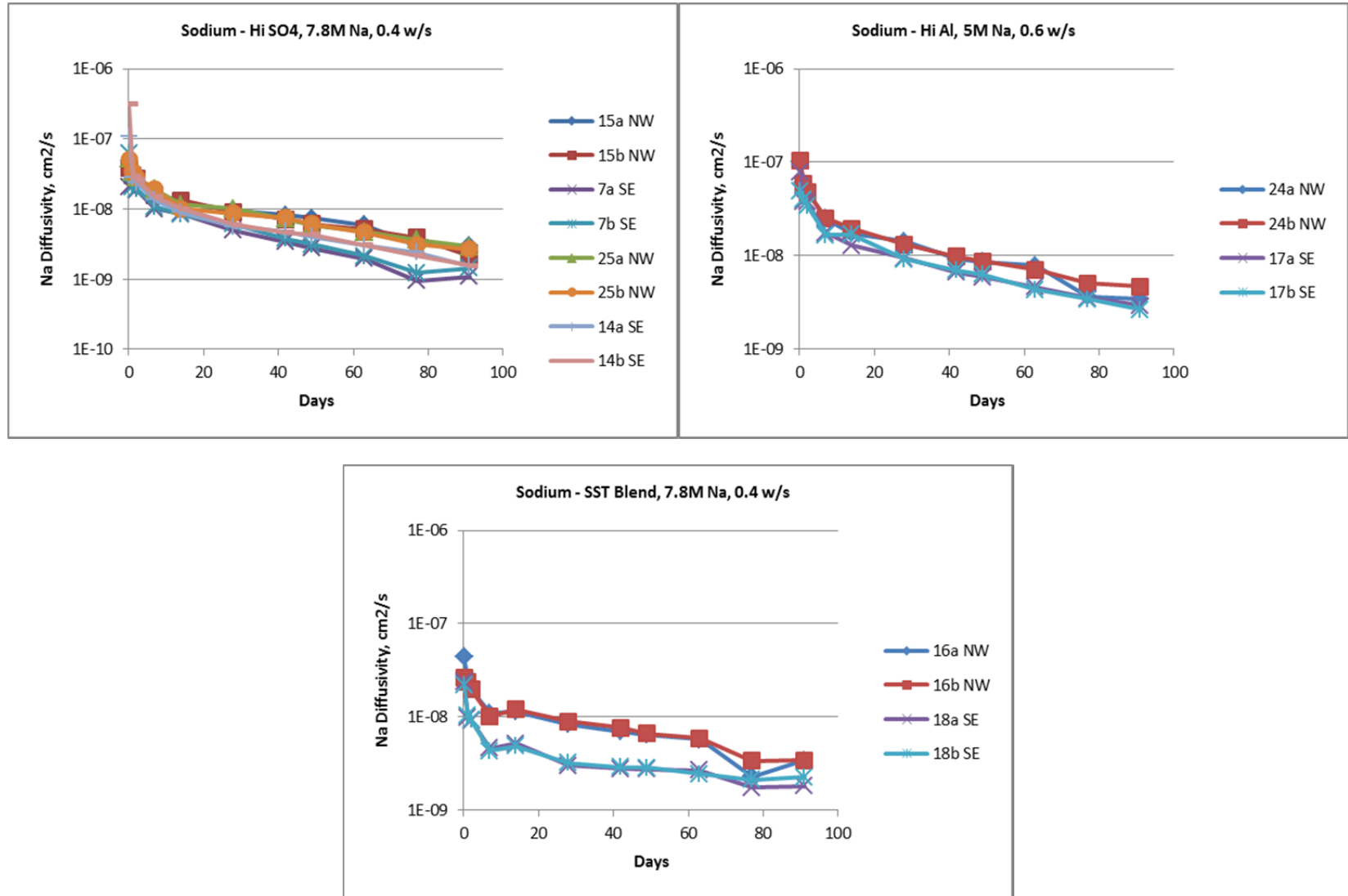


Figure 7-8. Impact of Dry Blend Mix on Sodium Effective Diffusivities

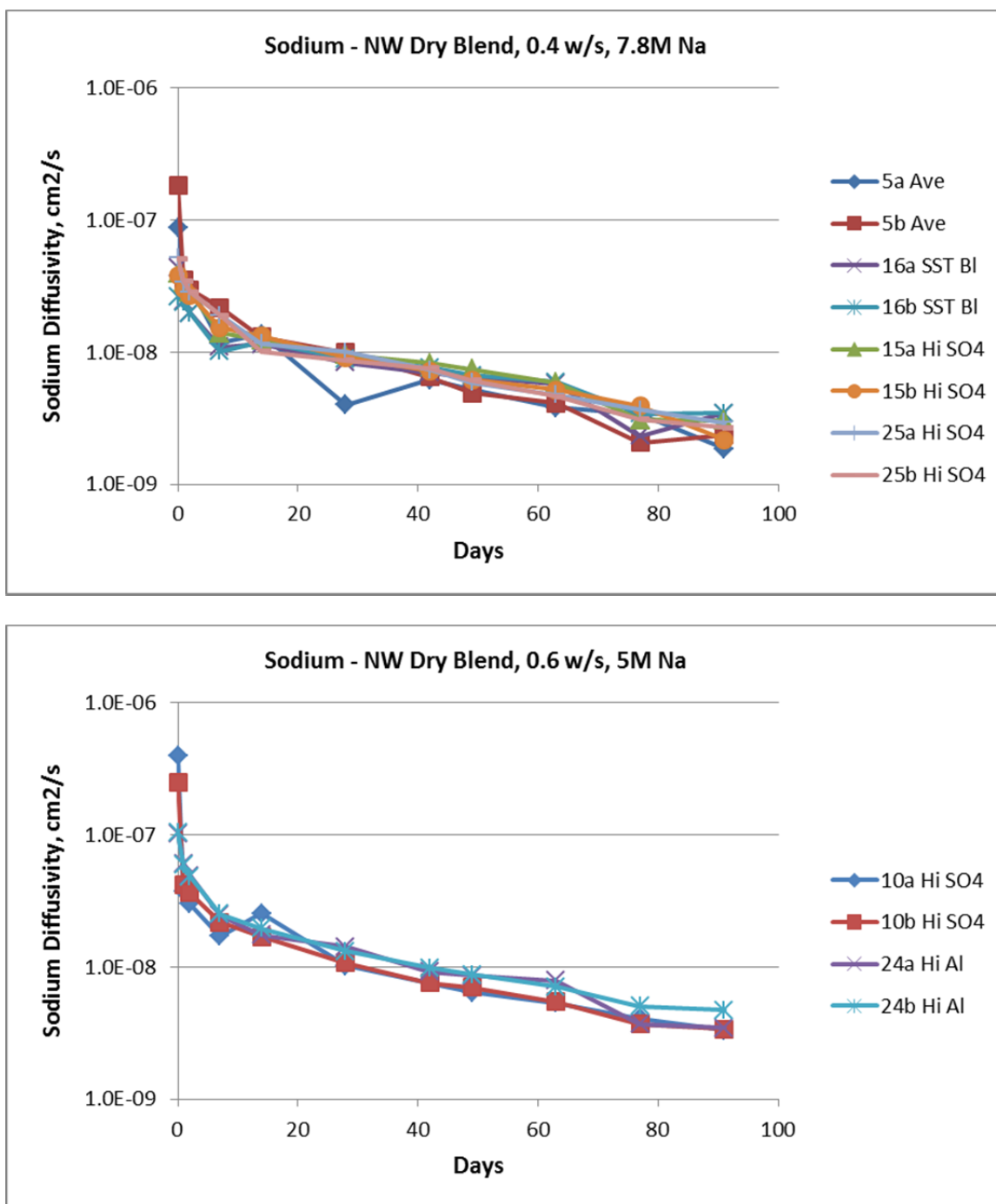


Figure 7-9. Impact of Waste Composition on Sodium Effective Diffusivities

7.5.4 Nitrate and Nitrite Effective Diffusivities

Statistical analysis of the leach test results for nitrate and nitrite showed that the effective diffusivities of these two constituents were highly correlated (see Figure 8-3 in Section 8). Therefore, this section covers both anions and the discussion focuses on nitrates as representing

both species. Figure 8-3 also shows that the Na leach test results are strongly correlated with the nitrate and nitrite leach test results, so the results described here are very similar to those for Na described above.

Figure 7-10 shows the range of nitrate effective diffusivities observed among the 26 Cast Stone mixes in the screening test matrix. As with Na, at the extremes, the nitrate and nitrite diffusivities are bounded by Mix 23 on the high end and Mix 1 on the low end. Because of processing properties, neither of these mixes would be considered for actual production. Excluding these two mixes, the nitrate and nitrite diffusivities are bounded by Mixes 21 and 8. At 63 days, the range of nitrate effective diffusivities among the remaining 24 mixes is between 2.3×10^{-9} and 1.0×10^{-8} cm²/s (Mixes 19 and 21, respectively). For nitrite, the range of effective diffusivities for these remaining 24 mixes at 63 days is between 2.2×10^{-9} and 9.6×10^{-9} cm²/s (Mixes 8 and 11, respectively).

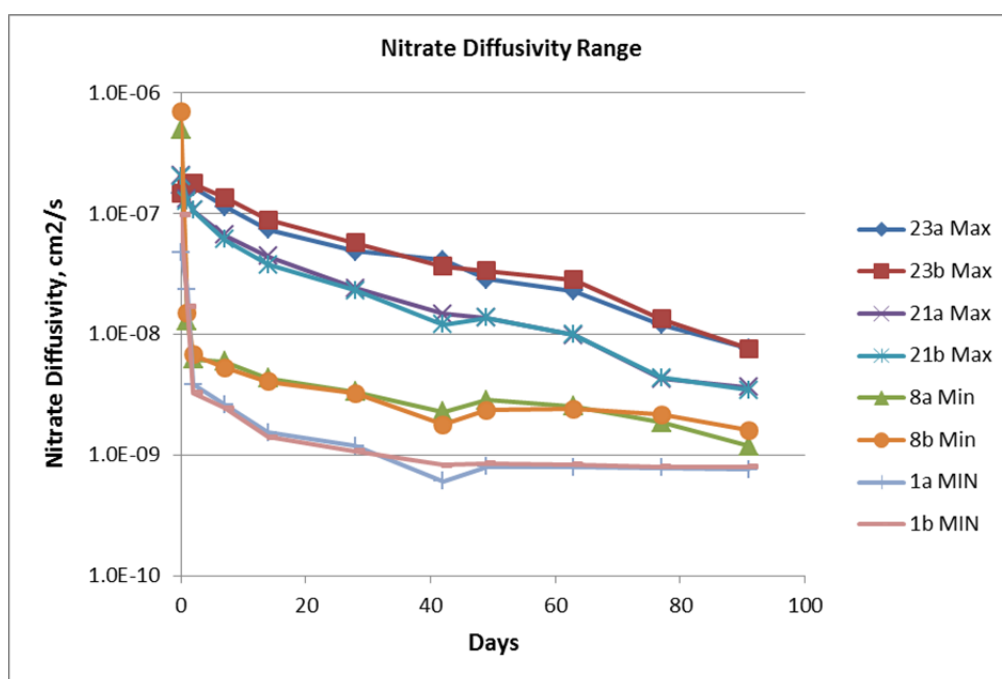


Figure 7-10. Range of Nitrate Effective Diffusivities

Figure 7-11 compares the effective diffusivities for mixes with the NW dry blend components and mixes with the SE dry blend components for the High SO₄, High Al, and SST Blend simulants at the same Na molarities and water-to-dry-blend solids ratios. Other than for the SST Blend, the source of the dry blend components does not appear to have any significant impact on the nitrate diffusivity values. For the SST Blend, the Cast Stone monoliths prepared with the NW-NW dry blend appear to leach slightly more nitrate than the Cast Stone made with SE-SE dry blend.

Figure 7-12 shows the nitrate effective diffusivities for the NW dry blend components and the four simulants used in the screening test matrix. The different simulant compositions do not appear to have any impact on nitrate diffusivity. All three nitrate diffusivity figures show that the nitrate effective diffusion coefficients for the duplicate samples (a and b) for each mix yield very similar values.

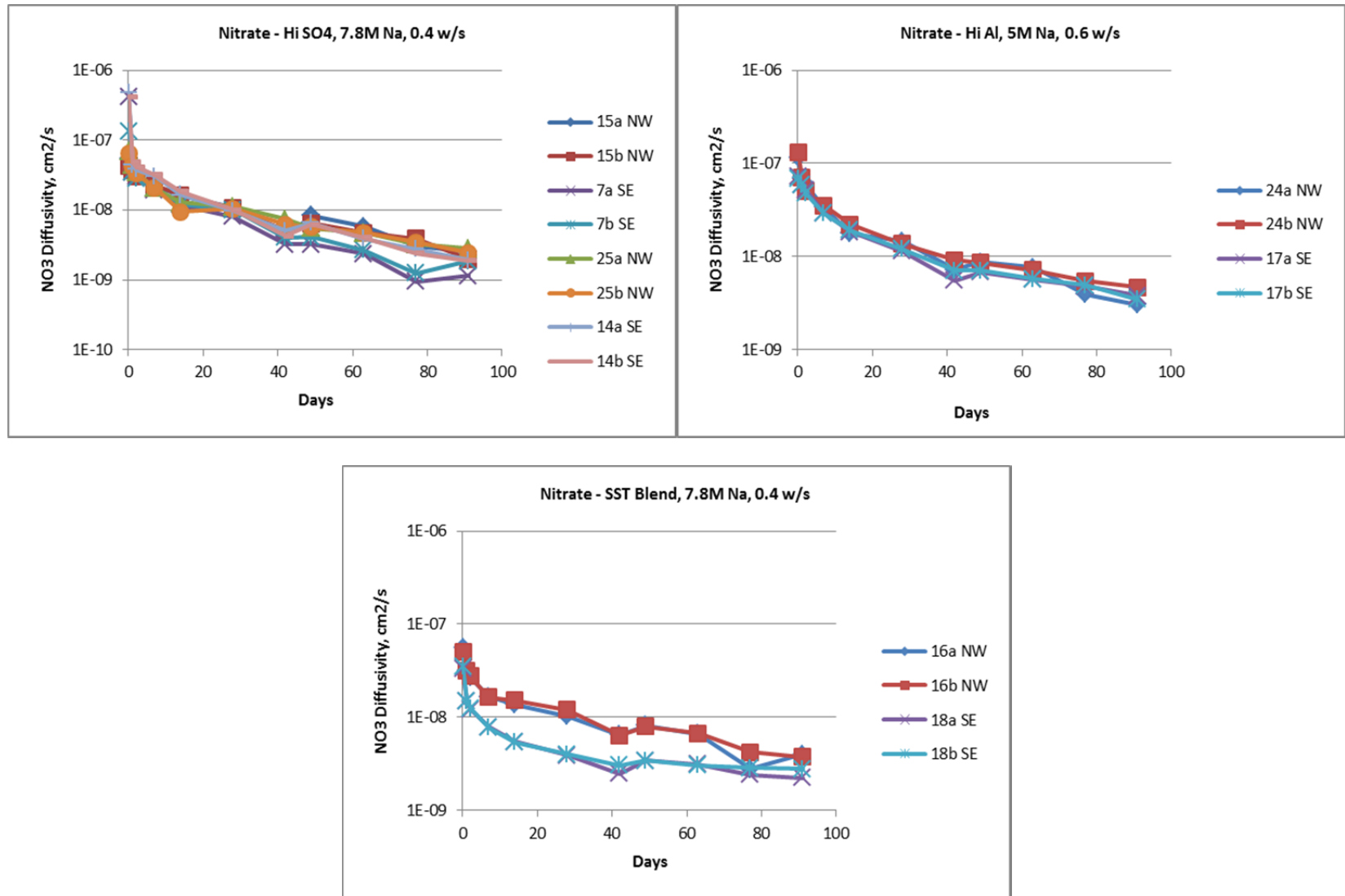


Figure 7-11. Impact of Dry Blend Mix on Nitrate Effective Diffusivities

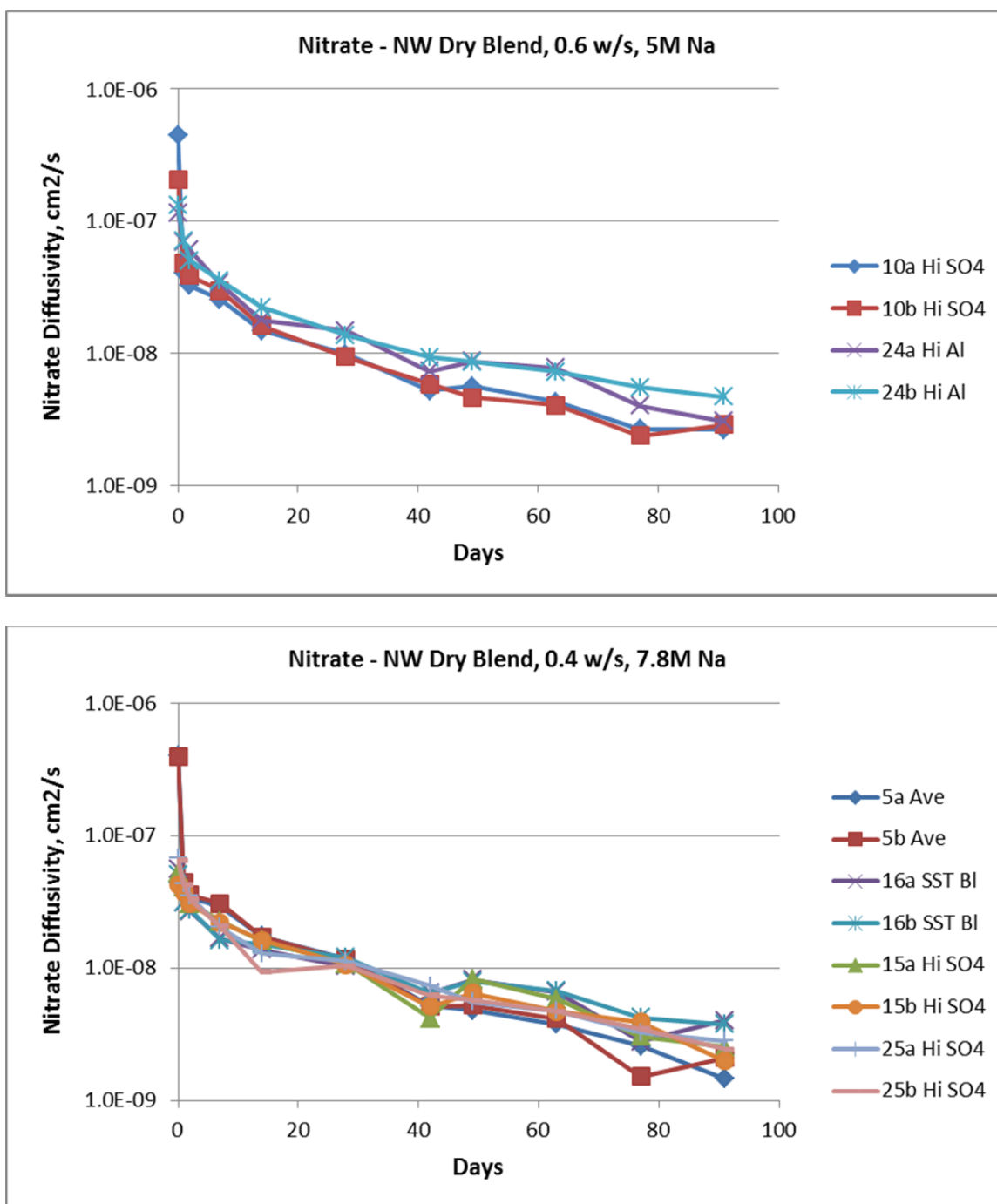


Figure 7-12. Impact of Waste Composition on Nitrate Effective Diffusivities

7.5.5 Iodine Effective Diffusivities

Figure 7-13 shows the range of I (added as iodide) effective diffusivities observed among the 26 Cast Stone mixes in the screening test matrix. As with Na, nitrate, and nitrite discussed above, the I diffusivities are bounded by Mix 23 on the high end and Mix 1 on the low end. Because of processing properties, neither of these mixes would be considered for actual production.

Excluding these two mixes, the I diffusivities are approximately bounded by Mixes 21 and 8. At 63 days, the range of I effective diffusivities among the 24 remaining mixes is between 2.0×10^{-9} and 1.2×10^{-8} cm²/s (Mixes 7 and 11, respectively).

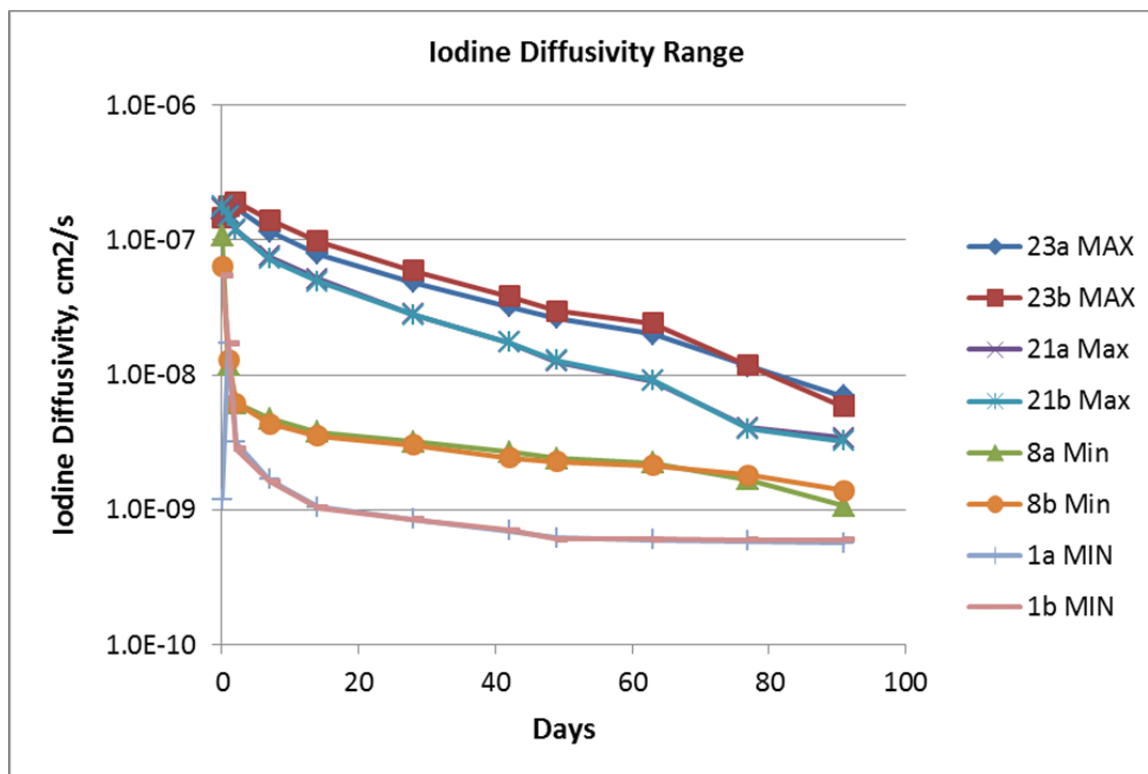


Figure 7-13. Range of Iodine Effective Diffusivities

Figure 7-14 compares the iodide effective diffusivities for mixes with the NW dry blend components and mixes with the SE dry blend components for the High SO₄, High Al, and SST Blend simulants at the same Na molarities and water-to-dry-blend solids ratios. Other than for the SST Blend, the source of the dry blend components does not appear to have any impact on the iodide effective diffusivities. For the SST Blend, the NW-NW dry blend Cast Stone monoliths appear to leach slightly more I than the Cast Stone made with SE-SE dry blend.

Figure 7-15 shows the I effective diffusivities for the NW dry blend components and the four simulants used in the screening test matrix. The different simulant compositions do not appear to have any impact on the I diffusivity at 7.8 M Na. There appears to be a slight difference in the I effective diffusivities between the High SO₄ and High Al wastes at 5 M Na. All three iodine diffusivity figures show that the I effective diffusion coefficients for the duplicate samples (a and b) for each mix yield very similar values.

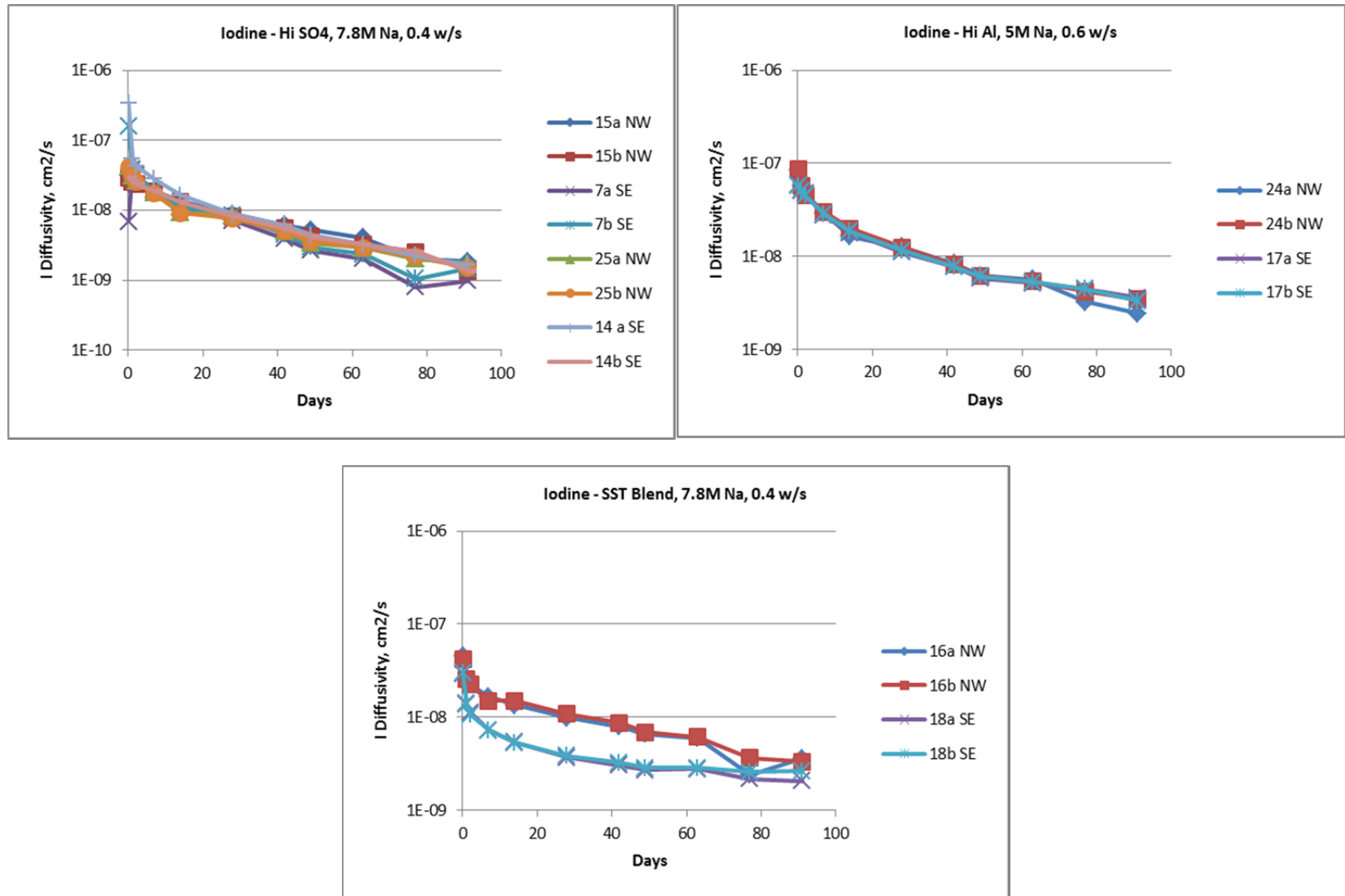


Figure 7-14. Impact of Dry Blend Mix on Iodine Effective Diffusivities

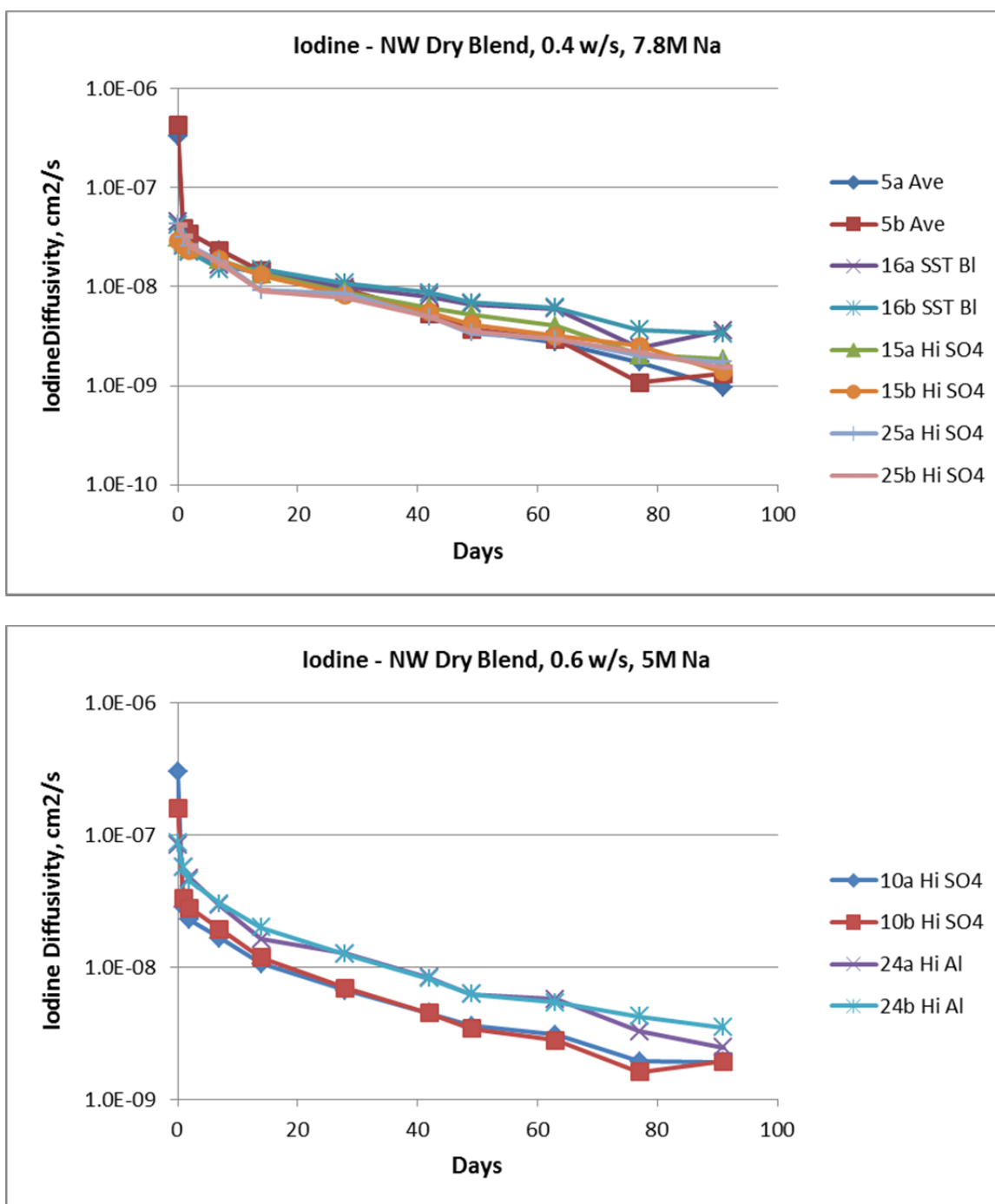


Figure 7-15. Impact of Waste Composition on Iodine Effective Diffusivities

7.5.6 Technetium Effective Diffusivities

Figure 7-16 shows the range of Tc effective diffusivities observed among the 26 Cast Stone mixes in the screening test matrix. Mix 23 had the highest Tc diffusivities of the 26 mixes. Because of processing properties, this mix would not be considered for actual production. Unlike the Na, nitrate/nitrite, and I diffusivities discussed above where Mix 1 represented a lower bound,

Mix 18 had the lowest Tc diffusivities from 28 through 63 days. At 63 days, the range of Tc effective diffusivities among the 25 mixes is between 6.8×10^{-12} and 2.3×10^{-10} cm²/s (Mixes 18 and 8, respectively).

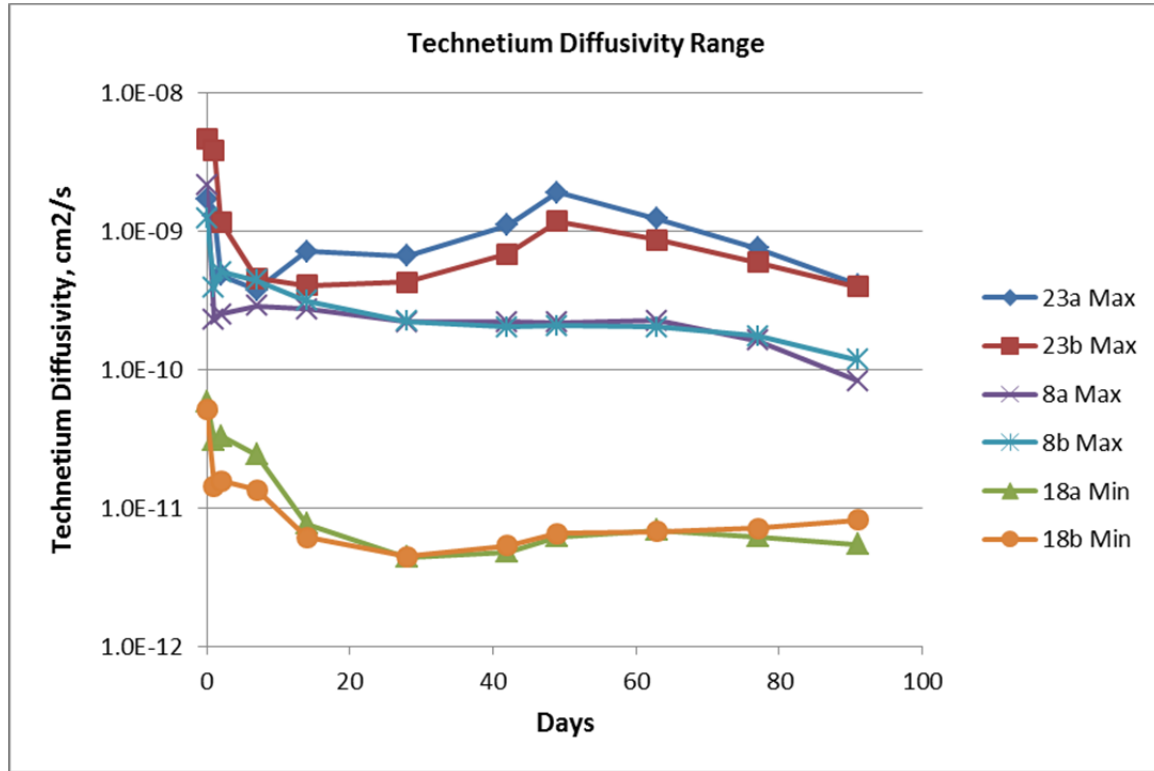


Figure 7-16. Range of Technetium Effective Diffusivities

Figure 7-17 compares the effective diffusivities for mixes with NW dry blend components and mixes with the SE dry blend components for the High SO₄, High Al, and SST Blend simulants at the same Na molarities and water-to-dry-blend solids ratios. The source of the dry blend components appears to have an impact, but the trend is not consistent across the three simulants. Technetium diffusivities are lower for the SE dry blend components with the High SO₄ and SST Blend simulants. However, for the High Al simulant, the Tc diffusivities are lower Cast Stone prepared with the NW dry blend ingredients.

Figure 7-18 shows the Tc effective diffusivities for the NW dry blend components and the four simulants used in the screening test matrix. All three Tc diffusivity figures show in general that the Tc effective diffusion coefficients for the duplicate samples (a and b) for each mix yield very similar values. The Tc effective diffusion coefficient values for a few of the mixes at particular leach times show some variation between the two duplicates, but the differences are small and show no time or mix consistency and thus are not considered practically important.

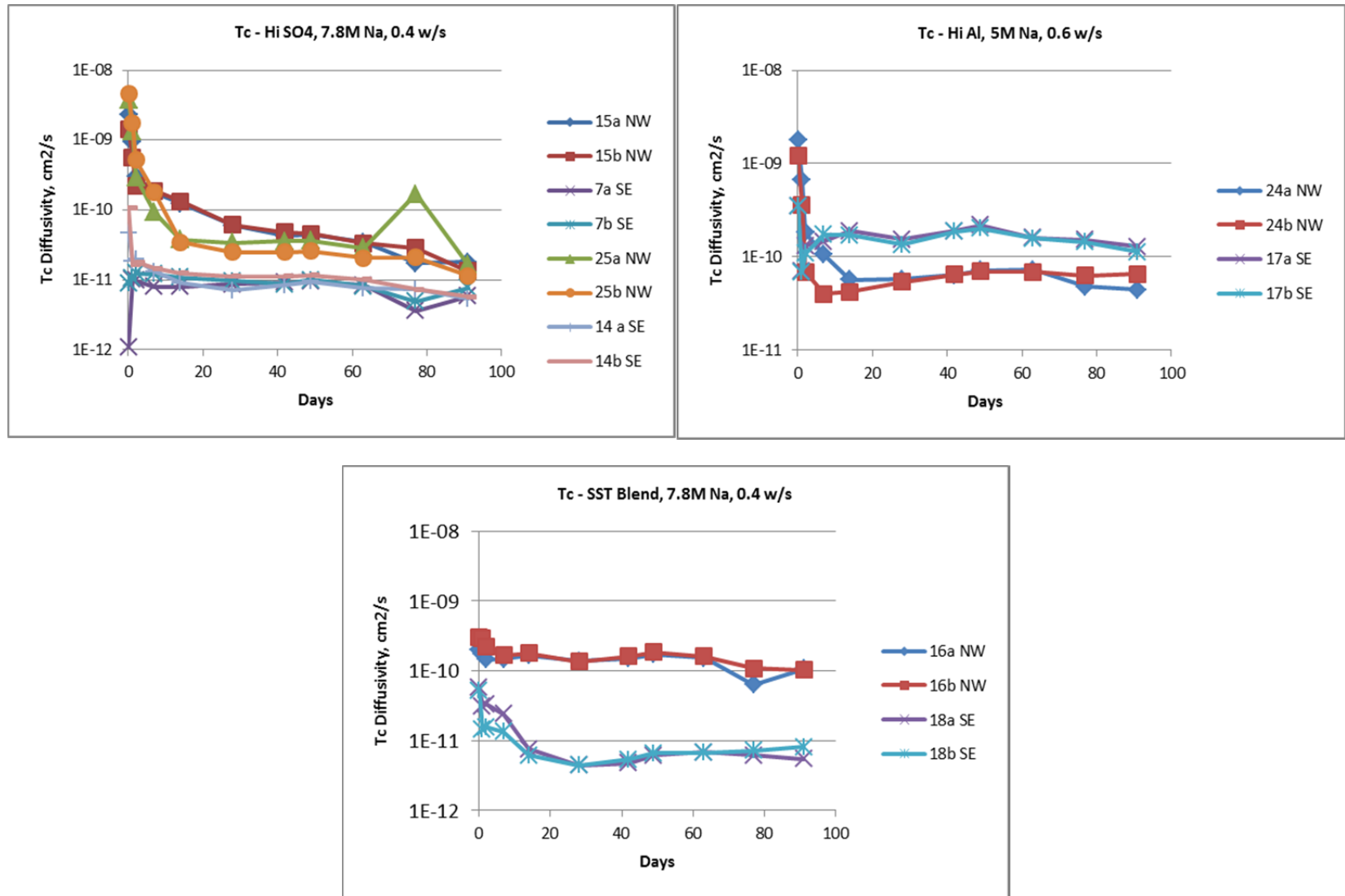


Figure 7-17. Impact of Dry Blend Mix on Technetium Effective Diffusivities

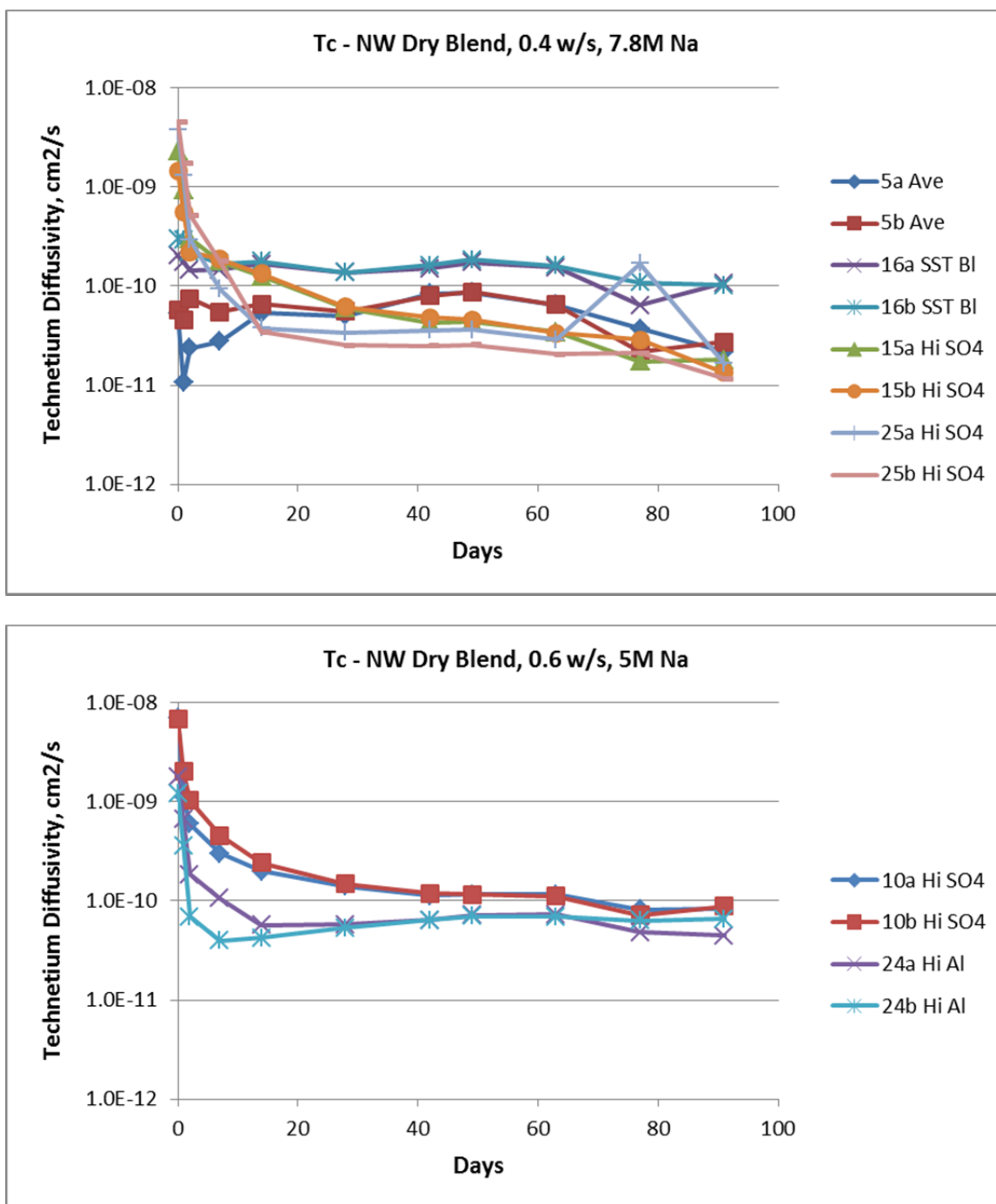


Figure 7-18. Impact of Waste Composition on Technetium Effective Diffusivities

7.5.7 Chromium Effective Diffusivities

Figure 7-19 shows the range of Cr effective diffusivities observed among the 26 Cast Stone mixes in the screening test matrix. At longer times, Mix 23 has the highest Cr diffusivities, but the differences between the next highest mixes (8 and 10) are not as great as those observed for the other species leached. As mentioned previously, Mix 23 would not be considered a viable recipe

for producing Cast Stone on a full scale. One replicate pair of mixes (#7 and #14) had the lowest Cr diffusivities. At 63 days, the range of Cr effective diffusivities among the 24 viable mixes is between 1.0×10^{-14} and 8.3×10^{-13} cm²/s (Mixes 7 and 10, respectively).

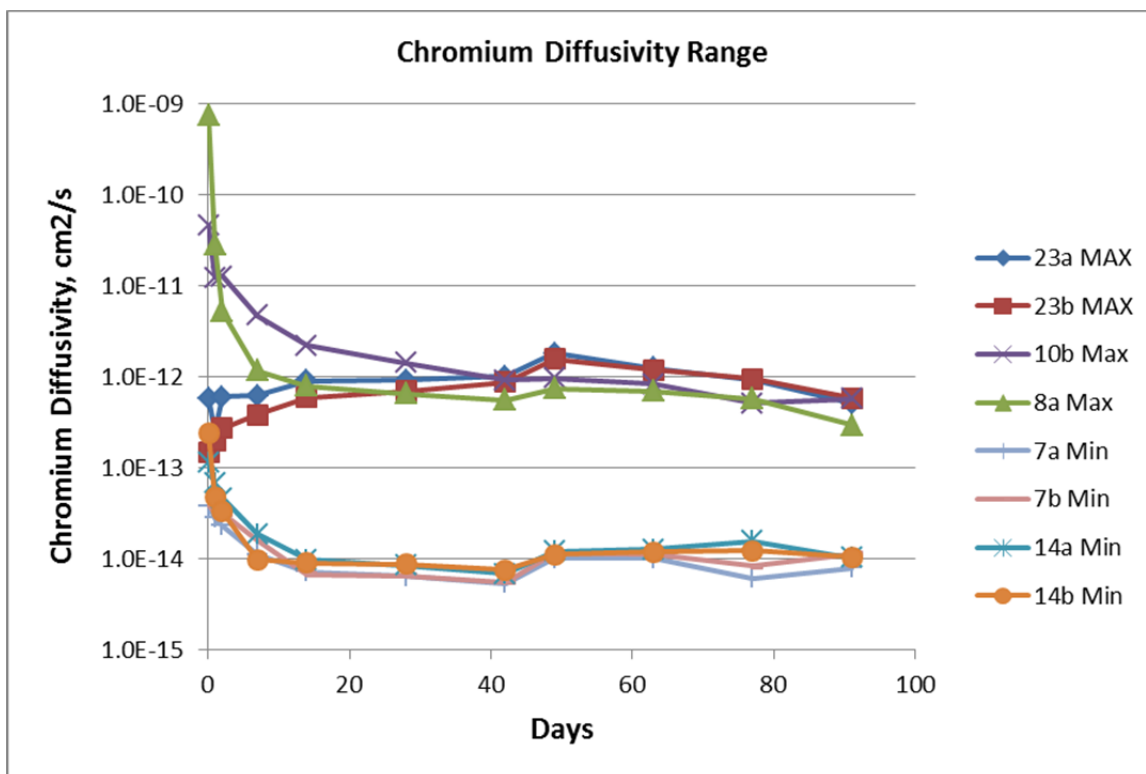


Figure 7-19. Range of Chromium Effective Diffusivities

Figure 7-20 compares the effective diffusivities of mixes with the NW dry blend components and mixes with the SE dry blend components for the High SO₄, High Al, and SST Blend simulants at the same Na molarities and water-to-dry-blend solids ratios. The source of the dry blend components appears to have an impact, but the trend is not consistent across all the simulants. Chromium diffusivities are lower for the SE dry blend components with the High SO₄ and SST Blend simulants. However, for the High Al simulant, the Cr diffusivities are indistinguishable between the two dry blend mixes. It should be noted that for the SST Blend, the 49- and 63-day Cr concentrations for the Mix 18 leachates were below analytical detection limits.

Figure 7-21 shows the Cr effective diffusivities for the NW dry blend components and the four simulants used in the screening test matrix. Although there are differences in the Cr diffusivities during the early part of the leach tests, at 49 and 63 days there does not appear to be any significant difference due to the simulant compositions at 7.8 M Na. There appears to be a slight difference in the Cr effective diffusivities between the High SO₄ and High Al wastes at 5 M Na. All three Cr diffusivity figures show that the effective diffusion coefficients for the duplicate samples (a and b) for each mix beyond the first 21 days of leaching yield very similar values.

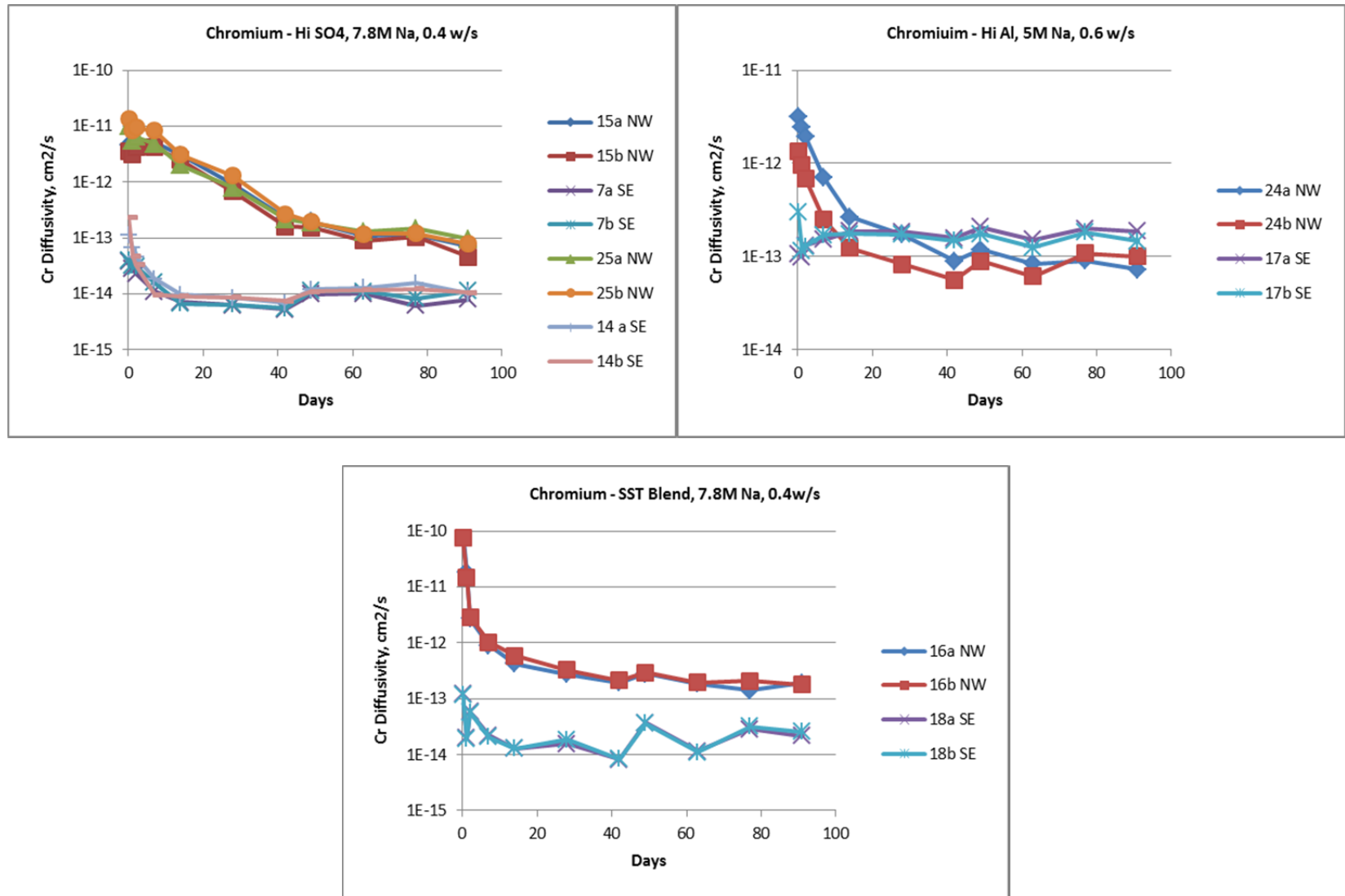


Figure 7-20. Impact of Dry Blend Mix on Chromium Effective Diffusivities

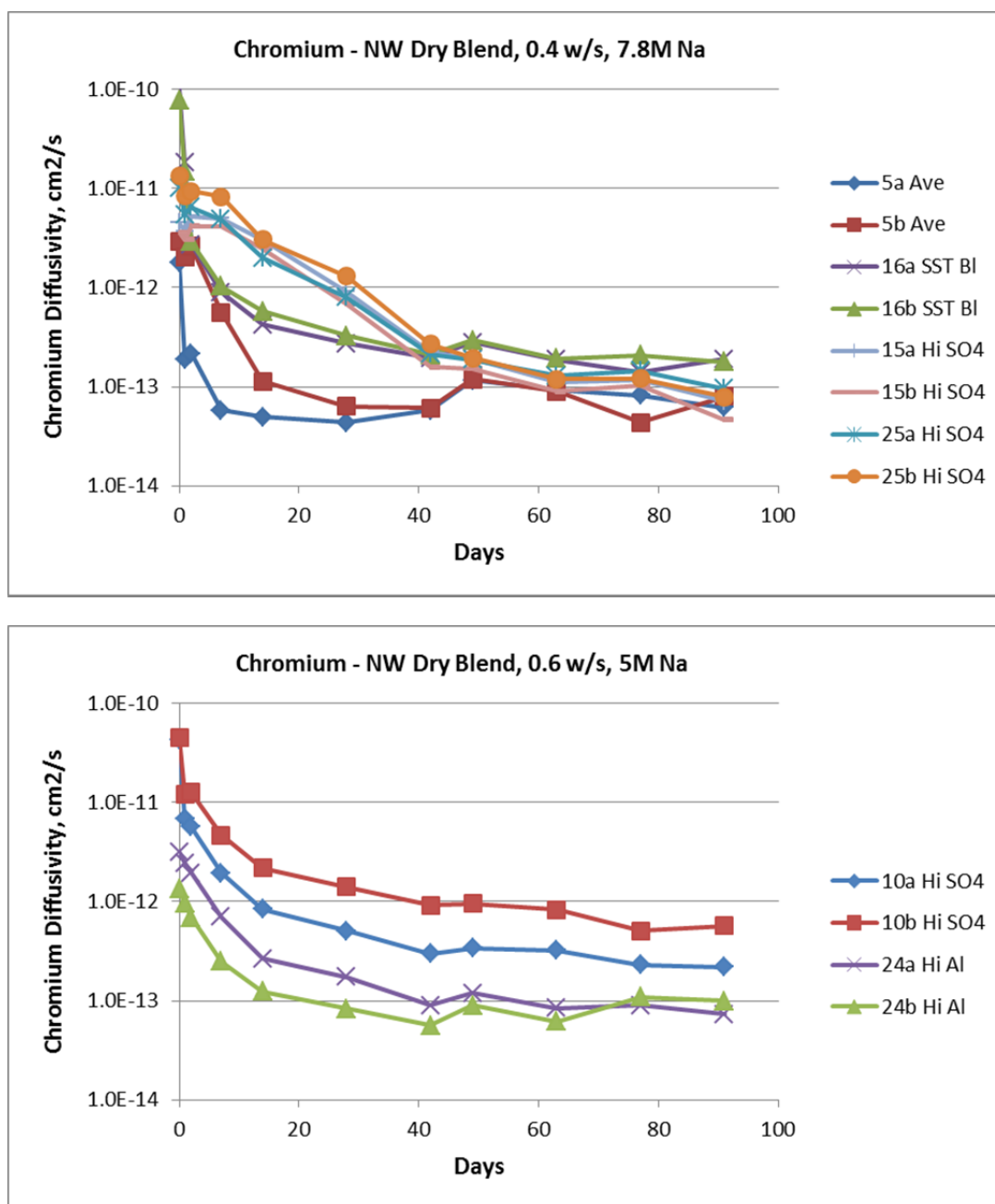


Figure 7-21. Impact of Waste Composition on Chromium Effective Diffusivities

7.5.8 Uranium Effective Diffusivities

Figure 7-22 shows the range of U calculated effective diffusivities observed among the 26 Cast Stone mixes in the screening test matrix. Uranium was not detected in most of the leachates through 91 days of testing for the 26 mixes. The curves labeled “U < Min a” and “U < Min b” are the minimum effective diffusivities calculated from the analytical detection limits for U in the tests. Only Mixes 1, 8, 16, 18, and 23 had measureable U concentrations for the leachates for the entire 91 days of the test. Figure 7-23 shows the U effective diffusivities for these five mixes. At 63 days, the diffusivities were in the range of 1.1×10^{-16} and 6.0×10^{-16} cm²/s. As mentioned,

Mix 23 had processability issues and would not be a candidate for full production of Cast Stone. All three U diffusivity figures show that the effective diffusion coefficients for the duplicate samples (a and b) for each mix yield very similar values.

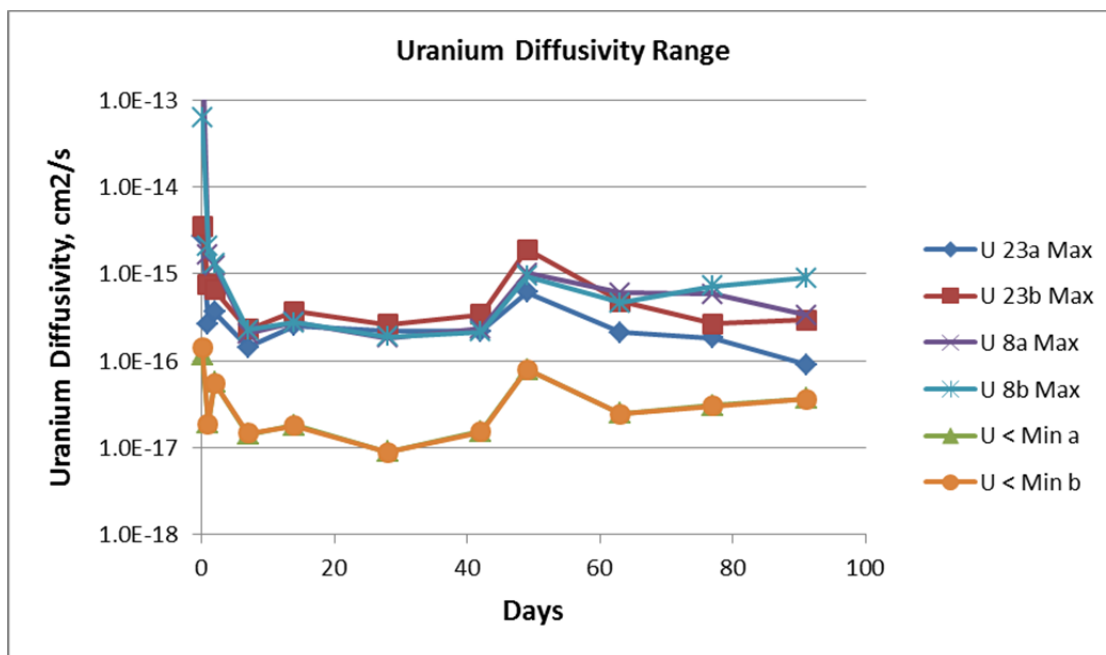


Figure 7-22. Range of Uranium Diffusivities

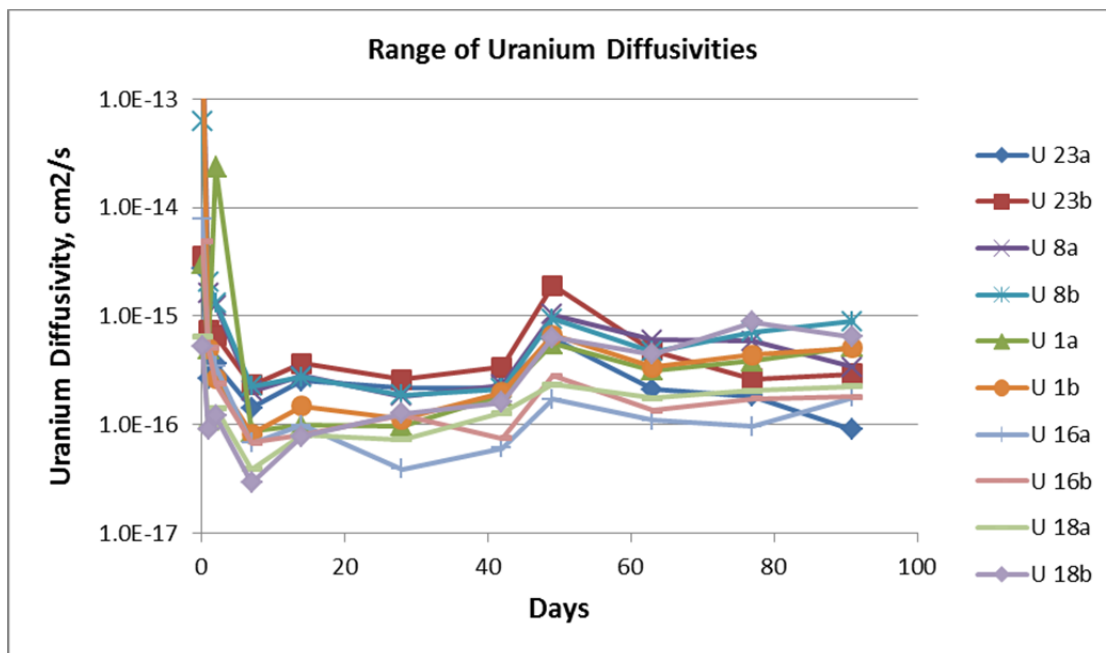


Figure 7-23. Range of Uranium Diffusivities for Specimens with Measurable Uranium Concentrations in all the Leachates

8.0 Statistical Analysis of the Effects of Screening Test Parameters on Cast Stone Properties

Statistical analyses were performed for several Cast Stone processing and cured waste form properties. The objectives of the statistical analyses were to 1) determine whether individual test parameters and their two-parameter interactions have statistically significant effects on a given property, and 2) assess the relative magnitudes of these effects. Sections 8.1 to 8.3 describe the background and two approaches used for statistical analyses of data from the Cast Stone screening tests. Section 8.4 discusses cautions that should be kept in mind when considering results of the two statistical analysis approaches described in Sections 8.2 and 8.3. Section 8.5 presents estimates of the experimental and measurement uncertainties in Cast Stone properties based on the five pairs of replicate tests. The results of applying the two statistical data analysis approaches are discussed for Cast Stone processing properties in Section 8.6 and cured properties in Section 8.7. Finally, Section 8.8 summarizes the results of the statistical analyses.

8.1 Statistical Analysis Approaches

Typically in screening experiments, the goal of the data analysis is to determine which test parameters have statistically significant effects and which do not. Fortunately, the total number of tests that could be performed in the test matrix was sufficient to assess not only the individual effects of the parameters, but also the effects of some pre-specified two-parameter interactions. By pre-specifying some two-parameter interactions of interest, it was assumed the remaining two-parameter interactions (as well as higher-order interactions) were not significant. As described in Section A.3 in Appendix A, some individual-parameter effects and two-parameter interactions were assumed to be not significant, and then the test matrix was developed to support estimating the remaining individual-parameter effects and two-parameter interactions.

Assessing individual-parameter effects and some two-parameter interactions with a limited number of experimental tests comes with a price. The price is that the parameter effects (individual and/or interaction) assumed to be not significant are partially aliased (i.e., confounded) with the parameter effects assumed to be significant. If the assumptions about nonsignificant effects are wrong, the results of the statistical data analyses could be misleading.

As discussed in Appendix A, the screening test matrix in Table 2-1 was developed under the following assumptions:

- A. There is no difference in the effects of the Average and SST Blend simulants on Cast Stone properties.
- B. Interactions of the Average and SST Blend simulants with other test parameters are not significant.
- C. Interactions of BFS with other test parameters are not significant.

These assumptions were assessed as part of the data analysis methods described in Sections 8.2 and 8.3. As discussed subsequently, the critical assumption that the BFS interactions are not significant is not supported by the statistical analyses performed.

Two approaches to statistical analyses of the data were performed to assess whether the test parameters and their two-parameter interactions have statistically significant effects on each of several Cast Stone properties. The first approach involved fitting (for each property) a “Full Model” that allows for individual effects of the five test parameters (simulant, sodium molarity,

fly ash source, BFS source, and mix ratio) and two-parameter interactions of all parameters except those involving BFS. Hence, the Full Model is larger than the model used to develop the experimental design, and provides for assessing Assumptions A and B above. The Full Model approach to statistical analyses of the Cast Stone screening test data is described in Section 8.2.

The second approach to statistical analysis involves using stepwise regression to build a model consisting of statistically significant individual parameter effects and two-parameter interaction effects. This second approach allows for selecting two-parameter interactions involving BFS as well as other two-parameter interactions. Hence, the second approach provides for assessing Assumption C above. This second approach to statistical analyses of the Cast Stone screening test data, referred to as the “Stepwise Model” approach, is described in Section 8.3.

8.2 Full Model Approach to Statistical Analysis of the Screening Test Data

This section discusses the Full Model and the corresponding data analysis approach.

8.2.1 Full Model

With screening test data collected according to Table 2-1 or Table C-1, it is possible to fit a statistical model that includes 1) all individual parameter effects, and 2) all two-parameter interactions, except those involving the BFS source. This model is given by

$$\begin{aligned} y_{hijkm} = & \mu + Sim_h + NaMol_i + FA_j + BFS_k + MR_m + (Sim_h * NaMol_i) \\ & + (Sim_h * FA_j) + (Sim_h * MR_m) + (NaMol_i * FA_j) \\ & + (NaMol_i * MR_m) + (FA_j * MR_m) + \varepsilon_{hijkm} \end{aligned} \quad (8-1)$$

- where
- y_{hijkm} = measured response value for the h^{th} level of *Sim*, i^{th} level of *NaMol*, j^{th} level of *FA*, k^{th} level of *BFS*, m^{th} level of *MR*, and a random error
 - μ = true, unknown mean of the response (y) over all combinations of the test parameters
 - Sim_h = effect of the h^{th} level of simulant type, with four levels (Average, Blend, HiAl, and HiSO₄)
 - $NaMol_i$ = effect of the i^{th} level of sodium molarity, with two levels (low = 5.0, high = 7.8)
 - FA_j = effect of the j^{th} level of fly ash, with two levels (low Ca, high Ca)
 - BFS_k = effect of the k^{th} level of blast furnace slag, with two levels (NW, SE)
 - MR_m = effect of the m^{th} level of mix ratio, with two levels (low = 0.4, high = 0.6)
 - ε_{hijkm} = random error in performing a test combination for the h^{th} level of *Sim*, i^{th} level of *NaMol*, j^{th} level of *FA*, k^{th} level of *BFS*, and m^{th} level of *MR* and measuring the response value. The random errors are assumed to be normally distributed with mean 0 and standard deviation σ .

and the notation “P1 * P2” denotes the two-parameter interaction involving parameters P1 and P2.

The model of Equation (8-1) is referred to as an analysis of variance (ANOVA) model in which the effects associated with each model term sum to zero. The parameters *NaMol*, *FA*, *BFS*, and *MR* each only have two levels. Hence, the effect of one level of each test parameter must be equal to the negative of the effect of the second level. Because *Sim* has four levels, the sum of the effects of those four levels must sum to zero. The effects of two-parameter interactions must also sum to zero over each of the two parameters. The subsets of parameter effects summing to zero is necessary to have an effect for each level of each parameter or interaction component, given that the model contains an overall mean term.

Note that the model in Equation (8-1) has a few differences compared to the model in Equation (A-2) in Appendix A.

- The model is written as an ANOVA model rather than a regression model. In the regression model of Equation (A-2), the parameters are either numeric or coded to take numeric values, and every model term has a numeric coefficient that can be estimated from experimental data. In the ANOVA model in Equation (8-1), each level of a parameter or component of an interaction has a numeric effect, but the effects must sum to zero for a given parameter and for each of the two parameters in a two-parameter interaction.
- The model does not assume that the Average and Blend simulants have the same effect on the Cast Stone properties and that the difference between these two simulants does not depend on (i.e., interact with) other test parameters. In this sense the model in Equation (8-1) is larger than the model in Equation (A-2).
- The model does not contain a Block term (see Appendix A for the explanation of the two blocks in the testing). Preliminary data analyses considered models with Block terms, but there was no strong evidence of a block effect for any of the Cast Stone properties statistically analyzed. Hence, the final models used for the data analyses do not contain such terms.

The model in Equation (8-1) is referred to as the Full Model because it provides for estimating and evaluating all the parameter effects thought to be significant plus effects involving differences between the Average and Blend simulants (which were assumed to be nonsignificant in developing the test matrix). In addition, the Full Model allows evaluating the correctness of Assumptions A and B discussed in Section 8.1.

8.2.2 Fitting the Full Model and Summaries of Results

The relationship between each response variable and the five test parameters is quantified with a fit of the Full Model to the experimental data using ordinary least squares (OLS). OLS rest on two key assumptions:

- Errors are independent and identically distributed (i.e., have constant variance).
- The two-parameter interactions involving BFS (which were excluded from the Full Model) and higher-order interactions have no significant effects on the response variables, as emphasized in Section 8.1.

For each Cast Stone property, the structure of the errors was evaluated with diagnostic plots, Box-Cox transformations, and Levene's test for homogeneity of variance (Draper and Smith 1998). In

addition, the fact that the run order of the experiment was randomized provides some assurance of error independence. Diagnostic evaluations indicated that log-transformations on two properties (plastic viscosity and compressive strength) produce a more constant variance (see discussion of the results for those properties in Sections 8.6.1 and 8.7.1, respectively).

Table 8-1 provides an overview of the Full Model relative to the number of data points available to estimate the effects of individual parameters and two-parameter interactions depending on whether a Cast Stone property has data corresponding to Table 2-1 (26 mixes) or Table C-1 (38 mixes). When a property has data corresponding to Table 2-1 with 26 tests, the Full Model will almost be fitted exactly (except for replicate variation) because it contains 20 free parameter effects¹ to be estimated using 21 distinct data points (because 5 of the 26 data points in the test matrix are replicates). Hence, there is only one degree of freedom (DOF) for assessing model lack-of-fit (LOF). For some Cast Stone Properties, a missing property value or removing one outlier led to a data set with 25 tests, in which case there are no DOF for assessing model LOF. For the Cast Stone properties with data for 38 mixes, there are 11 DOF for assessing model LOF. The five replicates for data according to Table 2-1, and the seven replicates for data according to Table C-1, provide for estimating the pure-error standard deviation (denoted in this report as $RMSE_{PE}$).

Table 8-1. Summary of the Full Model Relative to Available Data

Model Term	# Model Effects	DOF ^(a)		
		Table 2-1, 26 Tests	Table 2-1, 25 Tests	Table 2-1, Table C-1, 38 Tests
μ (mean)	1	1	1	1
Individual Parameter Effects				
Sim	4	3	3	3
NaMol	2	1	1	1
FA	2	1	1	1
BFS	2	1	1	1
MR	2	1	1	1
Two-Parameter Interactions				
Sim*NaMol	8	3	3	3
Sim*FA	8	3	3	3
Sim*MR	8	3	3	3
NaMol*FA	4	1	1	1
NaMol*MR	4	1	1	1
FA*MR	4	1	1	1
Error				
Lack of Fit	-	1	0	11
Replicates	-	5	5	7
Total	49	26 ^(b)	25	38
(a) DOF = degrees of freedom. The DOF are less than the number of effects because of subsets of effects summing to zero.				
(b) Total DOF = the number of data points.				

¹ As mentioned in Section 8.1 and subsequently illustrated with examples in Section 8.2.4, subsets of parameter effects sum to zero in the Full Model. The number of “free parameter effects” is the number obtained by subtracting from the total number of parameter effects the number of sum-to-zero constraints that apply to the parameter effects.

The test matrices in Table 2-1 and Table C-1 do not ensure that estimates of the effects in the Full Model are orthogonal, which means that two or more effects in the model may have some degree of linear relationship (referred to as collinearity). Use of OLS attempts to account for the collinearities that exist between the model effects, but may not be entirely successful. The possible negative consequences of collinearities include 1) inflating uncertainties of effect estimates, and 2) causing one effect to be assessed as statistically significant when it is actually one or more other effects that are the true (unknown) significant effects. Table 8-2 lists the largest (in absolute value) pairwise correlations among the terms included the Full Model. As seen in Table 8-2, several terms exhibit pairwise correlations greater in absolute value than 0.50. However, it should be noted the vast majority of the pairwise correlations are below 0.33.

Table 8-2. Largest Correlations Between Pairs of Terms in the Full Model for the Property “Heat Generation at 300 hr” and Leachability Properties. Correlations for other properties differ slightly from these because of missing values and different data sets for different properties. (For Information Only)

Heat Generation at 300 Hours			Leachability Indices		
Pairwise Correlation	Pairs of Model Terms ^(a)		Pairwise Correlation	Pairs of Model Terms ^(b)	
-0.75	SimHiSO4*NaMol	FlyAsh*MixRatio	0.81	SimBlend*NaMol	SimBlend*MixRat
0.75	SimSO4*NaMol	FlyAsh*MixRatio	-0.78	SimBlend*NaMol	SimHiAl*NaMol
-0.72	SimBlend*NaMol	FlyAsh*MixRatio	-0.78	SimBlend*MixRat	SimHiAl*MixRat
-0.72	SimBlend*NaMol	SimHiAl*NaMol	-0.77	SimHiPSF*NaMol	FlyAsh*MixRat
0.71	NaMol*FlyAsh	FlyAsh*MixRatio	0.77	MixRat	SimBlend*MixRat
-0.70	SimHiAl*NaMol	SimHiSO4*NaMol	-0.73	SimBlend	SimBlend*MixRat
-0.69	SimHiSO4*MixRatio	NaMol*FlyAsh	0.70	SimHiAl*NaMol	FlyAsh*MixRat
-0.68	SimBlend*MixRatio	NaMol*FlyAsh	-0.69	SimBlend	MixRat
-0.65	SimHiSO4*FlyAsh	NaMol*MixRatio	0.68	SimHiAl*NaMol	NaMol*MixRat
0.63	SimHiAl*NaMol	SimHiAl*FlyAsh	-0.68	SimHiPSF*FlyAsh	NaMol*MixRat

(a) The notation SimHiAl denotes the high Al simulant, with similar notation for the SST Blend and high SO₄ simulants.

(b) These pairwise correlations apply for all component leachability indices that were statistically analyzed (Na, NO₃, I, Tc, Cr).

A fit of the Full Model to data for a Cast Stone property produces three sets of outputs for evaluation.

- Estimates of individual-parameter effects and two-parameter interactions, standard deviations of the estimates, and the statistical significance of the effect estimates.
- Summary statistics that quantify the adequacy of the Full Model fit.
- Table of effect differences that provides for assessing which two-level parameters have statistically significant effects and which of the four simulants have statistically different effects.

Each of these sets of outputs is discussed in the following subsections. In Sections 8.6 and 8.7, all three sets of output are summarized in one table (e.g., see Table 8-10 in Section 8.6.1).

8.2.3 Summary Statistics to Assess Model Adequacy

Several statistics used to summarize the adequacy of a model fit to data are discussed in this subsection. These statistics apply to fitting any model, including the Full Model (discussed in Section 8.2) and a Stepwise Model (discussed in Section 8.3).

The root mean squared error (RMSE) is given by

$$RMSE = \left(\frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{n - p} \right)^{0.5} \quad (8-2)$$

where y_i is the property value for the i^{th} data point, \hat{y}_i is the model-predicted property value for the i^{th} data point, n is the number of data points used to fit the model, and p is the number of coefficients (or free parameter effects, see Section 8.2.2). If a property is mathematically transformed to satisfy the OLS regression assumptions (see Section 8.2.2), then the transformed values are used in Equation (8-2) as well as the following equations. RMSE includes variation due to model LOF as well as variation among replicate tests (referred to as *pure error*).

The RMSE for pure error is given by

$$RMSE_{PE} = \left(\frac{\sum_{k=1}^K \sum_{i=1}^{n_k} (y_{ki} - \bar{y}_k)^2}{\sum_{k=1}^K (n_k - 1)} \right)^{0.5} \quad (8-3)$$

where y_{ki} is the property value for the i^{th} data point in the k^{th} set of replicates, \bar{y}_k is the mean of the property values for the k^{th} set of replicates, and n_k is the number of replicates in the k^{th} set of replicates.

The R^2 statistic is defined by

$$R^2 = 1 - \frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{\sum_{i=1}^n (y_i - \bar{y})^2} \quad (8-4)$$

where \bar{y} is the mean of the property values over all n data points, and the other notation is as defined following Equation (8-2). R^2 quantifies the fraction of variation in values of a response variable for a data set that is accounted for by a model fitted to the data. Hence, R^2 takes values between 0 and 1. R^2 cannot equal 1 when there are replicate data (a model can never account for replicate variability). However, R^2 will increase and approach its maximum possible value as the number of parameter effects in a model approaches the number of distinct data points (i.e., without counting replicate pairs twice).

8.2.4 Estimates of Parameter Effects for the Full Model

As discussed in Section 8.2.2, there are 20 free parameter effects out of a total of 49 in the Full Model. Evaluating the results of fitting the Full Model is therefore simplified by concentrating on the free parameter effects. For all individual parameter effects and two-parameter interactions not involving the Simulant parameter, each model term is described by only one free parameter effect. Hence, determining the statistical significance of one of these model terms reduces to an evaluation of its one free parameter effect. For example, Table 8-8 in Section 8.6.1 contains the results for fitting the Full Model to $\ln(\text{Plastic Viscosity})$ data. In Table 8-8, the estimate of the free parameter effect associated with MixRatio is 0.770. This estimate actually refers to the first level of MixRatio, so 0.770 really represents the estimated effect of MixRatio = 0.4 on $\ln(\text{Plastic Viscosity})$. The estimate for MixRatio = 0.6 is therefore -0.770 , the negative of the effect for the first level (see the discussion of why these effects must sum to zero in Section 8.2.1). For a two-parameter interaction, such as NaMol*FlyAsh, the estimate is associated with the first level of each of the two parameters. Thus the estimate of -0.023 in Table 8-8 actually represents the effect of NaMol = 5.0 and FlyAsh = NW. Estimates of all two-parameter interaction effects associated with NaMol*FlyAsh are as shown in Table 8-3.

Table 8-3. Estimate of All Two-Parameter Interaction Effects Associated with the NaMol*FlyAsh Interaction

NaMol	FlyAsh	
	NW	SE
5.0	-0.023	0.023
7.8	0.023	-0.023

For model terms that involve the Simulant parameter (either individually or in interactions), the interpretations are more complex. For the Simulant model term, four estimated effects are produced (see Table 8-8), each associated with one of the four simulants. These effect estimates are constrained to sum to zero as discussed in Section 8.2.1. For an interaction involving Simulant, half the estimates (four of eight) are produced. For example, the four estimates listed in Table 8-8 for “Simulant*NaMol” are associated with the first level of NaMol = 5.0. The complete list of parameter effect estimates is obtained by taking the negatives of listed estimates as shown in Table 8-4.

Table 8-4. Complete List of Parameter Effect Estimates Associated with the “Simulant*NaMol” Interaction

NaMol	Simulant			
	Average	Blend	HiAl	HiSO ₄
5.0	-0.053	-0.173	-0.026	0.251
7.8	0.053	0.173	0.026	-0.251

8.2.5 Assessment of Whether the Four Simulants Have Different Effects on Cast Stone Properties

Because the Simulant parameter has four levels (i.e., four simulants), it is not sufficient to examine the statistical significance of the effect of each simulant individually. It is of interest to know whether the effects of the four simulants on a Cast Stone property are statistically different.

To accomplish this, all pairwise differences of effects of the four simulants are considered and p-values¹ are calculated to determine which simulants have statistically different effects. Because there are six pairwise comparisons of four estimates, a multiple comparison procedure (MCP) is used to produce the p-values that limit the probability of a wrong decision over all six of the comparisons. The MCP procedure used is a generalization of the Tukey Honestly Significant Difference (HSD) procedure, which is discussed by Bretz et al. (2010, 2013) and Hothorn et al. (2013). A generalization of the standard version of the Tukey HSD procedure was used because the assumptions of the standard version (independent estimates, equal variances) are not satisfied.

The last column of Table 8-10 illustrates a high-level summary of the results from the Tukey HSD procedure for the “Heat Generation at 300 Hours” property. Simulants with the same letter (A and B) are not statistically different with the probability 0.05 of an incorrect conclusion of a difference over all six pairwise differences. Hence, simulants that do not share any of the letters listed have statistically different effects. As an example, consider the summary of the results for the individual effects of the four simulants. The simulants with the letter A (Average, SST Blend, and Hi SO₄) do not have statistically different effects on “Heat Generation at 300 Hours”. Similarly, the simulants with the letter B (Average, Hi Al, and Hi SO₄) do not have statistically different effects on the property. Only the SST Blend and Hi Al simulants do not share any of the letters listed, so they have statistically different effects on the property. This is consistent with the effect estimate for “SST Blend” having the largest negative effect among the simulants (–25.86) and “Hi Al” having the largest positive effect (17.97).

8.3 Stepwise Regression Modeling Approach, Including the Possibility of Interactions Involving BFS

To evaluate the validity of Assumption C in Section 8.1 (that no interactions involving BFS are statistically significant), stepwise regression (Draper and Smith 1998) was used. Stepwise regression is a procedure that starts with no terms in a model or some terms forced into the model, and iteratively builds a model by adding or removing the term that most improves the optimality criterion being used to quantify the model goodness-of-fit.

One significant difference in a regression model (e.g., a Stepwise Model) compared to an ANOVA model (e.g., the Full Model) is that all of the parameters in a regression model must have numeric values and not have exact constraints (such as groups of parameter effects summing to zero as occur with the Full (ANOVA) model). Because the four possible simulants (Average, SST Blend, Hi Al, and Hi SO₄) are categorical values of the Simulant parameter, it is not possible for them to all appear in a regression model at the same time. The Stepwise Models for Cast Stone properties were constructed by omitting all terms involving the Average simulant. Hence, the individual effect of the SST Blend, Hi Al, or Hi SO₄ that is statistically significant actually corresponds to that simulant having a statistically different effect than the Average simulant.

For the Cast Stone screening data, the model terms corresponding to all individual parameter effects (except the Average simulant) were forced into the model, and all two-parameter interactions (including those with BFS as one of the parameters and those involving the Average simulant) were candidates for addition to the model. During each iteration of the stepwise regression, the term that most improves the model fit is added to the model, provided the

¹ A p-value is the probability of being wrong in concluding a parameter effect is not statistically significant. Small p-values (e.g., < 0.05) involve a small probability of incorrectly concluding a parameter effect is not statistically significant. Hence, parameter effects with p-values < 0.05 are declared as statistically significant.

coefficient of the term is statistically significant ($p\text{-value} \leq 0.05$). After a term is added to the model in an iteration, all of the terms added to the model are statistically assessed to make sure they are still statistically significant. Because of collinearities between model terms resulting from a particular test matrix, it is possible that a term that was statistically significant when first added to the model during an earlier iteration may no longer be statistically significant at a later iteration. If this occurs, the term is removed from the model and the stepwise regression process continues with the next iteration of adding the best remaining term. The stepwise regression process terminates when no statistically significant term can be added and all terms in the model are statistically significant. The stepwise model development was performed using the R software (R Core Team 2013, Crawley 2007).

An example of the output of the stepwise modeling approach is given in Table 8-9, which is discussed in Section 8.6.1. The statistics discussed in Section 8.2.3 for summarizing the model fit to the data are included in the results table for each stepwise regression model in this report.

8.4 Cautions Regarding Interpreting the Results of Fitting the Full Model and Stepwise Models

This section discusses cautions regarding interpreting the results of fitting a Full and Stepwise Models to Cast Stone property data.

There are two main cautions associated with interpreting the results of a fitted Full Model.

Caution FM1: For Cast Stone properties that had 26 mixes tested, some properties had missing data for one test or one outlier was removed before fitting models. In such cases, the number of free parameter effects (as discussed in Section 8.2.5) in the Full Model is exactly the same as the number of distinct mixes tested (20) and R^2 will achieve its maximum possible value. It is impossible to statistically assess the model LOF in this case. For Cast Stone properties that had 26 mixes tested without a missing data point or a removed outlier, the number of free parameter effects in the Full Model is one less than the number of distinct mixes tested (21) and R^2 will be very close to its maximum possible value. While it is possible to statistically assess the model LOF in this latter case, it is less likely that the LOF will be statistically significant with only one “extra” distinct mix. Hence, it must be recognized in assessing the results of fitting the Full Model to Cast Stone properties with data for 26 or 25 mixes that the R^2 value will be very close or equal to its maximum possible value and LOF tests may be of limited value or not possible.

Caution FM2: The tabular outputs of fitting the Full Model and a Stepwise Model to each Cast Stone property (for which statistical modeling was conducted) include p-values for assessing the statistical significance of each model term. The Full Model may include many terms associated with parameters or two-parameter interactions that are not statistically significant. Not only can this inappropriately inflate R^2 , it can cause terms with statistically significant effects to appear to have statistically nonsignificant effects. The latter can occur because of collinearities (i.e., correlations) among parameter individual effects and two-parameter interactions. However, if terms in a fitted Full Model are statistically significant, it generally can be believed that they do have significant effects.

There are also two main cautions associated with interpreting the results of a fitted Stepwise Model.

Caution SM1: There are collinearities (correlations) among the individual parameter terms and two-parameter interaction terms selected for a Stepwise Model, and these depend on the number of tests and the parameter combinations tested. Hence, a resulting Stepwise Model may not contain all of the true (unknown) individual parameter and two-parameter interaction terms with significant effects. Instead, a Stepwise Model could contain terms that are correlated with the true, unknown parameter effects.

Caution SM2: Stepwise regression procedures tend to include too many terms in models that are not among the true (unknown) terms.

One way to “protect” against SM2 is to use a tighter criterion to control model terms that are added to or remain in a model. In this report a p-value < 0.05 is used to both determine what terms are added to a model and which terms are removed. However, the results of Stepwise Models in this report include the p-values of terms in the model, so the viewer can always impose a tighter constraint (e.g., p-value < 0.01) to increase the chance that terms declared statistically significant correspond to true (unknown) effects of parameters on a Cast Stone property.

There are both pros and cons to the Full Model and Stepwise Model approaches chosen to statistically analyze selected Cast Stone properties. The Full Model approach avoids the concerns SM1 and SM2 associated with stepwise regression, but has the Caution FM1 and FM2 concerns. On the other hand, the stepwise regression approach avoids the Caution FM1 and FM2 concerns by building models that only contain statistically significant terms, but then has the concerns SM1 and SM2. Ultimately, both the Full Model and Stepwise Model approaches are useful, provided that the respective concerns are kept in mind when considering the model fits to Cast Stone property data (in Sections 8.6 and 8.7). The results of both approaches should be considered along with subject matter knowledge and experience to reach conclusions about which individual parameter effects and two-parameter effects are statistically significant and also meaningful from a subject matter perspective.

8.5 Estimates of Testing and Measurement Uncertainties of Cast Stone Properties from Replicate Tests

As discussed in Section 2.1, there are five pairs of replicate tests for Cast Stone properties for which 26 tests were performed (see Table 2-1), and there are seven pairs of replicate tests for Cast Stone properties for which 38 tests were performed (see Table C-1). The replicate pairs provide for calculating the uncertainty of Cast Stone property values due to the testing and measurement processes. Before presenting and discussing quantitative estimates of the combined testing and measurement uncertainties for Cast Stone properties, it is important to emphasize the difference between *replicates*, and what in this report are referred to as *repeats*. More specifically, for some locations in the report, repeats are referred to as *duplicates* or *triplicates*.

Repeats (which are different than replicates) are tests performed on two or three samples of a given Cast Stone mix. That is, the mix was made up only once, and two or three samples of it were tested to provide duplicate or triplicate results. On the other hand, replicates involved completely remaking a Cast Stone mix and measuring properties at a different time in the run order of tests. Typically, replicates are expected to have additional uncertainty over and above the uncertainty from repeats, because of making a mix and measuring properties in different

periods of time, potentially with different personnel and/or equipment. For Cast Stone properties that had repeat tests, the means and standard deviations of the repeat property values are listed in tables of Appendices C and D. The means over repeats were used as the property values in statistical data analyses.

Table 8-5 to Table 8-7 summarize (for each Cast Stone property) the uncertainties as SDs and %RSDs for each replicate pair, and also pooled (i.e., combined) over the five or seven replicate

Table 8-5. Replicate Property Values, Standard Deviations (SDs), and Percent Relative Standard Deviations (%RSDs), Along with Pooled SDs and %RSDs Using All Replicate Pairs (For Information Only)

Replicate Pair Mix #s	Plastic Viscosity (cP)	Heat Generation		Compressive Strength, PNNL (psi)
		at 300 hr (J/g)	Time to Peak (hr)	
2	86.0	382.3	73.87	4825
13	92.0	351.8	71.93	4840
SD	4.24	21.57	1.372	10.61
%RSD	4.77	5.88	1.88	0.22
3	80.0	394.0	53.48	2500
22	91.0	397.0	55.70	2397
SD	7.78	2.12	1.570	72.83
%RSD	9.10	0.54	2.88	2.97
7	558.0	334.6	47.27	6444
14	602.0	340.9	45.92	6669
SD	31.11	4.45	0.955	159.10
%RSD	5.36	1.32	2.05	2.43
15	278.0	314.0	52.93	4977
25	287.0	291.0	53.25	6179
SD	6.36	16.26	0.226	849.94
%RSD	2.25	5.38	0.43	15.24
21	93.0	271.0	51.78	1359
26	91.0	272.0	50.72	947
SD	1.41	0.71	0.750	291.33
%RSD	1.54	0.26	1.46	25.27
27 (3a)	88.0	325.2	53.48	(a)
36 (22a)	85.0	327.0	55.70	(a)
SD	2.12	1.27	1.570	(a)
%RSD	2.45	0.39	2.88	(a)
33 (15a)	202.0	318.0	52.93	(a)
38 (25a)	236.0	323.0	53.25	(a)
SD	24.04	3.54	0.226	(a)
%RSD	10.98	1.10	0.43	(a)
Pooled^(b)				
SD	15.45	10.48	1.095	409.39
%RSD	6.19	3.09	1.96	13.31
(a) Not applicable, because these mixes are ones SRNL mis-batched and tested.				
(b) Pooled SDs and %RSDs were calculated by squaring the SDs and %RSDs for each replicate pair, averaging the squared values over the five replicate sets, then taking the square root. The formula is simpler than the general formula because each SD or %RSD is calculated from the same number of values (two).				

pairs (as applicable for a given property). The SDs and %RSDs are useful as quantitative measures of the uncertainty inherent in the Cast Stone property data from testing, and also provide the basis for assessing whether the effects of parameters on properties are statistically significant. Table 8-5 presents the replicate property values and uncertainties for all properties that were chosen for statistical analysis except properties associated with EPA Method 1315. Table 8-6 and Table 8-7 present replicate property values and uncertainties for effective diffusion coefficients and leachability indices, respectively, associated with EPA Method 1315 for leach intervals between 28 and 63 days cumulative time.

Table 8-6. Replicate Values, Standard Deviations (SDs), and Percent Relative Standard Deviations (%RSDs) for Effective Diffusion Coefficients from EPA 1315 Leach Tests, Along with and Pooled SDs and %RSDs Using All Replicate Pairs (For Information Only)

Replicate Pair Mix #s	Effective Diffusion Coefficient (cm ² /s)					
	I	Tc	Na	NO ₃	NO ₂	Cr
2	7.02E-09	5.99E-11	6.92E-09	6.65E-09	7.50E-09	8.22E-14
13	8.48E-09	8.27E-11	6.44E-09	8.75E-09	7.47E-09	5.51E-14
SD	1.03E-09	1.61E-11	3.39E-10	1.48E-09	2.12E-11	1.92E-14
%RSD	1.33E+01	2.26E+01	5.08E+00	1.93E+01	2.83E-01	2.79E+01
3	4.98E-09	3.47E-11	4.99E-09	4.60E-09	5.44E-09	1.64E-13
22	4.42E-09	2.48E-11	4.84E-09	4.32E-09	4.14E-09	1.79E-13
SD	3.96E-10	7.00E-12	1.06E-10	1.98E-10	9.19E-10	1.06E-14
%RSD	8.43E+00	2.35E+01	2.16E+00	4.44E+00	1.92E+01	6.18E+00
7	4.25E-09	9.06E-12	3.52E-09	4.72E-09	4.68E-09	8.28E-15
14	5.70E-09	9.61E-12	4.44E-09	6.26E-09	5.93E-09	9.90E-15
SD	1.03E-09	3.89E-13	6.51E-10	1.09E-09	8.84E-10	1.15E-15
%RSD	2.06E+01	4.17E+00	1.63E+01	1.98E+01	1.67E+01	1.26E+01
15	5.67E-09	4.62E-11	7.33E-09	7.01E-09	7.17E-09	3.16E-13
25	4.87E-09	2.88E-11	6.86E-09	6.99E-09	5.80E-09	4.02E-13
SD	5.66E-10	1.23E-11	3.32E-10	1.41E-11	9.69E-10	6.08E-14
%RSD	1.07E+01	3.28E+01	4.68E+00	2.02E-01	1.49E+01	1.69E+01
21	1.68E-08	9.44E-11	1.05E-08	1.52E-08	1.38E-08	1.51E-13
26	7.30E-09	1.66E-10	8.07E-09	7.85E-09	7.09E-09	1.02E-13
SD	6.72E-09	5.06E-11	1.72E-09	5.20E-09	4.74E-09	3.46E-14
%RSD	5.57E+01	3.89E+01	1.85E+01	4.51E+01	4.54E+01	2.74E+01
Pooled^(a)						
SD	3.09E-09	2.46E-11	8.50E-10	2.47E-09	2.24E-09	3.28E-14
%RSD	2.79E+01	2.71E+01	1.15E+01	2.37E+01	2.42E+01	2.01E+01
(a) Pooled SDs and %RSDs were calculated by squaring the SDs and %RSDs, averaging the squared values over the five replicate sets, then taking the square root. The formula is simpler than the general formula because each SD or %RSD is calculated from the same number of values (two).						

Table 8-7. Replicate Values, Standard Deviations (SDs), and Percent Relative Standard Deviations (%RSDs) for Leachability Index Calculated from Effective Diffusion Coefficients from EPA 1315 Leach Tests, Along with and Pooled SDs and %RSDs Using All Replicate Pairs (For Information Only)

Replicate Pair Mix #s	Leachability Index ($-\log_{10}(\text{cm}^2/\text{s})$)					
	I	Tc	Na	NO ₃	NO ₂	Cr
2	8.180	10.230	8.200	8.200	8.170	13.090
13	8.090	10.090	8.210	8.090	8.140	13.270
SD	0.0636	0.0990	0.0071	0.0778	0.0212	0.1273
%RSD	0.7823	0.9744	0.0862	0.9550	0.2601	0.9657
3	8.300	10.460	8.320	8.340	8.270	12.800
22	8.360	10.630	8.320	8.370	8.390	12.770
SD	0.0424	0.1202	0.0000	0.0212	0.0849	0.0212
%RSD	0.5093	1.1400	0.0000	0.2539	1.0186	0.1659
7	8.430	11.050	8.480	8.380	8.380	14.110
14	8.270	11.030	8.360	8.230	8.250	14.020
SD	0.1131	0.0141	0.0849	0.1061	0.0919	0.0636
%RSD	1.3549	0.1281	1.0078	1.2771	1.1055	0.4525
15	8.270	10.340	8.140	8.180	8.170	12.650
25	8.340	10.550	8.180	8.180	8.250	12.560
SD	0.0495	0.1485	0.0283	0.0000	0.0566	0.0636
%RSD	0.5960	1.4217	0.3466	0.0000	0.6890	0.5049
21	7.810	10.160	8.000	7.850	7.880	12.860
26	8.170	9.880	8.110	8.130	8.160	13.030
SD	0.2546	0.1980	0.0778	0.1980	0.1980	0.1202
%RSD	3.1860	1.9759	0.9656	2.4780	2.4687	0.9286
Pooled^(a)						
SD	0.1311	0.1309	0.0531	0.1067	0.1082	0.0885
%RSD	1.6256	1.2799	0.6443	1.3227	1.3339	0.6756
(a) Pooled SDs and %RSDs were calculated by squaring the SDs and %RSDs, averaging the squared values over the five replicate sets, then taking the square root. The formula is simpler than the general formula because each SD or %RSD is calculated from the same number of values (two).						

8.6 Results of Statistical Analyses – Processing Properties

The results of the statistical analysis methods described in Sections 8.1 to 8.3 are presented and discussed in Section 8.6.1 for plastic viscosity and in Section 8.6.2 for two heat-generation properties.

8.6.1 Slurry Rheology—Plastic Viscosity

Two slurry/paste rheology properties (Yield Stress and Plastic Viscosity) were measured for the 38 Cast Stone mixes listed in Table C-1, with the data summarized in Table C-5 in Appendix C. No values of the two properties were obtained for Mix 1 because the Cast Stone paste was too thick. A scatterplot of the data for these two properties is shown in Figure 8-1. It is not clear whether there is 1) a fairly strong curvilinear relationship between these two properties with the data for Mix/Test 20, 28, and 35 being outliers, or 2) a strong linear relationship for lower values of the two parameters, but essentially no relationship for large values. Note that Mixes 20, 28, and 35 are more clearly potential outliers for Yield Stress (since they are the largest values observed). Because only Plastic Viscosity was selected for statistical analyses, and Mixes 20, 28,

and 35 have values of that property in the middle of its range of values, they are less likely to be outliers, but it is still possible. SRNL who performed the Plastic Viscosity tests found no clear reason for these tests to be questionable, so they were retained in the statistical data analyses.

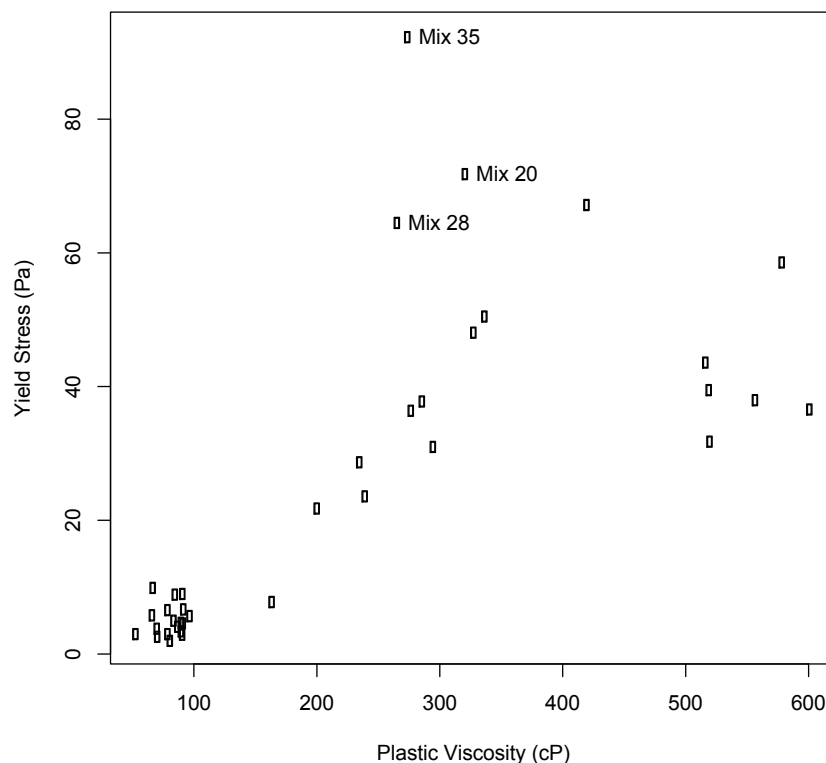


Figure 8-1. Scatterplot of Yield Stress versus Plastic Viscosity for the 38 Cast Stone Mixes. Outlying data points are labeled with the mix/test number. (For Information Only)

The Plastic Viscosity values for the 38 Cast Stone mixes listed in Table C-5 range from 54 to 602 cP with a mean of 228 cP. Table 8-5 lists the Plastic Viscosity values for the seven replicate pairs and presents uncertainties in the plastic viscosity values as calculated from the replicates. The pooled (i.e., combined) uncertainty estimates using all replicate pairs are SD = 15.45 cP and %RSD = 6.19. The range in Plastic Viscosity values (over a factor of 10) must be a result of variations in the test parameters, because the pooled %RSD is about 6%. The balance of this section presents and discusses for Plastic Viscosity the results of applying the statistical analysis methods described in Sections 8.1 to 8.3. Note that the natural logarithm of Plastic Viscosity [denoted $\ln(\text{Plastic Viscosity})$] was statistically analyzed because modeling diagnostics indicated that the transformation was necessary to satisfy the constant variance assumption for OLS regression (see Section 8.2.2). Viscosities of many kinds of materials are traditionally modeled using the natural logarithm transformation.

Table 8-8 summarizes the results of fitting the Full Model to the $\ln(\text{Plastic Viscosity})$ data and statistically assessing the effects of individual parameters and two-parameter interactions (except

Table 8-8. In(Plastic Viscosity): Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

In(Plastic Viscosity) Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Signif- icance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	5.2330	0.0428	<0.0001	****	
Simulant					
Average	0.0172	0.0605	0.7794		A
SST Blend	-0.0017	0.0570	0.9771		A
Hi Al	-0.0874	0.0601	0.1641		A
Hi SO ₄	0.0718	0.0803	0.3838		A
NaMol	0.0031	0.0455	0.9461		
FlyAsh	0.2278	0.0377	<0.0001	****	
BFS	0.0741	0.0311	0.0291	*	
MixRatio	0.7696	0.0364	<0.0001	****	
Simulant*NaMol					
Average	-0.0526	0.0851	0.5451		A
SST Blend	-0.1726	0.0933	0.0817		A
Hi Al	-0.0255	0.0847	0.7670		A
Hi SO ₄	0.2507	0.1154	0.0443	*(f)	A
Simulant*FlyAsh					
Average	-0.0594	0.0687	0.3990		A
SST Blend	0.0680	0.0762	0.3844		A
Hi Al	-0.1474	0.0725	0.0579		A
Hi SO ₄	0.1388	0.0760	0.0855		A
Simulant*MixRatio					
Average	-0.0508	0.0828	0.5475		A
SST Blend	-0.0735	0.0857	0.4036		A
Hi Al	-0.0235	0.0681	0.7338		A
Hi SO ₄	0.1478	0.0878	0.1107		A
NaMol*FlyAsh	-0.0232	0.0639	0.7206		
NaMol*MixRatio	-0.0071	0.0549	0.8988		
FlyAsh*MixRatio	0.0713	0.0678	0.3075		
Summary Statistics of Model Fit					
RMSE = 0.1616		RMSE _{PE} = 0.0627	R ² = 0.9799	LOF p-value = 0.0024	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					
(f) It is possible that a simulant has a statistically significant effect with p-value < 0.05 but that no simulants have statistically significant differences when assessed with the MCP, which controls the probability of a wrong decision over all possible comparisons.					

interactions involving BFS), as discussed in Section 8.2. Keep in mind that the Caution FM2 discussed in Section 8.4 applies to the discussion of the results in Table 8-8. The Full Model fits the $\ln(\text{Plastic Viscosity})$ data very well, accounting for a high fraction of the variation in the data ($R^2 = 0.9799$). However, the $\text{RMSE} = 0.1616$ is significantly larger than the $\text{RMSE}_{\text{PE}} = 0.0627$, and hence the model LOF is statistically significant ($p\text{-value} = 0.002$). Still, the Full Model with $R^2 = 0.9799$ can be used to assess which parameters have statistically significant effects on Plastic Viscosity. Table 8-8 shows that FlyAsh and MixRatio have highly significant effects ($p\text{-values} < 0.001$ confidence) and that the effect of BFS is significant ($p\text{-value} < 0.05$). The $(\text{SimHiSO}_4) * \text{NaMol}$ interaction is also statistically significant ($p\text{-value} < 0.05$). The last column in Table 8-8 shows that there are no statistically significant differences between the simulants for either individual effects or for interactions of other parameters with simulant.

Table 8-9 summarizes the results of using the stepwise regression approach described in Section 8.3. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-9. The Stepwise Model fits the $\ln(\text{Plastic Viscosity})$ data reasonably well, accounting for a high fraction of the variation in the data ($R^2 = 0.9572$). This R^2 is smaller than for the Full Model ($R^2 = 0.9799$), but that may be because the latter value is inflated per SM2. The $\text{RMSE} = 0.1835$ is significantly larger than the $\text{RMSE}_{\text{PE}} = 0.0627$, and hence the model LOF is highly statistically significant ($p\text{-value} = 0.0016$). Still, the Stepwise Model in Table 8-9 with $R^2 = 0.9572$ can be used to assess the parameters that have statistically significant effects on plastic viscosity. Table 8-9 shows that several individual parameters have statistically significant effects, including FlyAsh and MixRatio ($p\text{-values} < 0.0001$). The only interaction that is statistically significant is FlyAsh*MixRatio ($p\text{-value} < 0.001$). No interactions involving BFS are statistically significant, suggesting that BFS interactions do not appear to affect Plastic Viscosity.

Table 8-9. $\ln(\text{Plastic Viscosity})$: Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

In(Plastic Viscosity) Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	5.2110	0.0331	157.47	0.0000	****
SimBlend	0.0206	0.0589	0.35	0.7289	
SimHiAl	-0.0145	0.0548	-0.26	0.7932	
SimHiSO4	-0.0610	0.0531	-1.15	0.2604	
NaMol	-0.0341	0.0335	-1.02	0.3170	
FlyAsh	0.2054	0.0322	6.38	0.0000	****
BFS	0.0535	0.0308	1.74	0.0932	
MixRatio	0.7577	0.0330	22.96	0.0000	****
FlyAsh*MixRatio	0.1178	0.0321	3.67	0.0010	***
Summary Statistics for Model Fit					
RMSE = 0.1835		RMSE _{PE} = 0.0627		R ² = 0.9572	
				LOF p-value = 0.0016	
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

8.6.2 Heat Generation

The results of statistical analyses for two heat-generation properties are discussed in this subsection. Heat Generation at 300 Hours (HG300) is covered in Section 8.6.2.1, while Time to Peak Heat Generation (TPHG) is covered in Section 8.6.2.2. A third property, Heat Generation at Peak (HGP), was also measured but was not statistically analyzed. The values of all three properties for 38 Cast Stone mixes given in Table C-1 are listed in Table C-8 in Appendix C. Figure 8-2 shows a scatterplot matrix graph of the three heat-generation properties. This figure shows no strong correlations between pairs of the three properties when all data are considered. However, if the outlying data points are ignored, the property pairs (HG300, TPHG) and (HG300, HGP) do exhibit positive correlations. See Section 6.4 for further discussion of the approach and results from measuring these properties.

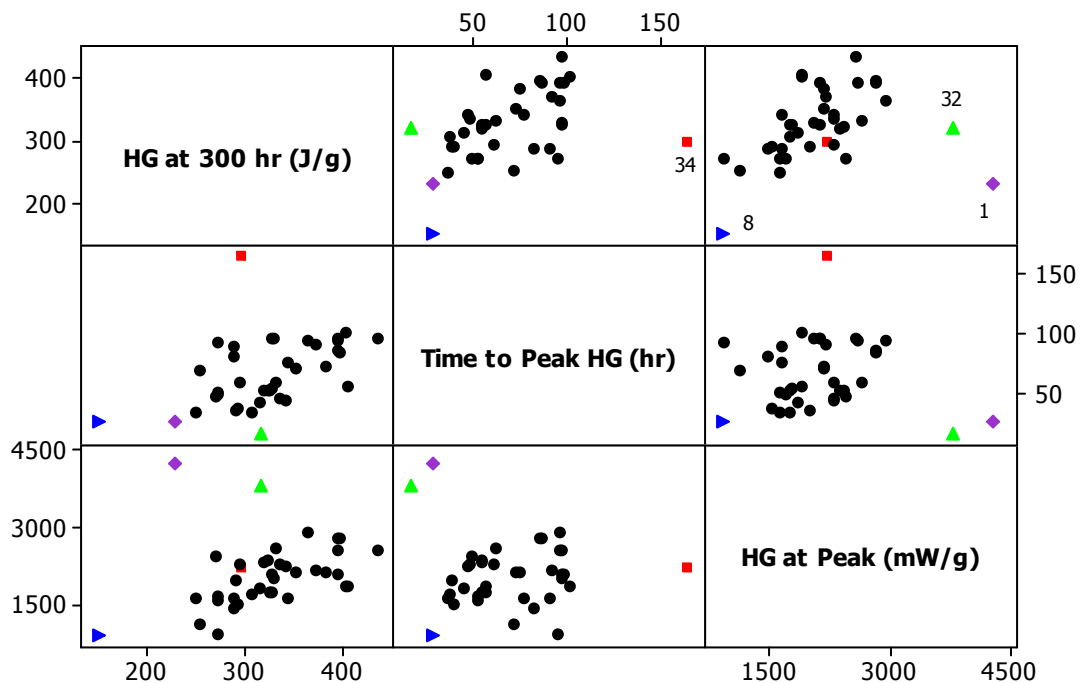


Figure 8-2. Scatterplot Matrix of Data on Three Heat Generation (HG) Properties for the 38 Cast Stone Mixes. Certain outlying points are plotted with color symbols and their mix numbers are identified in one of the plots. (For Information Only)

8.6.2.1 Heat Generation at 300 Hours

Table C-8 in Appendix C lists the HG300 values for 38 Cast Stone mixes in Table C-1, which range from 150 to 434 J/g. Figure 8-2 shows the distribution of the HG300 values. Table 8-5 lists the HG300 values for the seven replicate pairs and presents uncertainties in the HG300 values as calculated from the replicates. The pooled (i.e., combined) uncertainty estimates using all replicate pairs are $SD = 10.48$ J/g and $\%RSD = 3.09$, which are relatively small. The range in HG300 values (nearly a factor of three) must be a result of variations in the test parameters, because the pooled $\%RSD$ is only 3%. The balance of this section presents and discusses the results of the statistical analysis methods described in Sections 8.1 to 8.3.

Table 8-10 summarizes the results of fitting the Full Model to the HG300 data and statistically assessing the effects of individual parameters and their two-parameter interactions, as discussed in Section 8.2. Keep in mind that Caution FM2 discussed in Section 8.4 applies to the results in Table 8-10. The Full Model fits the HG300 data moderately well, accounting for a large fraction of the variation in the data, indicated by $R^2 = 0.9298$. However, the $RMSE = 21.716$ is approximately twice the $RMSE_{PE} = 10.478$, and hence the model LOF is statistically significant (p-value = 0.0107). However, the R^2 is large enough that the Full Model in Table 8-10 can be used to assess which parameters have statistically significant effects. Table 8-10 shows that the individual parameter effects for FlyAsh, BFS, and MixRatio are highly statistically significant (p-values < 0.0001). Also statistically significant are the effects for the SST Blend (p-value < 0.01) and Hi Al simulants (p-value < 0.05). As noted in Section 8.3, this means that the SST Blend and Hi Al simulants have statistically different effects than the Average simulant. None of the two-parameter interaction terms are statistically significant (i.e., all p-values > 0.05).

Table 8-11 summarizes the results of using the stepwise regression approach described in Section 8.3. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-11. Because more terms are available for the fit, the Stepwise Model fits the HG300 data better than the Full Model, producing a higher $R^2 = 0.9526$ and a model LOF that is just barely significant (p-value = 0.0452). However, the R^2 is large enough that the Stepwise Model in Table 8-11 can be used to assess which parameters have statistically significant effects. Table 8-11 shows that the individual parameter effects for FlyAsh, BFS, and MixRatio are highly statistically significant (p-values < 0.0001). Also statistically significant are the effects for the SST Blend and Hi Al simulants (p-values < 0.01). As noted above, this means that the SST Blend and Hi Al simulants have statistically different effects than the Average simulant. The Stepwise Model also shows that two two-parameter interaction terms are statistically significant: FlyAsh*BFS (p-values < 0.01) and NaMol*MixRatio (p-value < 0.05).

The principal objective for fitting the Stepwise Model was to evaluate the importance of two-parameter interactions involving BFS. Only one such interaction term is statistically significant. Still, this indicates that BFS has both an individual effect as well as an interactive effect on HG300.

Table 8-10. Heat Generation at 300 Hours: Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

Heat Generation at 300 hr Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Significance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	313.2296	4.4728	<0.0001	****	
Simulant					
Average	6.0579	7.2914	0.4170		AB
SST Blend	-25.8594	7.3297	0.0024	**	A
Hi Al	17.9693	7.6501	0.0304	*	B
Hi SO ₄	1.8323	6.7630	0.7895		AB
NaMol	-4.5302	4.2384	0.2993		
FlyAsh	-21.9614	4.1933	0.0001	****	
BFS	-27.7803	4.0530	<0.0001	****	
MixRatio	-32.6905	4.1331	<0.0001	****	
Simulant*NaMol					
Average	13.4088	10.7610	0.2287		A
SST Blend	-20.1723	11.3380	0.0921		A
Hi Al	5.4440	11.2370	0.6339		A
Hi SO ₄	1.3196	10.8555	0.9046		A
Simulant*FlyAsh					
Average	-2.5102	9.2256	0.7886		A
SST Blend	-5.0343	9.4196	0.5996		A
Hi Al	0.5534	9.6513	0.9549		A
Hi SO ₄	6.9911	8.5171	0.4225		A
Simulant*MixRatio					
Average	3.1876	11.1304	0.7778		A
SST Blend	-12.6828	10.5125	0.2433		A
Hi Al	2.0524	8.9021	0.8203		A
Hi SO ₄	7.4427	9.6008	0.4483		A
NaMol*FlyAsh	1.2445	8.0205	0.8784		
NaMol*MixRatio	-13.8591	6.9293	0.0608		
FlyAsh*MixRatio	9.0312	9.1049	0.3344		
Summary Statistics of Model Fit					
RMSE = 21.716		RMSE _{PE} = 10.478	R ² = 0.9298	LOF p-value = 0.0107	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					

Table 8-11. Heat Generation at 300 Hours: Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

Heat Generation at 300 hr Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	315.939	3.237	97.61	<0.0001	****
SimBlend	-20.599	5.780	-3.56	0.0013	**
SimHiAl	18.235	5.378	3.39	0.0021	**
SimHiSO4	3.022	5.070	0.60	0.5559	
NaMol	-4.228	3.121	-1.35	0.1863	
FlyAsh	-19.727	3.071	-6.42	<0.0001	****
BFS	-34.737	3.404	-10.21	<0.0001	****
MixRatio	-31.335	3.071	-10.20	<0.0001	****
FlyAsh*BFS	-11.658	3.404	-3.43	0.0019	**
NaMol*MixRatio	-8.632	3.611	-2.39	0.0238	*
Summary Statistics for Model Fit					
RMSE = 17.930		RMSE _{PE} = 10.478		R ² = 0.9526	LOF p-value = 0.0452
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

8.6.2.2 Time to Peak Heat Generation

Table C-8 in Appendix C lists the TPHG values for 38 Cast Stone mixes in Table C-1, which range from 15.97 to 164.07 hr. Figure 8-2 shows the distribution of the TPHG values. The largest TPHG value of 164.07 hr (for Mix 34) is considerably larger than the next largest value of 100.90 hr (for Mix #37). Because Mix 34 was identified as a significant outlier in preliminary fits of the Full Model and Stepwise Model, the TPHG value for Mix 34 was removed for the model fits presented in this section. Table 8-5 lists the TPHG values for the seven replicate pairs and presents uncertainties in the TPHG values as calculated from the replicates. The pooled (i.e., combined) uncertainty estimates using all replicate pairs are SD = 1.10 hr and %RSD = 1.96, which are relatively small. The range in TPHG values (over a factor of 10) must be a result of variations in the test parameters, because the pooled %RSD is about 2%. The balance of this section presents and discusses the results of the statistical analysis methods described in Sections 8.1 to 8.3.

Table 8-12 presents the results of fitting the Full Model to the TPHG data and statistically assessing the effects of individual parameters and their two-parameter interactions, as discussed in Section 8.2. Keep in mind that Cautions FM1 and FM2 discussed in Section 8.4 apply to the results in Table 8-12. The Full Model fits TPHG data with RMSE = 9.926, which is over four times RMSE_{PE} = 2.125. Hence, the model has a highly statistically significant LOF (p-value =

Table 8-12. Time to Peak Heat Generation: Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

Time to Peak Heat Generation Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Significance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	64.969	2.102	<0.0001	****	
Simulant					
Average	3.315	3.368	0.3389		A
SST Blend	-21.794	3.505	<0.0001	****	B
Hi Al	21.963	3.511	<0.0001	****	C
Hi SO ₄	-3.483	3.099	0.2766		A
NaMol	3.053	1.979	0.1413		
FlyAsh	-8.099	1.972	0.0007	***	
BFS	-7.982	1.899	0.0006	***	
MixRatio	-4.215	1.935	0.0437	*	
Simulant*NaMol					
Average	11.845	4.922	0.0278	*	A
SST Blend	-16.273	5.303	0.0070	**	B
Hi Al	5.070	5.164	0.3399		AB
Hi SO ₄	-0.643	4.977	0.8988		AB
Simulant*FlyAsh					
Average	-0.323	4.308	0.9411		A
SST Blend	-4.199	4.722	0.3862		A
Hi Al	10.671	4.469	0.0288	*(f)	A
Hi SO ₄	-6.148	3.908	0.1340		A
Simulant*MixRatio					
Average	2.883	5.087	0.5783		A
SST Blend	5.677	4.889	0.2616		A
Hi Al	0.691	4.083	0.8676		A
Hi SO ₄	-9.250	4.425	0.0519		A
NaMol*FlyAsh	-2.149	3.672	0.5662		
NaMol*MixRatio	5.853	3.195	0.0845		
FlyAsh*MixRatio	3.173	4.161	0.4562		
Summary Statistics of Model Fit					
RMSE = 9.926		RMSE _{PE} = 2.125	R ² = 0.9226	LOF p-value = 4.38E-05	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					
(f) It is possible that a simulant has a statistically significant effect with p-value < 0.05 but that no simulants have statistically significant differences when assessed with the MCP, which controls the probability of a wrong decision over all possible comparisons.					

4.38E-05). Still, the Full Model accounts for a substantial portion of variability in the TPHG data (after removing the data for Mix 34), as indicated by $R^2 = 0.9226$. Table 8-12 shows that the individual parameter effects for FlyAsh and BFS are statistically significant (p-value < 0.001) as is the effect for MixRatio (p-value < 0.05). For the individual simulant effects, the effects of the SST Blend and Hi Al simulants are statistically significant (p-values < 0.0001). The last column of Table 8-12 shows that the effects of SimAve and SimHiSO4 are not statistically different, but that SimBlend, SimHiAl, and pair of SimAve and SimHiSO4 have statistically different effects. Three two-parameter interaction terms involving simulants have statistically significant effects: SimBlend*NaMol (p-values < 0.01), SimAve*NaMol (p-value < 0.05), and SimHiAl*FlyAsh (p-value < 0.05). The last column of Table 8-12 for the Simulant*NaMol interaction shows that only SimAve and SimBlend have statistically different effects.

Table 8-13 summarizes the results of using the stepwise regression approach described in Section 8.3. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-13. The Stepwise Model for TPHG has $R^2 = 0.8958$ that is smaller than for the Full Model (0.9226), but that may be a result of the latter being inflated per Caution FM2 in Section 8.4. Also, RMSE = 9.498 is much larger than $RMSE_{PE} = 2.125$, such that the Stepwise Model LOF is highly significant (p-value = 8.72E-05). Table 8-13 indicates that six

Table 8-13. Time to Peak Heat Generation: Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

Heat Generation, Time to Peak Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	64.192	1.703	37.68	<0.0001	****
SimBlend	-21.506	3.240	-6.64	<0.0001	****
SimHiAl	24.127	2.974	8.11	<0.0001	****
SimHiSO4	-3.659	2.709	-1.35	0.1889	
NaMol	3.733	1.783	2.09	0.0466	*
FlyAsh	-8.026	1.716	-4.68	0.0001	****
BFS	-7.051	1.672	-4.22	0.0003	***
MixRatio	-5.625	1.667	-3.37	0.0024	**
SimBlend*NaMol	-12.693	2.578	-4.92	<0.0001	****
SimHiSO4*MixRatio	-14.994	4.054	-3.70	0.0011	**
SimAve*MixRatio	-4.519	1.659	-2.72	0.0116	*
SimHiAl*FlyAsh	6.034	2.476	2.44	0.0223	*
Summary Statistics for Model Fit					
RMSE = 9.498		RMSE _{PE} = 2.125		R ² = 0.8958	LOF p-value = 8.72E-05
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

individual parameter effects are statistically significant, including SimBlend, SimHiAl, and FlyAsh (p-values < 0.0001), BFS (p-value < 0.001), MixRatio (p-value < 0.01), and NaMol (p-value < 0.05). The statistically significant two-parameter interaction terms are SimBlend*NaMol (p-value < 0.0001), SimHiSO4*MixRatio (p-value < 0.01), SimAve*MixRatio (p-value < 0.05), and SimHiAl*FlyAsh (p-value < 0.05). Finally, no interactions involving BFS are statistically significant, suggesting that BFS interactions do not appear to affect TPHG.

8.7 Results of Statistical Analyses – Cured Waste Form Properties

The results of the statistical analysis methods described in Sections 8.1 to 8.3 are presented and discussed in Section 8.7.1 for compressive strength and in Section 8.7.2 for EPA Method 1315 leach results.

8.7.1 Compressive Strength

PNNL measured compressive strength on triplicate cylindrical samples made using each of the 26 mixes in the Cast Stone screening test matrix in Table 2-1, as discussed in Section 7.2. The means and SDs of Compressive Strength values over triplicate specimens for each of the 26 Cast Stone mixes are listed in Table D-2 in Appendix D. The means of Compressive Strength over the triplicate specimens vary from 857 to 8034 psi with a mean of 4159 psi. Table 8-5 presents uncertainties in the Compressive Strength values (means over triplicate specimens) as calculated from replicates. The estimated uncertainties using all replicate pairs are SD = 409.4 psi and %RSD = 13.3. The natural logarithm of Compressive Strength was statistically analyzed because the standard deviation seemed to be proportional to the magnitude of values. The balance of this section presents and discusses the results of the statistical analysis methods described in Sections 8.1 to 8.3.

Table 8-14 presents the results of fitting the Full Model to the $\ln(\text{Compressive Strength})$ data and statistically assessing the effects of individual parameters and their two-parameter interactions, as discussed in Section 8.2. Keep in mind that Cautions FM1 and FM2 discussed in Section 8.4 apply to the results in Table 8-14. The Full Model accounts for a large portion of the variability in the $\ln(\text{Compressive Strength})$ data, as indicated by $R^2 = 0.9826$. Also, $\text{RMSE} = 0.1759$ is close to $\text{RMSE}_{\text{PE}} = 0.1342$, such that the model LOF is not statistically significant (p-value = 0.0696). Table 8-14 indicates that individual parameter effects for four parameters are statistically significant, including MixRatio (p-value < 0.001) and NaMol, FlyAsh and BFS (p-values < 0.01). None of the terms involving simulant are statistically significant. The only statistically significant two-parameter interaction term is NaMol*MixRatio (p-value < 0.01).

Table 8-15 summarizes the results of using the stepwise regression approach described in Section 8.3. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-15. The Stepwise Model for $\ln(\text{Compressive Strength})$ has $R^2 = 0.9455$ that is smaller than for the Full Model (0.9826), but that may be a result of the latter being inflated per Cautions FM1 and FM2 in Section 8.4. Also, $\text{RMSE} = 0.1967$ is not enough larger than $\text{RMSE}_{\text{PE}} = 0.1342$, such that the Stepwise Model LOF is nonsignificant (p-value = 0.1404). Table 8-15 indicates that four individual parameter effects are statistically significant, including MixRatio and BFS (p-values < 0.0001) and NaMol and FlyAsh (p-values < 0.01). The statistically significant two-parameter interaction terms are NaMol*MixRatio (p-value < 0.0001), SimBlend*NaMol (p-value < 0.01), and SimHiAl*NaMol (p-value < 0.05). Finally, no interactions involving BFS are statistically significant, suggesting that BFS interactions do not appear to affect Compressive Strength.

Table 8-14. ln(Compressive Strength): Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

In(Compressive Strength) Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Signif-icance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	8.1770	0.0418	<0.0001	****	
Simulant					
Average	0.0192	0.0762	0.8094		A
SST Blend	0.0685	0.0705	0.3690		A
Hi Al	-0.1150	0.0710	0.1563		A
Hi SO ₄	0.0273	0.0762	0.7327		A
NaMol	0.1666	0.0404	0.0062	**	
FlyAsh	-0.1663	0.0396	0.0057	**	
BFS	-0.2483	0.0536	0.0036	**	
MixRatio	0.3381	0.0396	0.0001	***	
Simulant*NaMol					
Average	0.0387	0.1151	0.7482		A
SST Blend	-0.2288	0.1069	0.0760		A
Hi Al	0.1155	0.1025	0.3030		A
Hi SO ₄	0.0746	0.1089	0.5186		A
Simulant*FlyAsh					
Average	-0.0308	0.0856	0.7310		A
SST Blend	-0.0497	0.0913	0.6058		A
Hi Al	-0.0383	0.0943	0.6989		A
Hi SO ₄	0.1188	0.0835	0.2048		A
Simulant*MixRatio					
Average	-0.0558	0.1011	0.6012		A
SST Blend	-0.1586	0.0913	0.1331		A
Hi Al	0.0538	0.0774	0.5129		A
Hi SO ₄	0.1606	0.0835	0.1030		A
NaMol*FlyAsh	-0.0191	0.0703	0.7955		
NaMol*MixRatio	-0.2663	0.0668	0.0072	**	
FlyAsh*MixRatio	-0.0656	0.0893	0.4904		
Summary Statistics of Model Fit					
RMSE = 0.1759		RMSE _{PE} = 0.1342	R ² = 0.9826	LOF p-value = 0.0696	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					

Table 8-15. ln(Compressive Strength): Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

In(Compressive Strength) Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	8.1795	0.0409	199.90	<0.0001	****
SimBlend	0.0201	0.0753	0.27	0.7931	
SimHiAl	-0.1245	0.0708	-1.76	0.0990	
SimHiSO4	0.0776	0.0668	1.16	0.2635	
NaMol	0.1579	0.0417	3.78	0.0018	**
FlyAsh	-0.1640	0.0410	-4.00	0.0012	**
BFS	-0.2827	0.0428	-6.60	<0.0001	****
MixRatio	0.3519	0.0413	8.51	<0.0001	****
NaMol*MixRatio	-0.2394	0.0409	-5.85	<0.0001	****
SimBlend*NaMol	-0.2400	0.0718	-3.34	0.0044	**
SimHiAl*NaMol	0.1569	0.0680	2.31	0.0357	*
Summary Statistics for Model Fit					
RMSE = 0.1967		RMSE _{PE} = 0.1342	R ² = 0.9455	LOF p-value = 0.1404	
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

8.7.2 EPA 1315 Leach Test Results

The primary results from the EPA Method 1315 leach tests are effective diffusion coefficients (EDC) calculated as described in Section 7.5. In addition, leachability index (LI) values were calculated from EDCs using the transformation in Equation (7-2). One of the requirements of OLS model fitting methods used in the statistical analysis methods (see Section 8.1) is that the uncertainties in the property values must be relatively constant for all tests. This requirement is met for LI values but not EDC values. Hence, in this section it is the LI data that are statistically analyzed.

Table D-15 in Appendix D contains the duplicate (i.e., repeat) LI values for Na, NO₃, NO₂, I, Tc, Cr, and U, along with means and SDs of the duplicate values, for each of the 26 Cast Stone mixes in Table 2-1. The duplicate LI values were calculated by applying Equation (7-2) to the duplicate EDCs in Table D-14. Table D-15 lists the means and SDs calculated from duplicate LI values for each mix. The mean LI values are used as the property values for each of the 26 Cast Stone mixes for the balance of the data analyses in this subsection.

Figure 8-3 shows a scatterplot matrix of the LI values for Na, NO₃, NO₂, I, Tc, Cr, and U. Note that several components are strongly correlated, with the strongest correlation being for NO₃ and NO₂. Because of that, only the NO₃ data and not the NO₂ data are statistically analyzed

subsequently. Note that Na and I are also highly correlated with NO_3 and NO_2 . Na and I are also strongly correlated with each other, but not quite to the same degree as the other pairs of components discussed so far.

Also of note in Figure 8-3 are the results for Mix 1 (blue triangles) and Mix 23 (red closed squares). Mix 23 is the bottom-left-most data point in every plot in Figure 8-3, which indicates it has the lowest LI for every component. Equivalently, that means Mix 23 has the highest EDC for every component. Mix 23 has the most free-liquid (i.e., bleed water) of any of the 26 Cast Stone mixes for which it was measured, and is a poor mix that would not be used in reality. For that reason, it was decided to exclude Mix 23 from the statistical data analyses. Mix 1 has the highest LI values for Na, NO_3 , NO_2 , and I, but intermediate values for other components. Hence, it was decided to retain Mix 1 in the data for statistical analyses.

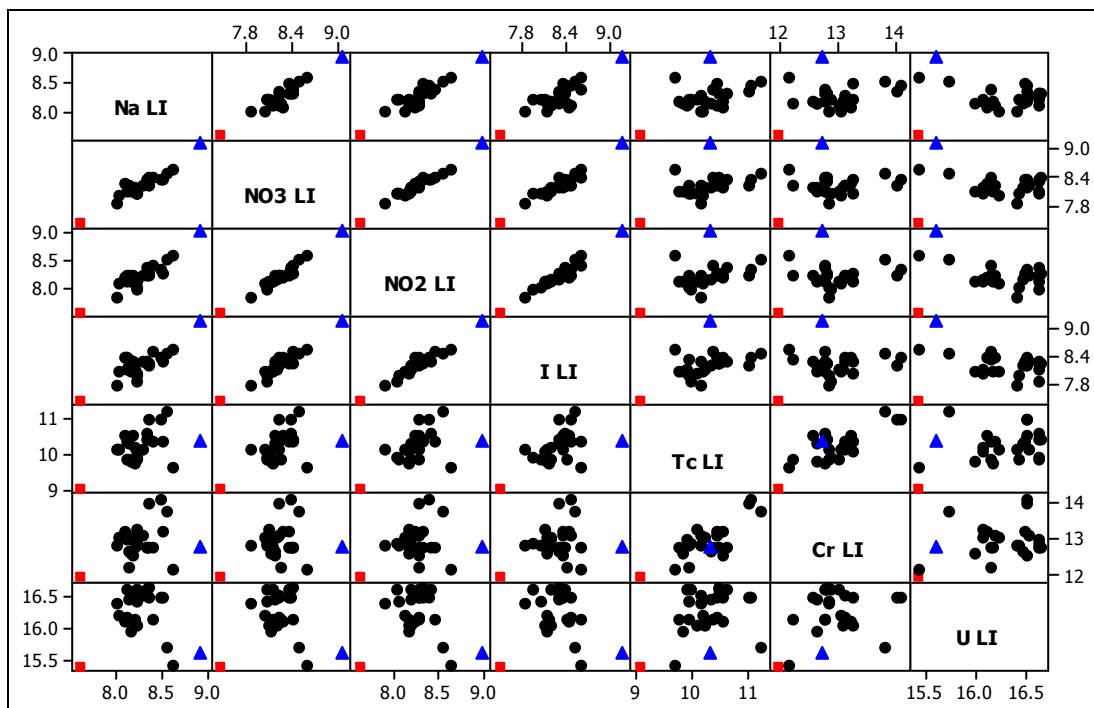


Figure 8-3. Scatterplot Matrix of EPA Method 1315 Leachability Indices for Na, NO_3 , NO_2 , I, Tc, Cr, and U. The red squares are the results for Mix 23, while the blue triangles are the results for Mix 1. (For Information Only)

As mentioned previously, it was decided to only statistically analyze NO_3 and not NO_2 because of how strongly correlated those two components are. Further, because the uranium concentrations in most of the leachates were below analytical detection limits and thus the EDCs were calculated as less-than values (LI as greater-than values), it was decided not to statistically analyze the U LI data. Hence the following subsections contain results of the statistical analyses discussed in Section 8.1 only for the LI data on Na, NO_3 , I, Tc, and Cr.

8.7.2.1 Na Leachability Index from EPA Method 1315

For the 26 Cast Stone mixes in Table 2-1, the means of Na LI values over duplicate (i.e., repeat) tests are given in Table D-15 in Appendix D. The mean values were used for the statistical

analyses reported in this section. Excluding the Na LI value for Mix 23, the remaining Na LI values range from 8.00 to 8.92 with a mean of 8.28. Table 8-7 lists the Na LI values for the five replicate pairs and presents uncertainties in the Na LI values as calculated from the replicates. The pooled (i.e., combined) uncertainty estimates using all replicate pairs are $SD = 0.053$ and $\%RSD = 0.64$, which are small. For one replicate pair (Mixes 3 and 22) the replicate values are identical, which may contribute to these values being underestimates of the uncertainties. The balance of this section presents and discusses the results of the statistical analysis methods described in Sections 8.1 to 8.3.

Table 8-16 presents the results of fitting the Full Model to the Na LI data and statistically assessing the effects of individual parameters and their two-parameter interactions, as discussed in Section 8.2. Keep in mind that the Cautions FM1 and FM2 discussed in Section 8.4 apply to the results in Table 8-16. The Full Model accounts for a large portion of the variability in the Na LI values, as indicated by the $R^2 = 0.9865$. The dropped data point for Mix 23 means that the Full Model fits the data exactly except for replicate variation, so it was not possible to assess the model LOF (see the discussion in Section 8.2.2). (Note that because the LI data for Mix 23 was omitted for all components, the Full Model LOF cannot be assessed for any of the component LIs.) Table 8-16 shows that of the non-simulant individual parameter effects, only FlyAsh is statistically significant ($p\text{-value} < 0.001$). The Average and SST Blend simulants have statistically significant effects ($p\text{-values} < 0.01$). Further, based on the last column of Table 8-16, the Average and SST Blend simulants have statistically different effects, as do the SST Blend and Hi Al simulants. There are six statistically significant two-parameter interaction terms of specific simulants with NaMol, FlyAsh, and MixRatio.

Table 8-17 summarizes the results of using the stepwise regression approach described in Section 8.3 to model Na LI. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that the cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-17. The Stepwise Model for Na LI has $R^2 = 0.8930$ that is smaller than $R^2 = 0.9865$ for the Full Model, but that may be a result of the latter being inflated per Cautions FM1 and FM2 in Section 8.4. The $RMSE = 0.0927$ is somewhat larger than $RMSE_{PE} = 0.0531$, but still the LOF for the Na LI Stepwise Model is nonsignificant ($p\text{-value} = 0.0617$). Table 8-17 indicates that three individual parameter effects are statistically significant, including MixRatio and FlyAsh ($p\text{-values} < 0.01$) and SimBlend ($p\text{-value} < 0.05$). There are four statistically significant two-parameter interaction terms, all involving simulants, including SimHiAl*NaMol and SimAve*NaMol ($p\text{-value} < 0.0001$), and SimHiSO4*MixRatio ($p\text{-value} < 0.01$), and SimHiSO4*FlyAsh ($p\text{-value} < 0.05$). Finally, no interactions involving BFS are statistically significant, suggesting that BFS interactions do not appear to affect Na LI.

Table 8-16. Na Leachability Index: Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

Na Leachability Index Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Significance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	8.3316	0.0166	<0.0001	****	
Simulant					
Average	-0.1141	0.0237	0.0048	**	A
SST Blend	0.1822	0.0365	0.0041	**	B
Hi Al	-0.0478	0.0225	0.0874		A
Hi SO ₄	-0.0203	0.0286	0.5094		AB
NaMol	0.0047	0.0151	0.7687		
FlyAsh	0.1147	0.0155	0.0007	***	
BFS	0.0075	0.0191	0.7114		
MixRatio	0.0122	0.0202	0.5720		
Simulant*NaMol					
Average	-0.0053	0.0397	0.8987		AB
SST Blend	-0.0603	0.0488	0.2717		A
Hi Al	-0.0891	0.0357	0.0549		A
Hi SO ₄	0.1547	0.0329	0.0053	**	B
Simulant*FlyAsh					
Average	-0.1166	0.0269	0.0075	**	A
SST Blend	0.1159	0.0359	0.0232	*	B
Hi Al	-0.0503	0.0291	0.1445		AB
Hi SO ₄	0.0509	0.0269	0.1169		B
Simulant*MixRatio					
Average	0.0153	0.0358	0.6866		AB
SST Blend	-0.2234	0.0619	0.0154	*	A
Hi Al	0.0791	0.0308	0.0500		AB
Hi SO ₄	0.1291	0.0303	0.0080	**	B
NaMol*FlyAsh	0.0800	0.0225	0.0164	*	
NaMol*MixRatio	0.0031	0.0215	0.8903		
FlyAsh*MixRatio	0.0519	0.0274	0.1172		
Summary Statistics of Model Fit					
RMSE = 0.0531		RMSE _{PE} = 0.0531	R ² = 0.9865	LOF p-value = NA ^(f)	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					
(f) The data value for Mix 23 was judged an outlier and not used in the model fit. Hence, it was not possible to statistically assess the model LOF.					

Table 8-17. Na Leachability Index: Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

Na Leachability Index Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	8.2929	0.0200	415.26	<0.0001	****
SimBlend	0.1001	0.0398	2.52	0.0258	*
SimHiAl	-0.0399	0.0341	-1.17	0.2623	
SimHiSO4	0.0207	0.0369	0.56	0.5842	
NaMol	0.0430	0.0207	2.08	0.0581	
FlyAsh	0.0723	0.0197	3.66	0.0029	**
BFS	-0.0263	0.0261	-1.01	0.3317	
MixRatio	0.0790	0.0205	3.86	0.0020	**
SimHiAl*NaMol	-0.2912	0.0502	-5.80	0.0001	****
SimHiSO4*MixRatio	0.1065	0.0284	3.75	0.0024	**
SimHiSO4*FlyAsh	0.0651	0.0282	2.31	0.0377	*
SimAve*NaMol	-0.1369	0.0214	-6.39	<0.0001	****
Summary Statistics for Model Fit					
RMSE = 0.0927		RMSE _{PE} = 0.0531	R ² = 0.8930	LOF p-value = 0.0617	
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

8.7.2.2 NO₃ Leachability Index from EPA Method 1315

For the 26 Cast Stone mixes in Table 2-1, the means of NO₃ LI values over duplicate (i.e., repeat) tests for the 26 Cast Stone mixes are given in Table D-15 in Appendix D. The mean values were used for the statistical analyses reported in this section. Excluding the NO₃ LI value for Mix 23, the remaining NO₃ LI values range from 7.85 to 9.07 with a mean of 8.27. Table 8-7 lists the NO₃ LI values for five replicate pairs and presents uncertainties in the NO₃ LI values as calculated from the replicates. The pooled (i.e., combined) uncertainty estimates using all replicate pairs are SD = 0.107 and %RSD = 1.32, which are small. For one replicate pair (Mixes 15 and 25) the replicate values are identical, which may contribute to these values being underestimates of the uncertainties. The balance of this section presents the results of the statistical analysis methods described in Sections 8.1 to 8.3.

Table 8-18 presents the results of fitting the Full Model to the NO₃ LI data and statistically assessing the effects of individual parameters and their two-parameter interactions, as discussed in Section 8.2. Keep in mind that Cautions FM1 and FM2 discussed in Section 8.4 apply to the results in Table 8-18. The Full Model accounts for a large portion of the variability in the NO₃ LI values, as indicated by the R² = 0.9562. The dropped data point for Mix 23 means that the Full Model fits the data exactly except for replicate variation, so it was not possible to assess the

Table 8-18. NO₃ Leachability Index: Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

NO ₃ Leachability Index Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Significance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	8.3494	0.0334	<0.0001	****	
Simulant					
Average	-0.1529	0.0476	0.0237	*(^f)	A
SST Blend	0.1931	0.0734	0.0464	*(^f)	A
Hi Al	-0.0579	0.0453	0.2575		A
Hi SO ₄	0.0176	0.0575	0.7715		A
NaMol	0.0370	0.0303	0.2769		
FlyAsh	0.0742	0.0312	0.0632		
BFS	0.0030	0.0385	0.9409		
MixRatio	0.0066	0.0405	0.8766		
Simulant*NaMol					
Average	0.0222	0.0797	0.7914		A
SST Blend	-0.1447	0.0982	0.2003		A
Hi Al	-0.0603	0.0718	0.4394		A
Hi SO ₄	0.1827	0.0661	0.0396	*(^f)	A
Simulant*FlyAsh					
Average	-0.1215	0.0541	0.0746		A
SST Blend	0.1565	0.0721	0.0822		A
Hi Al	-0.0690	0.0585	0.2913		A
Hi SO ₄	0.0340	0.0541	0.5571		A
Simulant*MixRatio					
Average	0.0449	0.0719	0.5601		A
SST Blend	-0.3101	0.1245	0.0551		A
Hi Al	0.1249	0.0618	0.0993		A
Hi SO ₄	0.1404	0.0609	0.0693		A
NaMol*FlyAsh	0.1080	0.0453	0.0628		
NaMol*MixRatio	0.0517	0.0433	0.2853		
FlyAsh*MixRatio	0.0652	0.0551	0.2898		
Summary Statistics of Model Fit					
RMSE = 0.1067		RMSE _{PE} = 0.1067	R ² = 0.9562	LOF p-value = NA ^(g)	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					
(f) It is possible that a simulant has a statistically significant effect with p-value < 0.05 but that no simulants have statistically significant differences when assessed with the MCP, which controls the probability of a wrong decision over all possible comparisons.					
(g) The data value for Mix 23 was judged an outlier and not used in the model fit. Hence, it was not possible to statistically assess the model LOF.					

model LOF. Table 8-18 shows that only three terms have statistically significant effects (p-values < 0.05) on NO₃ LI. Two of these terms correspond to the individual effects of the Average and SST Blend simulants (p-values < 0.05). The third term is an interaction of the HiSO₄ simulant with NaMol (p-value < 0.05).

Table 8-19 summarizes the results of using the stepwise regression approach described in Section 8.3 to model NO₃ LI. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that the cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-19. The Stepwise Model for NO₃ LI has $R^2 = 0.6801$ that is smaller than $R^2 = 0.9562$ for the Full Model, but that may be a result of the latter being inflated per Cautions FM1 and FM2 in Section 8.4. The RMSE = 0.1664 is somewhat larger than $RMSE_{PE} = 0.1067$, but still the LOF for the Na LI Stepwise Model is nonsignificant (p-value = 0.1087). Table 8-19 indicates that three individual parameter effects are statistically significant, including SimHiSO₄, MixRatio, and BFS (p-values < 0.05). There are two statistically significant two-parameter interaction terms, SimHiSO₄*NaMol (p-value < 0.001) and SimHiAl*NaMol (p-value < 0.05). Finally, no interactions involving BFS are statistically significant, suggesting that BFS interactions do not appear to affect NO₃ LI.

Table 8-19. NO₃ Leachability Index: Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

NO ₃ Leachability Index Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	8.3096	0.0355	234.10	0.0000	****
SimBlend	0.0417	0.0708	0.59	0.5651	
SimHiAl	-0.0671	0.0611	-1.10	0.2895	
SimHiSO4	0.1565	0.0644	2.43	0.0282	*
NaMol	0.0611	0.0357	1.71	0.1078	
FlyAsh	0.0234	0.0352	0.66	0.5173	
BFS	-0.1035	0.0389	-2.66	0.0178	*
MixRatio	0.0925	0.0361	2.56	0.0216	*
SimHiSO4*NaMol	0.2605	0.0587	4.44	0.0005	***
SimHiAl*NaMol	-0.1333	0.0565	-2.36	0.0323	*
Summary Statistics for Model Fit					
RMSE = 0.1664		RMSE _{PE} = 0.1067		R ² = 0.6801	
				LOF p-value = 0.1087	
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

8.7.2.3 Iodine Leachability Index from EPA Method 1315

For the 26 Cast Stone mixes in Table 2-1, the means of I LI values over duplicate (i.e., repeat) tests for the 26 Cast Stone mixes are given in Table D-15 in Appendix D. The mean values were used for the statistical analyses reported in this section. Excluding the I LI value for Mix 23, the remaining I LI values range from 7.81 to 9.16 with a mean of 8.30. Table 8-7 lists the I LI values for five replicate pairs and presents uncertainties in the I LI values as calculated from the replicates. The pooled (i.e., combined) uncertainty estimates using all replicate pairs are $SD = 0.131$ and $\%RSD = 1.63$, which are small. The balance of this section presents the results of the statistical analysis methods described in Sections 8.1 to 8.3.

Table 8-20 presents the results of fitting the Full Model to the I LI data and statistically assessing the effects of individual parameters and their two-parameter interactions, as discussed in Section 8.2. Keep in mind that Cautions FM1 and FM2 discussed in Section 8.4 apply to the results in Table 8-20. The Full Model accounts for a large portion of the variability in the I LI values, as indicated by the $R^2 = 0.9471$. The dropped data point for Mix 23 means that the Full Model fits the data exactly except for replicate variation, so it was not possible to assess the model LOF. Table 8-20 shows that only two terms have statistically significant effects on I LI. One is an individual parameter effect of the Average simulant, while the other is an interactive effect of the SST Blend simulant with MixRatio (p -values < 0.05).

Table 8-21 summarizes the results of using the stepwise regression approach described in Section 8.3 to model I LI. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that the cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-21. The Stepwise Model for I LI has $R^2 = 0.6812$ with $RMSE = 0.1798$ that is not much larger than $RMSE_{PE} = 0.1311$. Hence, the LOF for the I LI Stepwise Model is nonsignificant (p -value = 0.1869). Table 8-21 indicates that four individual parameter effects are statistically significant, including MixRatio (p -value < 0.01) as well as SimHiSO₄, NaMol, and BFS (p -values < 0.05). There is only one statistically significant two-parameter interaction term, SimHiSO₄*NaMol (p -value < 0.01). Finally, no interactions involving BFS are statistically significant, suggesting that BFS interactions do not appear to affect I LI.

Table 8-20. I Leachability Index: Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

I Leachability Index Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Significance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	8.4041	0.0410	<0.0001	****	
Simulant					
Average	-0.1666	0.0585	0.0359	*(f)	A
SST Blend	0.2297	0.0901	0.0513		A
Hi Al	-0.1103	0.0556	0.1042		A
Hi SO ₄	0.0472	0.0706	0.5335		A
NaMol	0.0747	0.0373	0.1013		
FlyAsh	0.0478	0.0383	0.2674		
BFS	0.0075	0.0473	0.8801		
MixRatio	0.0141	0.0498	0.7889		
Simulant*NaMol					
Average	0.0472	0.0979	0.6503		A
SST Blend	-0.1978	0.1205	0.1617		A
Hi Al	-0.0166	0.0881	0.8583		A
Hi SO ₄	0.1672	0.0811	0.0944		A
Simulant*FlyAsh					
Average	-0.1497	0.0664	0.0739		A
SST Blend	0.1928	0.0886	0.0815		A
Hi Al	-0.0634	0.0718	0.4177		A
Hi SO ₄	0.0203	0.0664	0.7720		A
Simulant*MixRatio					
Average	0.0909	0.0883	0.3505		A
SST Blend	-0.4078	0.1529	0.0445	*(f)	A
Hi Al	0.1647	0.0759	0.0822		A
Hi SO ₄	0.1522	0.0748	0.0975		A
NaMol*FlyAsh	0.1075	0.0556	0.1110		
NaMol*MixRatio	0.0481	0.0531	0.4067		
FlyAsh*MixRatio	0.0694	0.0677	0.3526		
Summary Statistics of Model Fit					
RMSE = 0.1311		RMSE _{PE} = 0.1311	R ² = 0.9471	LOF p-value = NA ^(g)	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					
(f) It is possible that a simulant has a statistically significant effect with p-value < 0.05 but that no simulants have statistically significant differences when assessed with the MCP, which controls the probability of a wrong decision over all possible comparisons.					
(g) The data value for Mix 23 was judged an outlier and not used in the model fit. Hence, it was not possible to statistically assess the model LOF.					

Table 8-21. I Leachability Index: Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

I Leachability Index Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	8.3393	0.0380	219.31	<0.0001	****
SimBlend	0.0483	0.0762	0.63	0.5356	
SimHiAl	-0.1009	0.0659	-1.53	0.1451	
SimHiSO4	0.1818	0.0682	2.67	0.0169	*
NaMol	0.1122	0.0386	2.91	0.0102	*
FlyAsh	-0.0041	0.0374	-0.11	0.9143	
BFS	-0.0986	0.0417	-2.36	0.0312	*
MixRatio	0.1200	0.0388	3.09	0.0070	**
SimHiSO4*NaMol	0.1818	0.0545	3.34	0.0042	**
Summary Statistics for Model Fit					
RMSE = 0.1798		RMSE _{PE} = 0.1311	R ² = 0.6812	LOF p-value = 0.1869	
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

8.7.2.4 Tc Leachability Index from EPA Method 1315

For the 26 Cast Stone mixes in Table 2-1, the means of Tc LI values over duplicate (i.e., repeat) tests for the 26 Cast Stone mixes are given in Table D-15 in Appendix D. The mean values were used for the statistical analyses reported in this section. Excluding the Tc LI value for Mix 23, the remaining Tc LI values range from 9.66 to 11.25 with a mean of 10.30. Table 8-7 lists the Tc LI values for five replicate pairs and presents uncertainties in the Tc LI values as calculated from the replicates. The pooled (i.e., combined) uncertainty estimates using all replicate pairs are SD = 0.131 and %RSD = 1.28, which are small. The balance of this section presents and discusses the results of the statistical analysis methods described in Sections 8.1 to 8.3.

Table 8-22 presents the results of fitting the Full Model to the Tc LI data and statistically assessing the effects of individual parameters and their two-parameter interactions, as discussed in Section 8.2. Keep in mind that Cautions FM1 and FM2 discussed in Section 8.4 apply to the results in Table 8-22. The Full Model accounts for a large portion of the variability in the Tc LI values, as indicated by the R² = 0.9784. The dropped data point for Mix 23 means that the Full Model fits the data exactly except for replicate variation, so it was not possible to assess the model LOF. Table 8-22 shows that the only significant individual parameter effect is NaMol (p-value < 0.01). The Simulant*NaMol, Simulant*FlyAsh, and Simulant*MixRatio groups of interaction terms each have one, three, and three statistically significant terms, respectively. The last column of Table 8-22 indicates which simulants are involved in the interactions. For the

Table 8-22. Tc Leachability Index: Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

Tc Leachability Index Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Signif- icance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	10.3153	0.0409	<0.0001	****	
Simulant					
Average	-0.1118	0.0584	0.1137		A
SST Blend	0.2034	0.0900	0.0733		A
Hi Al	-0.0706	0.0556	0.2600		A
Hi SO ₄	-0.0211	0.0705	0.7772		A
NaMol	-0.1942	0.0372	0.0034	**	
FlyAsh	0.0267	0.0383	0.5167		
BFS	-0.1155	0.0472	0.0582		
MixRatio	-0.0594	0.0497	0.2855		
Simulant*NaMol					
Average	0.1868	0.0978	0.1144		AB
SST Blend	-0.3212	0.1204	0.0445	*	A
Hi Al	0.0106	0.0880	0.9092		AB
Hi SO ₄	0.1238	0.0811	0.1872		B
Simulant*FlyAsh					
Average	-0.2638	0.0663	0.0106	*	A
SST Blend	0.4657	0.0885	0.0033	**	B
Hi Al	-0.2451	0.0718	0.0189	*	A
Hi SO ₄	0.0432	0.0663	0.5437		A
Simulant*MixRatio					
Average	-0.0696	0.0882	0.4662		AB
SST Blend	-0.5133	0.1527	0.0201	*	A
Hi Al	0.2142	0.0758	0.0369	*	AB
Hi SO ₄	0.3687	0.0747	0.0043	**	B
NaMol*FlyAsh	-0.0955	0.0555	0.1462		
NaMol*MixRatio	-0.0686	0.0531	0.2526		
FlyAsh*MixRatio	0.0166	0.0676	0.8156		
Summary Statistics of Model Fit					
RMSE = 0.1309		RMSE _{PE} = 0.1309	R ² = 0.9784	LOF p-value = NA ^(f)	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					
(f) The data value for Mix 23 was judged an outlier and not used in the model fit. Hence, it was not possible to statistically assess the model LOF.					

Simulant*NaMol interaction, the SST Blend and HiSO4 simulants have statistically different effects depending on the level of NaMol. For the Simulant*FlyAsh interaction, the SST Blend simulant has statistically different effects depending on the level of NaMol. For the Simulant*MixRatio interaction, the SST Blend and HiSO4 simulants have statistically different effects depending on the level of MixRatio. Note that SST Blend interacts with NaMol, FlyAsh, and MixRatio.

Table 8-23 summarizes the results of using the stepwise regression approach described in Section 8.3 to model Tc LI. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-23. The Stepwise Model for Tc LI has $R^2 = 0.9358$ that is smaller than $R^2 = 0.9784$ for the Full Model, but that may be a result of the latter being inflated per Cautions FM1 and FM2 in Section 8.4. The RMSE = 0.1351 is slightly larger than $RMSE_{PE} = 0.1309$, such that the LOF for the Tc LI Stepwise Model is highly nonsignificant (p-value = 0.4844). Table 8-23 shows statistically significant individual effects of MixRatio and BFS (p-values < 0.0001), SimHiSO4 (p-value < 0.001), FlyAsh p-value < 0.01) and SimBlend and NaMol (p-values < 0.05). As discussed in Section 8.3, the significant effects of SimHiSO4 and SimBlend actually correspond to the Hi SO₄ and SST Blend simulants having different effects than the Average simulant on Tc LI. There are three statistically significant interaction terms, two of which include BFS interacting with simulants (p-values < 0.0001). Hence, it appears BFS has a statistically significant effect individually and has statistically significant interaction effects involving simulants.

Table 8-23. Tc Leachability Index: Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

Tc Leachability Index Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	10.2574	0.0294	349.32	<0.0001	****
SimBlend	-0.1879	0.0662	-2.84	0.0132	*
SimHiAl	0.0373	0.0506	0.74	0.4733	
SimHiSO4	0.2482	0.0480	5.17	0.0001	***
NaMol	-0.0751	0.0318	-2.36	0.0333	*
FlyAsh	-0.0907	0.0293	-3.10	0.0079	**
BFS	-0.3306	0.0303	-10.91	<0.0001	****
MixRatio	0.2092	0.0312	6.69	<0.0001	****
SimHiAl*BFS	0.2985	0.0490	6.10	<0.0001	****
SimBlend*BFS	-0.4183	0.0710	-5.89	<0.0001	****
SimBlend*MixRatio	0.2018	0.0674	2.99	0.0097	**
Summary Statistics for Model Fit					
RMSE = 0.1351		RMSE _{PE} = 0.1309		R ² = 0.9358	LOF p-value = 0.4844
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

8.7.2.5 Cr Leachability Index from EPA Method 1315

For the 26 Cast Stone mixes in Table 2-1, the means of Cr LI values over duplicate (i.e., repeat) tests for the 26 Cast Stone mixes are given in Table D-15 in Appendix D. The mean values were used for the statistical analyses reported in this section. Excluding the Cr LI value for Mix 23, the remaining Cr LI values range from 12.15 to 14.11 with a mean of 12.98. Table 8-7 lists the Cr LI values for five replicate pairs and presents uncertainties in the Cr LI values as calculated from the replicates. The pooled (i.e., combined) uncertainty estimates using all replicate pairs are $SD = 0.088$ and $\%RSD = 0.67$, which are small. The balance of this section presents and discusses the results of the statistical analysis methods described in Sections 8.1 to 8.3.

Table 8-24 presents the results of fitting the Full Model to the Cr LI data and statistically assessing the effects of individual parameters and their two-parameter interactions, as discussed in Section 8.2. Keep in mind that Cautions FM1 and FM2 discussed in Section 8.4 apply to the results in Table 8-24. The Full Model accounts for a large portion of the variability in the Cr LI values, as indicated by the $R^2 = 0.9926$. The dropped data point for Mix 23 means that the Full Model fits the data exactly except for replicate variation, so it was not possible to assess the model LOF. Table 8-24 shows that three individual parameter effects are statistically significant, namely NaMol, FlyAsh, and BFS (p-values < 0.01). Interactions not involving Simulant that are statistically significant include NaMol*MixRatio (p-value < 0.001), NaMol*FlyAsh (p-value < 0.01), and FlyAsh*MixRatio (p-value < 0.05). The significant interactions involving Simulant include Simulant*NaMol, Simulant*FlyAsh, and Simulant*MixRatio. For Simulant*NaMol, only $HiSO_4$ has a significant effect, but no simulants have statistically different effects based on Tukey's HSD MCP results (see Section 8.2.5) shown in the last column of Table 8-24. For the interaction Simulant*FlyAsh, the simulants SST Blend and $HiSO_4$ interact differently with FlyAsh than the other two simulants. For the interaction, Simulant*MixRatio, the Average simulant interacts differently with MixRatio than the other three simulants, while the $HiSO_4$ simulant interacts differently with MixRatio than the other three simulants.

Table 8-25 summarizes the results of using the stepwise regression approach described in Section 8.3 to model Cr LI. That approach allows all two-parameter interactions to enter the model, including interactions involving BFS. Keep in mind that cautions SM1 and SM2 discussed in Section 8.4 apply to the results in Table 8-25. The Stepwise Model for Cr LI has $R^2 = 0.9834$ that is smaller than $R^2 = 0.9926$ for the Full Model, but that may be a result of the latter being inflated per Cautions FM1 and FM2 in Section 8.4. The $RMSE = 0.0935$ is similar in magnitude to $RMSE_{PE} = 0.0885$, such that the LOF for the Cr LI Stepwise Model is highly nonsignificant (p-value = 0.4127). Table 8-25 shows statistically significant individual effects of MixRatio and BFS (p-values < 0.0001), NaMol and FlyAsh (p-values < 0.001), and SimBlend and SimHiAl (p-values < 0.01). Based on the discussion in Section 8.3, the SimBlend and SimHiAl effects actually correspond to significant differences in the effects on Cr LI of the SST Blend and Hi Al simulants compared to the Average simulant. There are seven statistically significant interaction terms (see Table 8-25), three of which include BFS (interacting with MixRatio, SimHiAl, and SimBlend). Hence, it appears BFS has a statistically significant individual effect and has statistically significant interaction effects.

Table 8-24. Cr Leachability Index: Summary of Fitting the Full Model and Statistically Assessing the Effects of the Individual Test Parameters and Two-Parameter Interactions

Cr Leachability Index Model Term ^(a)	Effect Estimate ^(a)	SD(Effect Estimate) ^(b)	p-value ^(c)	Signif-icance ^(d)	Tukey's HSD MCP Results ^(e)
Intercept	12.8516	0.0277	<0.0001	****	
Simulant					
Average	0.0607	0.0395	0.1849		A
SST Blend	-0.0791	0.0609	0.2507		A
Hi Al	0.0869	0.0376	0.0686		A
Hi SO ₄	-0.0686	0.0477	0.2101		A
NaMol	-0.1148	0.0252	0.0060	**	
FlyAsh	0.1198	0.0259	0.0057	**	
BFS	-0.1530	0.0319	0.0049	**	
MixRatio	0.0456	0.0336	0.2334		
Simulant*NaMol					
Average	0.0299	0.0662	0.6699		A
SST Blend	-0.0231	0.0814	0.7884		A
Hi Al	-0.1513	0.0595	0.0518		A
Hi SO ₄	0.1444	0.0548	0.0463	*(^f)	A
Simulant*FlyAsh					
Average	-0.3132	0.0449	0.0009	***	A
SST Blend	0.2588	0.0598	0.0075	**	B
Hi Al	-0.3394	0.0485	0.0009	***	A
Hi SO ₄	0.3938	0.0449	0.0003	***	B
Simulant*MixRatio					
Average	-0.2008	0.0597	0.0200	*	A
SST Blend	-0.0421	0.1033	0.7007		AB
Hi Al	0.0079	0.0513	0.8831		AB
Hi SO ₄	0.2349	0.0505	0.0056	**	B
NaMol*FlyAsh	-0.2380	0.0376	0.0014	**	
NaMol*MixRatio	-0.3011	0.0359	0.0004	***	
FlyAsh*MixRatio	-0.1746	0.0457	0.0124	*	
Summary Statistics of Model Fit					
RMSE = 0.0885		RMSE _{PE} = 0.0885	R ² = 0.9926	LOF p-value = NA ^(g)	
(a) See Section 8.2.1 for discussion of the model terms and effect estimates.					
(b) The standard deviation of the effect estimate.					
(c) The probability of incorrectly concluding that the effect estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate effect estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					
(e) Tukey's HSD MCP is discussed in Section 8.2.5. For the Simulant parameter by itself, estimates of the simulant effects having the same letter (e.g., A, B, C) are not statistically different at the 0.05 level of significance (i.e., > 95% confidence) over all comparisons. For Simulant*Parameter interactions, simulants with the same letter (e.g., A, B, C) do not have statistically significant interactions with Parameter at the 0.05 level of significance (e.g., > 95% confidence) over all comparisons.					
(f) It is possible that a simulant has a statistically significant effect with p-value < 0.05 but that no simulants have statistically significant differences when assessed with the MCP, which controls the probability of a wrong decision over all possible comparisons.					
(g) The data value for Mix 23 was judged an outlier and not used in the model fit. Hence, it was not possible to statistically assess the model LOF.					

Table 8-25. Cr Leachability Index: Results of Using Stepwise Regression to Select Statistically Significant Individual Parameter and Two-Parameter Interaction Terms for the Stepwise Model^(a)

Cr Leachability Index Model Term ^(a)	Coefficient Estimate ^(a)	SD(Coeff. Estimate) ^(b)	t value	p-value Pr(> t) ^(c)	Statistical Significance ^(d)
Intercept	12.8967	0.0227	566.95	<0.0001	****
SimBlend	-0.1548	0.0408	-3.79	0.0035	**
SimHiAl	0.1554	0.0361	4.31	0.0015	**
SimHiSO4	-0.0267	0.0362	-0.74	0.4781	
NaMol	-0.1168	0.0217	-5.39	0.0003	***
FlyAsh	0.1179	0.0193	6.12	0.0001	***
BFS	-0.2472	0.0231	-10.71	<0.0001	****
MixRatio	0.1562	0.0218	7.16	<0.0001	****
BFS*MixRatio	-0.1037	0.0260	-3.99	0.0026	**
NaMol*MixRatio	-0.1982	0.0269	-7.37	<0.0001	****
SimHiSO4*FlyAsh	0.3047	0.0396	7.70	<0.0001	****
SimHiAl*FlyAsh	-0.2179	0.0351	-6.21	0.0001	***
SimHiAl*BFS	0.2220	0.0415	5.35	0.0003	***
SimBlend*BFS	-0.1655	0.0464	-3.56	0.0052	**
FlyAsh*MixRatio	-0.0595	0.0219	-2.72	0.0215	*
Summary Statistics for Model Fit					
RMSE = 0.0935		RMSE _{PE} = 0.0885		R ² = 0.9834	
				LOF p-value = 0.4127	
(a) See Section 8.3 for discussion of the model terms and why there are no terms involving the Average simulant.					
(b) The standard deviation of the coefficient estimate.					
(c) The probability of incorrectly concluding that the coefficient estimate is different from zero. Low values (e.g., < 0.05 and especially < 0.01) indicate coefficient estimates that are statistically different from zero.					
(d) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001. Also, note that the individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

8.8 Final Assessment

Section 8.8.1 assesses how well the Full and Stepwise Models fit the screening study data for the nine Cast Stone properties that were statistically analyzed. Section 8.8.2 discusses the statistically significant terms in the Full and Stepwise Models for each property.

8.8.1 Assessing the Fits of the Full and Stepwise Models

Table 8-26 summarizes the fits of the Full Models and Stepwise Models to the screening study data for the nine Cast Stone properties that were statistically analyzed. For comparison, R² statistics are also included for the Main Effects Model, which contains terms only for individual effects of the parameters (i.e., no interaction terms). Generally the Full Models and Stepwise Models have noticeably larger R² values than the Main Effects Model for the Cast Stone properties, which indicates that there are statistically significant interactions between parameters. The results in Table 8-26 are discussed in turn below for the Full Models, then the Stepwise Models.

Table 8-26. Summary Statistics from Full Model and Stepwise Model Fits to Cast Stone Screening Study Data

Property	RMSE Pure Error	RMSE Full Model	RMSE Stepwise Model	# Terms in Stepwise Model ^(a)	# Data for Fitting Models	R ² Main Effects Model ^(b)	R ² Full Model ^(c)	R ² Stepwise Model	p-value LOF Full Model ^(d)	p-value LOF Stepwise Model ^(d)
Plastic Viscosity	0.063	0.162	0.183	9	37 ^(e)	0.9366	0.9799	0.9572	0.0024*	0.0016*
Heat Generation at 300 Hours	10.478	21.716	17.930	10	38	0.8468	0.9298	0.9256	0.0107*	0.0452*
Time to Peak Heat Generation	2.125	9.926	9.498	12	37 ^(f)	0.4458	0.9226	0.8958	<0.0001*	<0.0001*
Compressive Strength	0.134	0.176	0.197	11	26	0.7508	0.9826	0.9455	0.0696	0.1404
Na Leachability Index	0.053	0.053	0.093	12	25 ^(g)	0.3403	0.9865	0.8930	NA ^(h)	0.0617
NO ₃ Leachability Index	0.107	0.107	0.166	10	25 ^(g)	0.2592	0.9562	0.6801	NA	0.1087
I Leachability Index	0.131	0.131	0.180	9	25 ^(g)	0.4591	0.9471	0.6812	NA	0.1869
Tc Leachability Index	0.131	0.131	0.135	11	25 ^(g)	0.6972	0.9784	0.9358	NA	0.4844
Cr Leachability Index	0.089	0.089	0.094	15	25 ^(g)	0.6321	0.9926	0.9834	NA	0.4127
<p>(a) The number of terms includes the intercept and all seven individual parameter effects, regardless of whether the latter are statistically significant or not.</p> <p>(b) The “main effects” model contains only terms for individual effects of the parameters, that is, no interaction terms.</p> <p>(c) As discussed in Section 8.4, these R² values may be inflated because of nonsignificant terms being in the Full Model.</p> <p>(d) * denotes the model LOF is statistically significant with p-value < 0.05.</p> <p>(e) Models were fit to 37 data points because the paste for Mix 1 was too thick to measure plastic viscosity.</p> <p>(f) Models were fit to 37 data points because the value for Mix 34 was an outlier that substantially changed the statistical results.</p> <p>(g) Models were fit to 25 data points for these properties (because the data for Mix 23 were judged to be outliers and were not used in the model fits).</p> <p>(h) NA = not applicable. The model LOF cannot be assessed because the Full Model contains the same number of free parameter effects as distinct data points (see Section 3.6.2).</p>										

Table 8-26 shows that the Full Model produces very high R^2 values for the nine Cast Stone properties. However, these values tend to be inflated because of containing many nonsignificant terms (see Cautions FM1 and FM2 in Section 8.4). For Compressive Strength in Table 8-26, the Full Model contains 20 independent parameters, but there are only 21 distinct mixes with data used to fit the model. For the component LIs (Na, NO₃, I, Tc, and Cr) in Table 8-26, there are only 20 distinct mixes with data to fit the model because of eliminating an outlier. Hence, there is only one DOF to assess LOF of the Full Model for Compressive Strength (which is not statistically significant) and no DOF to assess LOF of the Full Model for the LI components. There are sufficient DOF to assess LOF of the Full Model for ln(Plastic Viscosity), HG300, and TPHG. There are also sufficient DOF to assess the LOF of the Stepwise Models for all Cast Stone properties.

The results in Table 8-26 show that Stepwise Models have R^2 values ranging from 0.6801 to 0.9834 for the LI components, and from 0.8958 to 0.9572 for the other four properties. Table 8-26 also shows the Stepwise Models for Compressive Strength and the component LIs do not have statistically significant LOFs and hence are adequate approximations of the relationships between these properties and the test parameters. However, the results also show that the Stepwise Models (as well as the Full Models) for Plastic Viscosity, HG300, and TPHG have statistically significant LOFs. Ultimately though, the Full and Stepwise Models fit the data well enough to assess which components have significant effects (individual and interaction) on the Cast Stone properties.

8.8.2 Assessing the Statistically Significant Terms in the Full and Stepwise Models

Table 8-27 and Table 8-28 provide an overview of the Full Model terms that are statistically significant for each Cast Stone property that was modeled. The p-values presented in these two tables originate from the Full Model fit summaries presented previously in Table 8-8 through Table 8-24. The p-values < 0.05 are marked with an asterisk (*), which means the corresponding terms are statistically significant with > 95% confidence. While terms marked with an * are likely significant, as discussed in Section 8.4 terms not so marked may still be statistically significant if nonsignificant model terms were removed. That is one of the reasons the Stepwise Models were fit to the Cast Stone property data. All individual parameter effects and all except two two-parameter interaction effects are statistically significant in the Full Model for at least one property in Table 8-27 and Table 8-28. One exception is the individual effect of HiSO₄ in Table 8-27, but that may not be significant as a result of significant interactions involving HiSO₄. The other exception is the SimHiAl*NaMol interaction, although it is close to being significant for Na LI and Cr LI.

Discussion of Caution SM2 in Section 8.4 noted that one way to protect against the tendency of stepwise regression to include too many terms in a model is to use a more stringent threshold for assessing the statistical significance of terms. Suppose a more stringent threshold (than the $p < 0.05$ used in Table 8-27 and Table 8-28) for statistical significance was adopted for both the Full and Stepwise Models. With a p-value < 0.01 threshold, the SimAve*NaMol, SimAve*MixRatio, SimBlend*MixRatio, and FlyAsh*MixRatio terms in the Full Model would not be statistically significant for any property (see those columns in Table 8-27 and Table 8-28).

Table 8-27. Simulant Terms in the Full Model Identified as Statistically Significant by the Tukey Multiple Comparison Procedure. Table contains p-values for statistically testing the hypothesis that the “Term = 0”.

Property ^(a)	Simulant				Simulant*NaMol				Simulant*FlyAsh				Simulant*MixRatio			
	Ave.	Blend	HiAl	HiSO4	Ave.	Blend	HiAl	HiSO4	Ave.	Blend	HiAl	HiSO4	Ave.	Blend	HiAl	HiSO4
PV	0.779	0.977	0.164	0.384	0.545	0.082	0.767	0.044*	0.399	0.384	0.058	0.086	0.548	0.404	0.734	0.111
HG300	0.417	0.002*	0.030*	0.790	0.229	0.092	0.634	0.905	0.789	0.600	0.955	0.422	0.778	0.243	0.820	0.448
TPHG	0.339	<0.001*	<0.001*	0.277	0.028*	0.007*	0.340	0.899	0.941	0.386	0.029*	0.134	0.578	0.262	0.868	0.052
CS	0.809	0.369	0.156	0.733	0.748	0.076	0.303	0.519	0.731	0.606	0.699	0.205	0.601	0.133	0.513	0.103
Na LI	0.005*	0.004*	0.087	0.509	0.899	0.272	0.055	0.005*	0.007*	0.023*	0.144	0.117	0.687	0.015*	0.050*	0.008*
NO ₃ LI	0.024*	0.046*	0.257	0.772	0.791	0.200	0.439	0.040*	0.075	0.082	0.291	0.557	0.560	0.055	0.099	0.069
I LI	0.036*	0.051	0.104	0.534	0.650	0.162	0.858	0.094	0.074	0.082	0.418	0.772	0.350	0.044*	0.082	0.098
Tc LI	0.114	0.073	0.260	0.777	0.114	0.044*	0.909	0.187	0.011*	0.003*	0.019*	0.544	0.466	0.020*	0.037*	0.004*
Cr LI	0.185	0.251	0.069	0.210	0.670	0.788	0.052	0.046*	0.001*	0.008*	0.001*	<0.001*	0.020*	0.701	0.883	0.006*

(a) PV = Plastic Viscosity, HG300 = Heat Generation at 300 hr, TPHG = Time to Peak Heat Generation, CS = Compressive Strength, LI = Leachability Index.
 (b) An asterisk (*) denotes the term is statistically significant with p-value < 0.05 (95% confidence level).

Table 8-28. Full Model Terms not Involving Simulant Identified as Statistically Significant.
Table contains p-values for statistically testing the hypothesis that the “Term = 0”.

Property	NaMol	FlyAsh	BFS	MixRatio	NaMol * FlyAsh	NaMol* MixRatio	FlyAsh* MixRatio
Plastic Viscosity	0.9461	<0.0001*	0.0291*	<0.0001*	0.7206	0.8988	0.3075
Heat Generation at 300 hr	0.2993	<0.0001*	<0.0001*	<0.0001*	0.8784	0.0608	0.3344
Time to Peak Heat Generation	0.1413	0.0007*	0.0006*	0.0437*	0.5662	0.0845	0.4562
Compressive Strength	0.0062*	0.0057*	0.0036*	0.0001*	0.7955	0.0072*	0.4904
Na Leachability Index	0.7687	0.0007*	0.7114	0.5720	0.0164*	0.8903	0.1172
NO ₃ Leachability Index	0.2769	0.0632	0.9409	0.8766	0.0628	0.2853	0.2898
I Leachability Index	0.1013	0.2674	0.8801	0.7889	0.1110	0.4067	0.3526
Tc Leachability Index	0.0034*	0.5167	0.0582	0.2855	0.1462	0.2526	0.8156
Cr Leachability Index	0.0060*	0.0057*	0.0049*	0.2334	0.0014*	0.0004*	0.0124*
Note: An asterisk (*) denotes the term is statistically significant with p-value < 0.05 (95% confidence level).							

Table 8-29 and Table 8-30 summarize the statistically significant terms in the Stepwise Model for each of the nine Cast Stone properties that were statistically analyzed. The results in each table are discussed in turn.

The following observations about individual parameter effects are based on Table 8-29: 1) FlyAsh and MixRatio have statistically significant effects for all four properties, 2) BFS has a statistically significant effect for HG300, TPHG, and Compressive Strength, and 3) SimBlend and SimHiAl have statistically significant effects for HG300 and TPHG. The individual effects of SimBlend and SimHiAl may not be statistically significant for Compressive Strength because of interaction terms involving those two simulants. Table 8-29 also shows that a total of eight different interaction effects are statistically significant for one or more of the four properties. As explained in Caution SM1 of Section 8.4, the interactions in the Stepwise Models may be present because the two parameters interact, or because of correlations among the possible individual and interaction terms. Still, note that each of the four simulants and each of the remaining parameters (NaMol, FlyAsh, BFS, and MixRatio) is present in at least one interaction. MixRatio appears in four interactions, NaMol and FlyAsh each appear in three interactions, and BFS only appears in one interaction (HG300). As for the simulants, SimHiAl appears in two interactions, while SimAve, SimBlend, and SimHiSO4 each appear in one interaction.

Table 8-29. Summary of Statistically Significant Terms in Stepwise Models for the Cast Stone Physical Properties

Stepwise Model Term ^(a)	ln(Plastic Viscosity)	Heat Generation at 300 hr	Time to Peak Heat Generation	ln(Compressive Strength)
SimBlend		**	****	(c)
SimHiAl		**	****	(c)
SimHiSO4			(c)	
NaMol		(c)	*	**
FlyAsh	**** ^(b)	****	****	**
BFS		****	***	****
MixRatio	****	****	**	****
SimBlend*NaMol		(c)	****	**
SimHiAl*NaMol				*
SimHiAl*FlyAsh			*	
SimAve*MixRatio			*	
SimHiSO4*MixRatio			**	
NaMol*MixRatio		*		****
FlyAsh*MixRatio	***			
FlyAsh*BFS		**		
R ²	0.9572	0.9256	0.8958	0.9455
RMSE	0.1835	17.930	9.498	0.1967
RMSE _{PE}	0.0627	10.478	2.125	0.1342
Model LOF p-value	0.0016	0.0452	8.72E-05	0.1404
(a) As discussed in Section 8.3, the individual term for the Average simulant (SimAve) was not possible in the Stepwise Models.				
(b) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001.				
(c) The individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.				

The following observations about individual parameter effects on the component LIs are based on Table 8-30: 1) MixRatio has a statistically significant effect for all five component LIs, 2) BFS has a statistically significant effect for all except Na LI, 3) FlyAsh has a statistically significant effect for Na, Tc, and Cr LIs, 4) NaMol has statistically significant effects on I, Tc, and Cr LIs, 5) SimHiSO4 has statistically significant effects on NO₃, I, and Tc LIs and 6) SimBlend has statistically significant effects on Na, Tc, and Cr LIs. The (c) footnote in Table 8-30 marks other individual parameter effects that may not be statistically significant for component LIs because of interaction terms involving those parameters. Table 8-30 also shows that a total of 12 different interaction effects are statistically significant for one or more of the component LIs. As explained in Caution SM1 of Section 8.4, the interactions in the Stepwise Models may be present because the two parameters interact, or because of correlations among the possible individual and interaction terms. Still, note that each of the four simulants and each of the remaining parameters (NaMol, FlyAsh, BFS, and MixRatio) is present in at least one interaction. MixRatio appears in five interactions, NaMol appears in four interactions, while FlyAsh and BFS each appear in three interactions. As for the simulants, SimHiAl and SimHiSO4 each appear in three interactions, SimBlend appears in two interactions, and SimAve appears in only one interaction.

Table 8-30. Summary of Statistically Significant Terms in Stepwise Models for the Leachability Indices (LI)

Stepwise Model Term^(a)	Na LI	NO₃ LI	I LI	Tc LI	Cr LI
SimBlend	*			*	**
SimHiAl	(c)			(c)	**
SimHiSO4	(c)	*	*	***	(c)
NaMol	(c)	(c)	*	*	***
FlyAsh	**			**	***
BFS		*	*	****	****
MixRatio	**	*	**	****	****
SimAve*NaMol	****				
SimHiAl*NaMol	****	*			
SimHiSO4*NaMol		***	**		
SimHiAl*FlyAsh					***
SimHiSO4*FlyAsh	*				****
SimBlend*MixRatio				**	
SimHiSO4*MixRatio	**				
NaMol*MixRatio					****
FlyAsh*MixRatio					*
SimBlend*BFS				****	**
SimHiAl*BFS				****	***
BFS*MixRatio					**
R ²	0.8930	0.6801	0.6812	0.9358	0.9834
RMSE	0.0927	0.1664	0.1798	0.1351	0.0935
RMSE _{PE}	0.0531	0.1067	0.1311	0.1309	0.0885
Model LOF p-value	0.0617	0.1087	0.1869	0.4844	0.4127
(a) As discussed in Section 8.3, terms involving the Average simulant (SimAve) were excluded from the stepwise models.					
(b) * = p-value < 0.05, ** = p-value < 0.01, *** = p-value < 0.001, and **** = p-value < 0.0001.					
(c) The individual effect of a parameter may not be statistically significant if that parameter is involved in one or more statistically significant interactions.					

The objective of the Cast Stone screening study was to identify which parameters (individually or in interactions) affect which Cast Stone properties. Ideally, some parameters would be identified as not affecting any of the more important properties that were statistically analyzed, so that those parameters could be removed from consideration for future Cast Stone optimization studies. However, as noted in the preceding discussions of Table 8-29 and Table 8-30, every parameter has individual as well as interaction effects for at least one property. This includes BFS, which was believed not to interact with other parameters during the test matrix development (see Appendix A). Even though parameters like BFS and FlyAsh have statistically significant individual and interactive effects, the test results and statistical analyses of those results provide a basis for choosing a source of BFS and a source of FlyAsh to eliminate one or both of those parameters from a future formulation optimization study. Further, the statistical analyses in this section provide a basis for choosing the simulants and ranges of the NaMol and MixRatio parameters for a future formulation optimization study.

9.0 Conclusions

The LAW Cast Stone screening tests were a successful first step in developing and optimizing the Cast Stone waste form for immobilization of Hanford LAW. The screening tests included 26 mixes identified through a statistical experimental design and included four Hanford LAW simulants, two different sources each of fly ash and BFS used as dry blend ingredients, water-to-dry-blend mix ratios of 0.4 and 0.6, and waste concentrations for 5M and 7.8M sodium. An additional 12 Cast Stone mixes were investigated at SRNL. The freshly prepared Cast Stone slurry was characterized with respect to rheology, set time, hardening time, heat generation during curing, fresh density, and free liquids. The final Cast Stone cured specimens were characterized with respect to cured density, compressive strength, porosity, TCLP for meeting land disposal restrictions, and leachability of Na, NO₃, NO₂, iodine, technetium, chromium, and uranium. Statistical analyses of the rheology, heat generation, compressive strength, and leachability were conducted to determine whether individual test parameters and their two-parameter interactions have statistically significant effects on a given property, and to assess the relative magnitudes of these effects.

Plastic viscosity and Bingham Plastic yield stress were measured on the fresh Cast Stone slurry paste. Mix ratio and fly ash source were the primary contributors to the variations observed in the plastic viscosity, with both individual effects and the two-parameter interaction being statistically significant. Mixes with the 0.4 mix ratio were more viscous than the mixes with the higher water content at a mix ratio of 0.6. Plastic viscosities were in the range of 220 to 580 cP for the 0.4 mixes and 54 to 165 for the 0.6 mixes. Mixes prepared with the SE fly ash were more viscous than those prepared with the NW FA. There were no statistically significant differences in the plastic viscosities due to the different waste simulants.

Isothermal calorimetry was used to determine the heat of hydration of the Cast Stone slurries as the mixes cured. Heat generated over 300 hours was in the range of 150 to 434 J/g. Higher heat generation was observed for the 0.6 mix ratios than for the 0.4 mix ratios. The greater heat generated from the 0.6 mix ratios may indicate that there was not enough water in mixes with the lower mix ratio to fully hydrate the dry materials. Statistical analyses indicated that, in addition to mix ratio, the BFS and fly ash sources had statistically significant effects on both heat generation properties that were statistically analyzed (Heat Generation at 300 Hours, and Time to Peak Heat Generation). The time to reach the peak heat generation rate ranged from 16 to 164 hours. This property can affect the processing strategy for a given waste container if there are temperature limits associated with the chosen waste form.

Gel times for the Cast Stone slurries ranged from 1 minute to more than 2 hours. The gel times were typically higher for mixes with the 0.6 mix ratio than mixes with the 0.4 mix ratio and were highest for the 0.6 mix ratio and 7.8M Na simulants. Gel times can be useful in the design and implementation of the waste form to ensure even distribution in the container.

Densities were measured for both the “as prepared” Cast Stone slurry (fresh density) and after curing 28 days (cured density). The fresh densities were highest for mixes with the 0.4 mix ratio and were in the range of 1.87 to 1.96 g/cm³. Fresh densities for mixes with the 0.6 mix ratio were in the range of 1.73 to 1.83 g/cm³. Cured densities were in the range of 1.87 to 1.96 g/cm³ for Cast Stone specimens with the 0.4 mix ratio and 1.73 to 1.80 g/cm³ for specimens with the 0.6 mix ratio.

Residual free liquids were observed in 6 of the 38 mixes one day after preparation and only 3 of the 38 mixes had any residual free liquids after three days. The residual free liquid is an indication of settling that results from a longer gel time and lower Bingham Plastic yield stress. Settling is undesirable because the denser slag and cement can separate from the fly ash introducing a layered structure with variable properties.

Compressive strengths ranged from 5.8 to 62.0 MPa (850 to 8990 psi), and easily met the target minimum compressive strength of 3.4 MPa (500 psi). Statistical analyses indicated that the mix ratio, simulant sodium molarity, and sources of the fly ash and BFS were statistically significant parameters. The two-parameter interaction involving sodium molarity and mix ratio was also highly statistically significant. Lower compressive strengths tended to be from mixes with the 7.8M Na simulants and the 0.6 water-to-dry-blend mix ratio. The higher compressive strengths tended to be from mixes with the 5 M Na simulants and/or the 0.4 water-to-dry-blend ratio.

The EPA Method 1315 was used to measure the effective diffusivities of Tc, I, U, Cr, Na, and nitrates and nitrites. Effective diffusivities for Na, I, nitrate, and nitrite averaged over 28 to 91 days of leaching were all in the same range of 1×10^{-8} (LI = 8) to 2×10^{-9} (LI = 8.7) cm^2/s for all but one of the 26 Cast Stone mixes. Technetium effective diffusivities were in the range of 2×10^{-10} (LI = 9.7) to 6×10^{-12} (LI = 11.2) cm^2/s for the same 25 mixes and total leach interval. The sodium and technetium results are consistent with previous testing of Cast Stone waste forms with secondary waste simulants at 6M Na. Leach indices were 8.7 for sodium and 10.0 for technetium (Mattigod et al. 2011). Chromium effective diffusivities were in the range of 9×10^{-13} (LI = 12.0) to 8×10^{-15} (LI = 14.1) cm^2/s . Uranium was not detected in most of leachates from the 26 mixes, indicating that the U is retained in the Cast Stone and is not being released under the conditions of the leach test. The results of these leach tests are generally consistent with ANS/ANSI 16.1 test results reported by Lockrem (2005). Limited statistical analyses were conducted on the effective diffusivities, while more extensive statistical analyses (modeling) were performed for Na, NO_3 , I, Tc, and Cr LIs, both averaged over 28 to 63 days of cumulative leaching (the standard duration of the EPA method). The statistical analyses showed that the effective diffusivity results for Na, iodine, nitrates, and nitrites are strongly correlated. The statistical Stepwise Models for Na, NO_3 , I, Tc, and Cr LIs all had statistically non-significant lack-of-fits (see Table 8-30), indicating that they account for the variation in the test data after accounting for replicate testing and measurement uncertainty. Mix ratio has a statistically significant effect for all five component LIs, while the source of BFS has a statistically significant effect on all LIs except Na. The sodium molarity has individual or interaction effects on all five component LIs. The different simulants also were involved in statistically significant individual or two-parameter effects on all five component LIs.

The TCLP test was conducted to demonstrate that the Cast Stone waste form will meet land disposal restrictions. The simulants were spiked with Cr, Pb, Ni, and Cd, and the dry blend materials contributed As, Ba, and Se to the Cast Stone. All of the 26 mixes easily met the Universal Treatment Standards in 40 CFR 268.

Generally, for the properties for which statistical analyses were conducted, the most significant parameters in the screening test matrix were the mix ratio and the sources for the fly ash and BFS. The sodium molarity and simulant compositions were less significant unless interaction terms are considered (see Table 8-29 and Table 8-30).

The screening tests demonstrated that for the range of parameters studied, the formulations could be processed and would provide acceptable waste forms. With the exception of two mixes, the formulations have shown good processing and waste form properties and have not touched any unacceptable waste form boundaries. Mix 1 at the mix ratio of 0.4 set quickly and would not be considered an acceptable formulation. The other mix (Mix 23) at the 0.6 mix ratio had generally poorer waste form properties than the other formulations and would not be considered for use. The screening test results suggest that there are opportunities to increase waste loading by increasing the mix ratio and increasing the waste concentration.

10.0 Recommendations for the Next Phase of Development

The Cast Stone LAW screening tests documented in this report are the first step in a program to provide information to support the One-Time Hanford Tank Waste Supplemental Treatment Technologies Report and to support potential future implementation of Cast Stone to immobilize Hanford LAW. The next steps in the program include 1) optimization of the Cast Stone formulation, 2) waste form qualification to demonstrate that the optimized Cast Stone can meet anticipated waste acceptance criteria for the IDF, 3) demonstrating on an engineering scale the Cast Stone preparation process and characterizing the resulting large-scale casting of the Cast Stone waste form, 4) preparing and characterizing Cast Stone specimens with actual radioactive waste samples, and 5) conducting longer-term tests and characterizing the long-term performance of the Cast Stone waste form to support future performance assessment analyses. This program is well designed and should be continued as long as Cast Stone is being considered for the solidification of Hanford LAW.

The next step is to further optimize the Cast Stone formulation. The screening tests revealed opportunities to further increase the waste loading in the Cast Stone. Of the 26 original Cast Stone mixes in the screening test matrix, all but two mixes had acceptable processing and waste form properties. That is, the ranges of properties measured are similar enough that a processing/disposal facility can be designed to accommodate all but two of the tests compositions with the current experience. This suggests that the waste loading in the Cast Stone can be increased beyond the levels achieved in the screening tests. Waste loading can be increased by concentrating the wastes to higher sodium concentrations and by increasing the free water-to-dry blend mix ratio. It is reasonable to conduct additional formulation optimization testing with sodium molarities in the range of 7M to 10M and free water-to-dry blend mix ratios in the range of 0.5 to 0.7.

Cast Stone formulation optimization provides the opportunity to investigate methods to improve the retention of Tc and I in the waste form. One approach is to add “getter” materials to the Cast Stone mix. Getters are typically inorganic materials that selectively adsorb the specific contaminant of interest. Pierce et al. (2010) identify several potential getters including layered bismuth hydroxides, argentite (Ag_2S), silver-impregnated carbon, and Ag-zeolites as I getters; and nanoporous tin phosphates, Sn(II)-treated apatite, nano zero-valent iron (nano ZVI), and ground BFS as ^{99}Tc getters. Another approach is to add a material such as silica fume or a concrete waterproofing additive that effectively fills the porosity in the Cast Stone to effectively slow the diffusion processes. A third approach is to increase the slag content at the expense of the cement and a portion of the FA. The increased reactivity associated with additional slag could improve the physical properties of the Cast Stone and provide additional reductive capacity for retaining Tc.

One concern with the leach testing to date is that, in order to have measurable concentrations of iodine in the leachates, the iodine concentration in the Cast Stone specimens must be spiked to two orders of magnitude higher than the projected average concentration in the waste. This may lead to higher leach rates. Leach tests with a combination of iodine radiotracers and low concentrations of stable iodine representing the projected concentrations of ^{129}I in the waste would help resolve this issue. Alternative analytical methods of natural iodine detection are also available for investigation to reduce the initial concentration needed to provide detectability.

The optimization work should also consider different cement:FA:BFS dry-blend ratios. Testing to date has been at a fixed 8:45:47 blend ratio. Testing at blend ratios around this point is needed

to further demonstrate the robustness of the Cast Stone formulation to variability inherent in process control and potential process upsets. Testing could also consider a broader range of dry blend mixes. For example, if Tc is separated from the LAW, then there may not be a need for as much BFS, which could result in a cost saving if fly ash is substituted for some of the BFS in the dry blend. A program is under way to evaluate blends using less BFS to support Tc removal (Fox 2013).

To support the performance assessment for the IDF, long-term leach test data on the Cast Stone waste form are needed. The leach testing conducted as part of the screening tests provides an early opportunity to extend leach tests to longer times. Selected leach tests are being extended beyond the 91 days documented in this report. At the conclusion of the leach tests, the specimens should be characterized with respect to changes in chemical composition, mineralogy, and reductive capacity. Additional leach tests should be conducted with leachants representative of the pore waters expected to actually contact the waste forms in the disposal environment. The deionized water used in the screening test leach tests is thought to be more aggressive than what would be expected in the IDF disposal system. In addition to saturated leach testing, the effect of oxygen infiltration in IDF soil would provide data to evaluate retention/release behavior of redox-dependent constituents. Multi-component leach tests where Cast Stone is surrounded by IDF vadose zone sediments and leached in flow-through columns at unsaturated water conditions should be performed to simulate the actual IDF burial environment. McGrail et al. (2003) describe a strategy for assessing the performance of Cast Stone in the IDF.

To support the selection of Cast Stone as the supplemental waste form, future work should consider further evaluation of the engineering scale test. Additional testing at the engineering scale can be used to evaluate processing conditions and disposal options.

11.0 References

This list contains references for sources cited in the main text and in the appendixes.

10 CFR 830. Code of Federal Regulations, Title 10, Energy, Part 830, “Nuclear Safety Management.”

40 CFR 268. Code of Federal Regulations, Title 40, Protection of Environment, Part 268, “Land Disposal Restrictions.”

Amidon GE, PJ Secreast, and D Mudie. 2009. “Particle, Powder and Compact Characterization.” Chapter 8 in *Developing Solid Oral Dosage Forms: Pharmaceutical Theory & Practice*, Y Qiu, Y Chen, GGZ Zhang, L Liu, and W Porter (eds.), Academic Press (Elsevier), Burlington, Massachusetts, pp. 163–186.

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

ASME NQA-1-2000. 2001. *Quality Assurance Program for Nuclear Facilities*, American Society of Mechanical Engineers, New York.

ASME NQA-1-2004. 2004. *Quality Assurance Program for Nuclear Facilities*, American Society of Mechanical Engineers, New York.

ASTM C39. 2012. *Measuring Hydration Kinetics of Hydraulic Cementitious Mixtures Using Isothermal Calorimetry*. ASTM International, West Conshohocken, Pennsylvania.

ASTM C191. 2008. *Standard Test Methods for Time of Setting of Hydraulic Cement by Vicat Needle*. ASTM International, West Conshohocken, Pennsylvania.

ASTM C232. 2004. *Standard Test Methods for Bleeding of Concrete*. ASTM International, West Conshohocken, Pennsylvania.

ASTM C1231. 2012. *Standard Practice for Use of Unbonded Caps in Determination of Compressive Strength of Hardened Concrete Cylinders*. ASTM International, West Conshohocken, Pennsylvania.

ASTM C1679. 2009. *Standard Method for Measuring Hydration Kinetics of Hydraulic Cementitious Mixtures Using Isothermal Calorimetry*. ASTM International, West Conshohocken, Pennsylvania.

ASTM D1475. 1998. *Standard Test Method for Density of Liquid Coatings, Inks, and Related Products*. ASTM International, West Conshohocken, Pennsylvania.

ASTM D6103. 2004. *Standard Test Method for Flow Consistency of Controlled Low Strength Material (CLSM)*. ASTM International, West Conshohocken, Pennsylvania.

ASTM E1915-01. 2001. *Standard Test Methods for Analysis of Metal Bearing Ores and Related Materials by Combustion Infrared Absorption Spectrometry*. ASTM International, West Conshohocken, Pennsylvania.

Atkinson AC and AN Donev. 1992. *Optimal Experimental Design*, Oxford University Press, New York.

Bretz F, T Hothorn, and P Westfall. 2010. *Multiple Comparisons Using R*. Chapman & Hall/CRC, Boca Raton, Florida.

Bretz F, T Hothorn, and P Westfall. 2013. *On Multiple Comparisons in R*. Available at <http://ftp.auckland.ac.nz/software/CRAN/doc/vignettes/multcomp/Rmc.pdf>.

Brunauer S, PH Emmett, and E Teller. 1938. “Adsorption of Gases in Multimolecular Layers.” *J. Am. Chem. Soc.* 60:309.

Certa PJ, PA Empey, and MN Wells. 2011. *River Protection Project System Plan*. ORP-11242, Revision 6, Washington River Protection Solutions, LLC, Richland, Washington.

Cozzi AD, WE Daniel, and EK Hansen. 2013. *Sonic Velocities and other Properties of Fresh SRS Saltstone Grout*. SRNL-L3100-2013-00062, Revision 0, Savannah River National Laboratory, Aiken, South Carolina.

Cozzi AD, and BR Pickenheim. 2012. *Impact of Standing Bleed Water on Saltstone Placement*. SRNL-STI-2012-00546, Revision 0, Savannah River National Laboratory, Aiken, South Carolina.

Crank, J. 1975. *The Mathematics of Diffusion*, 2nd edition, Oxford University Press, New York.

Crawley MJ. 2007. *The R Book*. John Wiley & Sons, Ltd, The Atrium, Southern Gate, Chichester, West Sussex, England.

de Korte ACJ and HJH Brouwers. 2011. “Ultrasonic sound speed analysis of hydrating calcium sulphate hemihydrate.” *Journal of Materials Science* 46(22):7228–7239.

DOE. 2011, as amended. *Quality Assurance*. Order 414.1D, U.S. Department of Energy, Washington, D.C.

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

DOE/RL. 2007. *Hanford Analytical Services Quality Assurance Requirements Documents*. DOE/RL-96-68, Rev 3, U.S. Department of Energy Richland Operations Office, Richland, Washington.

Draper NR and H Smith. 1998. *Applied Regression Analysis*. Third Edition, John Wiley and Sons, Inc., New York, NY.

EPA. 1992. "Toxicity Characteristic Leaching Procedure." EPA Method 1311, Revision 0. In *Test Methods for Evaluating Solid Waste: Physical/Chemical Methods*. EPA SW 846, U.S. Environmental Protection Agency, Washington, D.C. Available at <http://www.epa.gov/epawaste/hazard/testmethods/sw846/pdfs/1311.pdf>.

EPA. 1996. "Microwave Accelerated Acid Digestion of Siliceous and Organically Based Matrices." EPA Method 3052 Rev. 0. In *Test Methods for Evaluating Solid Waste: Physical/Chemical Methods*. EPA SW 846, U.S. Environmental Protection Agency, Washington, D.C. Available at <http://www.epa.gov/epawaste/hazard/testmethods/sw846/pdfs/3052.pdf>.

EPA. 2007a. "Inductively Coupled Plasma-Atomic Emission Spectroscopy." EPA Method 6010C. In *Test Methods for Evaluating Solid Waste: Physical/Chemical Methods*. EPA SW 846, U.S. Environmental Protection Agency, Washington, D.C. Available at <http://www.epa.gov/epawaste/hazard/testmethods/sw846/pdfs/6010c.pdf>.

EPA. 2007b. "Inductively Coupled Plasma-Mass Spectrometry." EPA Method 6020A. In *Test Methods for Evaluating Solid Waste: Physical/Chemical Methods*. EPA SW 846, U.S. Environmental Protection Agency, Washington, D.C. Available at <http://www.epa.gov/epawaste/hazard/testmethods/sw846/pdfs/6020a.pdf>.

EPA. 2012. *Mass Transfer Rates of Constituents in Monolith or Compacted Granular Materials Using a Semi-Dynamic Tank Leaching Test*. EPA Draft Method 1315, U.S. Environmental Protection Agency, Washington, D.C. Current version available at <http://www.epa.gov/epawaste/hazard/testmethods/sw846/pdfs/1315.pdf>.

Fox KM. 2013. *Cast Stone Formulation at Higher Sodium Concentrations*. SRNL-STI-2013-00499, Revision 0, Savannah River National Laboratory, Aiken, South Carolina.

Gasper KA, KD Boomer, ME Johnson, GW Reddick, Jr., AF Choho, and JS Garfield. 2002. *Recommendation for Supplemental Technologies for Potential Mission Acceleration*, RPP-11261, Revision 0, CH2M HILL Hanford Group, Inc., Richland, Washington.

Hansen EK, DT Herman, and VJ Williams, 2006. *Saltstone Sheet Drain Performance Results*. WSRC-TR-2006-00058, Revision 0, Savannah River National Laboratory, Aiken, South Carolina.

Hansen EK, AD Marzolf, and KR Hera. 2012. *2012 SRNL-EM Vane Rheology Results*. SRNL-STI-2012-00519, Revision 0, Savannah River National Laboratory, Aiken, South Carolina.

Harbour JH, VJ Williams, and TB Edwards. 2007. *Heat of Hydration of Saltstone Mixes-Measurement by Isothermal Calorimetry*. WSRC-STI-2007-00263, Rev. 0, Savannah River National Laboratory, Aiken, South Carolina.

Hothorn T, F Bretz, and P Westfall. 2013. *Package 'multcomp' – Simultaneous Inference in General Parametric Models*. Available at <http://cran.r-project.org/web/packages/multcomp/multcomp.pdf>.

Lockrem LL. 2005. *Hanford Containerized Cast Stone Facility Task 1-Process Testing and Development Final Report*. RPP-RPT-26742 Revision 0, CH2M Hill Hanford Group, Inc., Richland, Washington.

Macosko CW. 1994. "Plastic Behavior." In *Rheology Principles, Measurements, and Applications*, Macosko CW (ed.). Advances in Interfacial Engineering Series; Wiley-VCH Inc., New York, New York; pp. 92–108.

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

McCarthy GJ, JK Solem, OE Manz, and DJ Hasset. 1989. "Use of a Database of Chemical, Mineralogical and Physical Properties of North American Fly Ash to Study the Nature of Fly Ash and Its Utilization as a Mineral Admixture in Concrete." In *MRS Proceedings* 178:3, Cambridge University Press.

McGrail BP, DH Bacon, RJ Serne, and EM Pierce. 2003. *A Strategy to Assess Performance of Selected Low-Activity Waste Forms in an Integrated Disposal Facility*. PNNL-14362, Pacific Northwest National Laboratory, Richland, Washington.

Pierce EM, SV Mattigod, RJ Serne, JP Icenhower, RD Scheele, W Um, N Qafoku, and JH Westsik, Jr. 2010. *Review of Potential Candidate Stabilization Technologies for Liquid and Solid Secondary Waste Streams*. PNNL-19122, Pacific Northwest National Laboratory, Richland, Washington.

R Core Team (2013). *R: A Language and Environment for Statistical Computing*. R Foundation for Statistical Computing, Vienna, Austria. ISBN 3-900051-07-0, URL <http://www.R-project.org/>.

Ramachandran VS and RF Feldman. 1995. "Concrete Science." In *Concrete Admixtures Handbook Properties, Science and Technology*, Second Edition, VS Ramachandran (ed.), Building Materials Science Series; Noyes Publications, Park Ridge, New Jersey; pp. 1–60.

Rapko BM, SI Sinkov, and TG Levitskaia. 2003. *Removal of ¹³⁷Cs from Dissolved Hanford Tank Saltcake by Treatment with IE-911*. PNNL-14250, Revision 1, Pacific Northwest National Laboratory, Richland, Washington.

Rassat SD, LA Mahoney, RL Russell, SA Bryan, and RL Sell. 2003. *Cold Dissolved Saltcake Simulant Development, Preparation, and Analysis*. PNNL-14194, Rev 1, Pacific Northwest National Laboratory, Richland, Washington.

Resource Conservation and Recovery Act of 1976. 1976. Public Law 94-580, as amended, 42 USC 6901 et seq. and 42 USC 6927(c) et seq.

Robeyst N, E Gruyaert, CU Grosse, and N De Belie. 2008. "Monitoring the setting of concrete containing blast-furnace slag by measuring the ultrasonic p-wave velocity." *Cement and Concrete Research* 38:1169–1176.

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

Savannah River National Laboratory (SRNL). 2010. *Lithium Tetraborate Fusion Method (U) Revision 1*. Procedure Manual L29, ITS-0070, Aiken, South Carolina.

Savannah River National Laboratory (SRNL). 2010. *Operation of the Quantachrome Multipycnometer Revision 0*. Procedure Manual L29, ITS-0168, Aiken, South Carolina.

Stat-Ease. 2010. *Design-Expert Version 8.0.7.1*. Stat-Ease, Inc., Minneapolis, Minnesota.

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

Valenta G and T Füllmann. 2004. “Advances in Quantitative XRD Analysis for Clinker, Cements, and Cementitious Additions. *Advances in X-ray Analysis* 47:287–296.

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

Appendix A. Statistical Design of the Screening Test Matrix

This appendix discusses how the screening test matrix was developed using statistical experimental design methods and software. The screening test matrix was developed in two “blocks” in case project funding might be limited and only the Block 1 tests could be performed. However, sufficient funding was provided, so both Block 1 and Block 2 of the experimental design were performed.

Section A.1 describes the optimal experimental design (OED) methodology used to develop the test matrix. Sections A.2 and A.3 describe how the two blocks of the test matrix were developed.

A.1 Optimal Experimental Design

The test matrix (both the Block 1 and Block 2 portions) was selected using OED methods (Atkinson and Donev 1992) implemented using the Design-Expert software (Stat-Ease 2010). OED seeks to select the tests in a test matrix so as to minimize or maximize a statistical criterion based on some measure of goodness of the test matrix. The optimality criteria considered in developing the Cast Stone screening experimental design (both Block 1 and Block 2) included

- D-optimality: Minimize $|(X'X)^{-1}|$
- I-optimality: Minimize Average($x'(X'X)^{-1}x$)

where X denotes the test matrix with columns expanded in the form of terms in a specified model form, the prime denotes matrix transpose, the superscripted -1 denotes the matrix inverse, the vertical bars denote the determinant of a matrix (a scalar), and x denotes a vector of test combinations (expanded in the form of the specified model) within the allowable set (referred to as the candidate set) of test combinations. The D-optimality criterion seeks to reduce the size of the confidence ellipsoid on estimates of the model coefficients, while the I-optimality criterion seeks to minimize the average variance of property predictions made with the specified model. For categorical parameters (or numeric parameters with discrete values), the average is over the allowable discrete combinations. For numeric parameters with continuous values allowed, the average is over the whole space of numeric combinations.

OED methods rely on optimization algorithms that do not guarantee “the” optimal solution on a given “try.” Hence, software typically makes many “tries” from different random starting designs and selects the best design from all the tries according to the chosen optimality criterion.

A.2 Screening Test Matrix, Block 1

The Block 1 test matrix focused on a screening assessment of the effects of the five individual test parameters discussed in Section 2.0. To enable the use of OED methods and software to develop the Block 1 test matrix, the following model for individual effects of the test parameters was specified:

$$y = \beta_0 + \beta_1(S_{SSTBlend-Avg} + \beta_2 S_{HighAl-Avg} + \beta_3 S_{HighSO4-Avg} + \beta_4 WC + \beta_5 FA + \beta_6 BFS + \beta_7 MR + \varepsilon \quad (A-1)$$

where y = a measured property (possibly mathematically transformed, e.g., logarithm transformation) of a Cast Stone waste form

$S_{SSTBlend-Avg}$ = 1 for the SST Blend simulant, = 0 otherwise

$S_{HighAl-Avg}$ = 1 for the High Al simulant, = 0 otherwise

- $S_{HiSO4-Avg}$ = 1 for the High SO_4 simulant, = 0 otherwise
- WC = waste concentration, quantified as Na molarity, where 5 M was coded as 0, and 7.8 M was coded as 1
- FA = fly ash, where SE (low Ca) = 0, NW (high Ca) = 1
- BFS = blast furnace slag, where NW = 0, SE = 1
- MR = mix ratio, quantified as the grams of free water in the waste simulant per gram of dry blend solids. MR = 0.4 was coded as 0, and MR = 0.6 was coded as 1.
- β_0, \dots, β_7 = model coefficients to be estimated from test data using least squares regression
- ε = random error from testing and measuring property y for a given test.

To use ordinary least squares (OLS) regression to fit the model coefficients to experimental data obtained from a test matrix, the random errors must be independently and identically distributed according to a normal distribution. One aspect of this assumption is that the random errors must have constant variance over all test combinations in the test matrix. Often when these assumptions are not satisfied for a given property, they will be satisfied for a mathematical transformation (e.g., logarithm) of the property. Typically, a commonly used transformation of the property has a subject-matter basis as well as the statistical basis. The assumption of equal variance was assessed as part of data analyses performed on the data from the test matrix, the results of which are discussed in Section 8.0.

Although the simulant, fly ash (FA), and blast furnace slag (BFS) test parameters are categorical, numeric coding for their settings was used to enable specifying a numeric model form in Equation (A-1). However, with this numeric coding convention, the FA and BFS parameters can only take the values of 0 and 1; intermediate values are not appropriate. The waste concentration (WC) and mix ratio (MR) test parameters are numeric, and conceptually could take a value anywhere in the selected testing range of each parameter. However, for the screening test matrix, it was decided that WC and MR would only take values equal to their lower and upper bounds. Those values were not decided upon at the time that the test matrix was constructed, so the 0 and 1 coded values of WC and MR (along with FA and BFS) were used to develop the test matrix.

Because the Simulant test parameter is categorical with four possible simulants, choices had to be made to enable numeric coding of the four simulants. The coding of the chosen Simulant test parameter involves comparing (via differencing) each of the SST Blend, High Al, and High SO_4 simulants to the Average simulant. Obviously, other comparisons could be made and may be of interest. However, this set of comparisons was chosen because it was believed that there would not be much difference in property values for the Average and SST Blend simulants, and so comparing those two simulants was of interest. It was then natural to also compare the High Al and High SO_4 simulants to the Average simulant.

With all of the test parameters (and simulant comparisons) coded 0 and 1 as discussed, note that the OLS estimate of the β_0 coefficient is the model-predicted property value when all parameters and simulant comparisons are set to their 0 coded values. That corresponds to the Average simulant, 5.0 Na molarity, southeast (SE; low-Ca) FA, northwest (NW) BFS, and MR = 0.4. The OLS estimates of β_1 , β_2 , and β_3 are predictions of the increase in the property value when changing from the Average Simulant to the SST Blend, High Al, and High SO_4 simulants,

respectively. The OLS estimates of β_4 , β_5 , β_6 , and β_7 are predictions of the increase in the property value when changing the WC, FA, BFS, and MR parameters, respectively, from their 0 to 1 coded values.

Block 1 of the test matrix was designed to include 12 distinct tests and 2 replicates, for a total of 14 tests. These numbers of tests and replicates provided a minimal basis for 1) fitting the model (A.1) to property data from the tests, 2) assessing the testing and measurement uncertainty in the property values, and 3) assessing the lack-of-fit (LOF) when fitting the model in Equation (A-1) to experimental data.

The Design-Expert software (Stat-Ease 2010) was used to first select the 12 distinct tests for Block 1 using the D-optimality criterion based on the model in Equation (A-1). The Design-Expert's default set of 128 candidate points was too large because of the special nature of coding the four simulants as three comparisons. Hence, a set of 64 candidate points was generated and supplied to Design-Expert to use in selecting the 12 test conditions. The number of candidate points results from $(4 \text{ simulants}) \times (2 \text{ WC values}) \times (2 \text{ FA values}) \times (2 \text{ BFS values}) \times (2 \text{ MR values}) = 64$ combinations. After selecting the 12 distinct tests for Block 1 of the test matrix, a separate run was made in Design-Expert to select 2 of the 12 design points as replicates, again using D-optimality as the OED criterion based on the model in Equation (A-1). The resulting 12 tests plus 2 replicates are listed as Tests 1 through 14 in Table 2-1 of Section 2.0 in the main body of this report.

A.3 Screening Test Matrix, Block 2

Block 2 of the test matrix was developed using OED methodology to augment the Block 1 test matrix with additional tests to support the assessment of whether selected pairs of parameters have significant two-parameter interactive effects on Cast Stone properties. A two-parameter interaction occurs when the effect of one parameter on a property depends of the value of a second parameter.

As guidance for developing the test matrix, it was thought that the Average and SST Blend simulants would have similar effects on the Cast Stone properties, because each simulant was an estimate of the average LAW composition. Hence, it was thought that interactions of other parameters with the difference in the effects of the SST Blend and Average simulants would not be significant. Those interactions are the first three listed in the left column of Table A-1. Further, it was believed that there would not be much difference in the effects of the two sources of BFS on Cast Stone properties. Hence, it was believed that interactions of other parameters with BFS would not be significant. Those interactions are the final six interactions listed in the left column of Table A-1.

Table A-1. Two-Parameter Interactions Thought Not to Have Significant Effects and the Remaining Two-Parameter Interactions Planned for in Constructing Block 2 of the Test Matrix

Two-Parameter Interactions Thought Not to Have Significant Effects	Remaining Two-Parameter Interactions Planned for in the Test Matrix
(SST Blend vs. Average)*Waste Concentration	(High Al vs. Average)*Waste Concentration
(SST Blend vs. Average)*Fly Ash	(High SO ₄ vs. Average)*Waste Concentration
(SST Blend vs. Average)*Mix Ratio	(High Al vs. Average)*Fly Ash
BFS*(High Al vs. Average)	(High SO ₄ vs. Average)*Fly Ash
BFS*(High SO ₄ vs. Average)	(High Al vs. Average)*Mix Ratio
BFS*(SST Blend vs. Average)	(High SO ₄ vs. Average)*Mix Ratio
BFS*Waste Concentration	Fly Ash*Waste Concentration
BFS*Fly Ash	Waste Concentration*Mix Ratio
BFS*Mix Ratio	Fly Ash*Mix Ratio

The two-parameter interactions remaining (nine of them) are listed in the right column of Table A-1. The model listed in Equation (A-1) was expanded to include terms for these nine interactions:

$$\begin{aligned}
 y = & \beta_0 + \beta_1(S_{SSTBlend-Avg} + \beta_2 S_{HighAl-Avg} + \beta_3 S_{HighSO4-Avg} \\
 & + \beta_4 WC + \beta_5 FA + \beta_6 BFS + \beta_7 MR + \beta_8 (S_{HighAl-Avg})^*(WC) \\
 & + \beta_9 (S_{HighSO4-Avg})^*(WC) + \beta_{10} (S_{HighAl-Avg})^*(FA) \\
 & + \beta_{11} (S_{HighSO4-Avg})^*(FA) + \beta_{12} (S_{HighAl-Avg})^*(MR) \\
 & + \beta_{13} (S_{HighSO4-Avg})^*(MR) + \beta_{14} (FA)^*(WC) + \beta_{15} (WC)^*(MR) \\
 & + \beta_{16} (FA)^*(MR) + \beta_{17} Block + \varepsilon
 \end{aligned}
 \tag{A-2}$$

where β_8 to β_{16} are model coefficients of the interaction terms, β_{17} is the model coefficient of the “block effect” term, and all other notation is the same as defined for Equation (A-1).

Block 2 of the test matrix was designed to contain 10 distinct test combinations and two replicates, so that together the whole test matrix (Blocks 1 and 2) contained 22 distinct test combinations and 4 replicates for a total of 26 tests. Design-Expert (Stat-Ease 2010) was used to select the 10 additional distinct test combinations to augment the 14 tests in Block 1, based on D-optimality and the model in Equation (A-2). The 10 additional test combinations were selected from the subset of the 64 candidate points discussed previously that were not selected for Block 1. Finally, Design-Expert was used to select the two replicates for Block 2 from among the 10 distinct test combinations chosen for Block 2. The resulting $10 + 2 = 12$ tests in Block 2 of the test matrix are listed as Tests 15–26 in Table 2-1 of Section 2.0.

Appendix B. Dry Blend Materials Characterization

Appendix B provides additional information about the characteristics of the individual blast furnace slag (BFS), fly ash (FA), and ordinary portland cement (OPC) dry blend components used in preparing the Cast Stone specimens used in these screening tests. This appendix also describes the chemical analysis methods used to characterize the waste simulants, dry materials, Cast Stone specimens, and EPA Draft Method 1315 leach tests.

B.1 Blast Furnace Slag Properties

Figure B-1 shows scanning electron microscopy (SEM) micrographs of the slags used in this study. The delay in onset of the reaction of slags with gypsum added is demonstrated in the isothermal calorimeter data shown in Figure B-2. For the SE slag, the onset of the heat generation peak is consistent with previously reported results (Harbour et al. 2007). Calorimeter data in Figure B-3 show that the total heat generated by the NW slag exceeds that of the SE slag after 80 hours. For the SE slag, the total heat generated is comparable with previously reported results over a similar time period (Harbour et al. 2007). Figure B-4 and Figure B-5 show the volumetric particle size distribution and thermal gravimetric analyses of the two slags. The surface areas of the NW and SE slags are 3.03 and 2.87 m²/g as measured by Brunauer–Emmett–Teller (BET) analysis (Brunauer et al. 1938). The true particle density and the aerated and tap densities for the slags are listed in Table B-1 (see Section B.10 of the methods used). The table includes the calculated Hausner ratio, the tap density divided by the aerated density. The Hausner ratio is related to flowability. A Hausner ratio <1.46 is considered poor to very poor (Amidon et al. 2009).

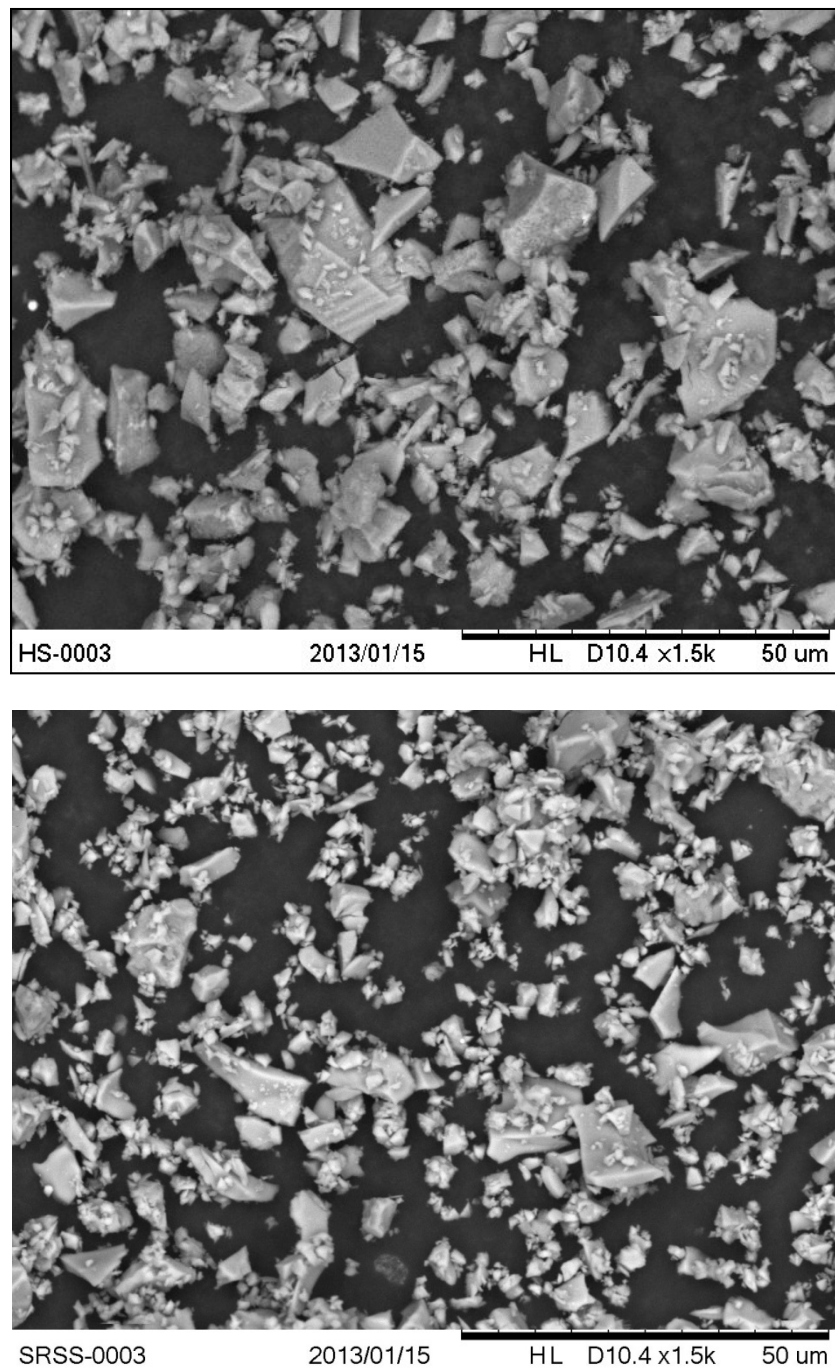


Figure B-1. Micrographs (1500x) of the Northwest (top) and Southeast (bottom) Blast Furnace Slags

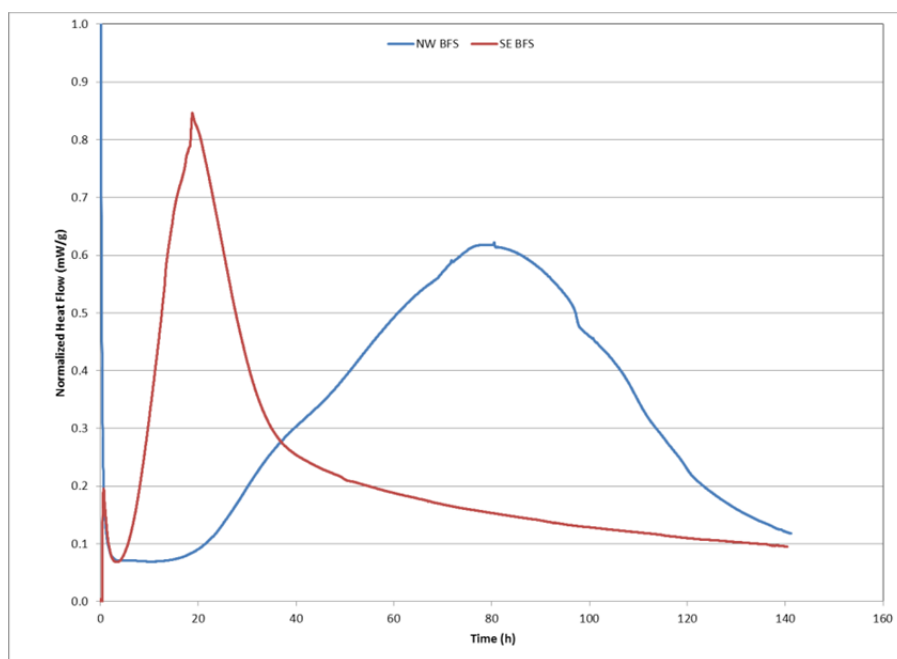


Figure B-2. Normalized Heat Flow Blast Furnace Slags (BFSs) Mixed with Water (1:0.4 BFS:Water)

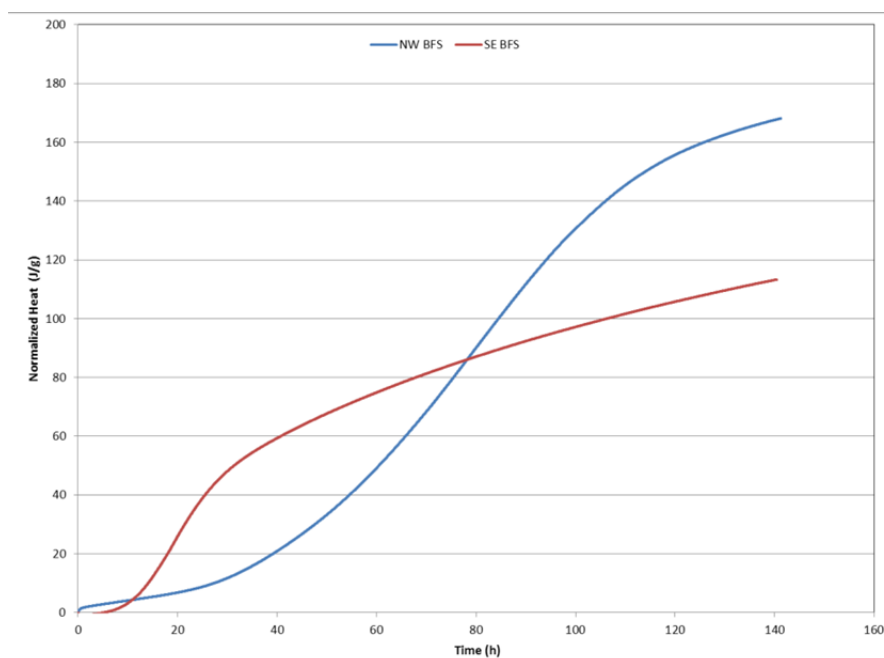


Figure B-3. Normalized Total Heat Generated by Blast Furnace Slags with Water (1:0.4 BFS:Water)

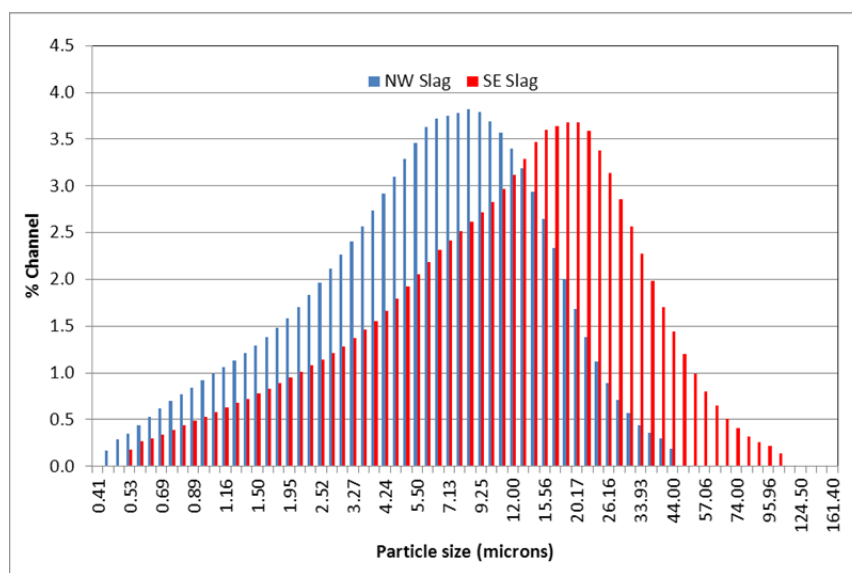


Figure B-4. Volumetric Particle Size Distribution of Blast Furnace Slags

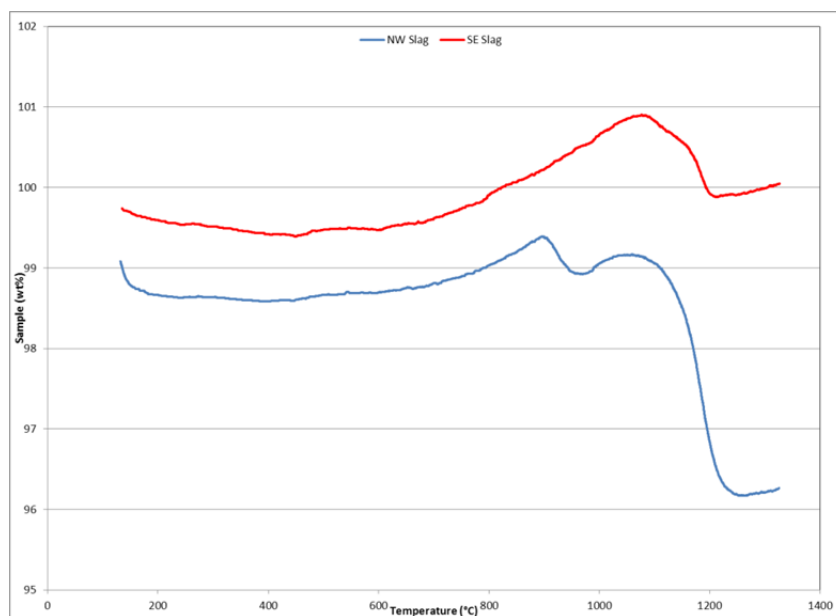


Figure B-5. Thermal Gravimetric Analysis of the Blast Furnace Slags

Table B-1. Density and Hausner Ratio of Blast Furnace Slags

Slag Source	True Density (g/ml)	Aerated Density (g/ml)	Tap Density (g/ml)	Hausner Ratio
Northwest	2.881	0.78	1.12	1.44
Southeast	2.907	0.82	1.37	1.68

B.2 Fly Ash Properties

Figure B-6 shows SEM micrographs of the FAs used in this study. Figure B-7 and Figure B-8 show that in water, only the NW sourced fly ash produced any reaction. The minimal pozzolanic activity in the SE sourced fly ash has also been observed with salt solutions (Harbour et al. 2007). Figure B-9 and Figure B-10 show the volumetric particle size distribution and thermal gravimetric analyses of the two FAs. The surface areas of the NW and SE fly ashes are 0.79 and 0.87 m²/g as measured by BET analysis. The true density and the aerated and tap densities for the slags are listed in Table B-2.

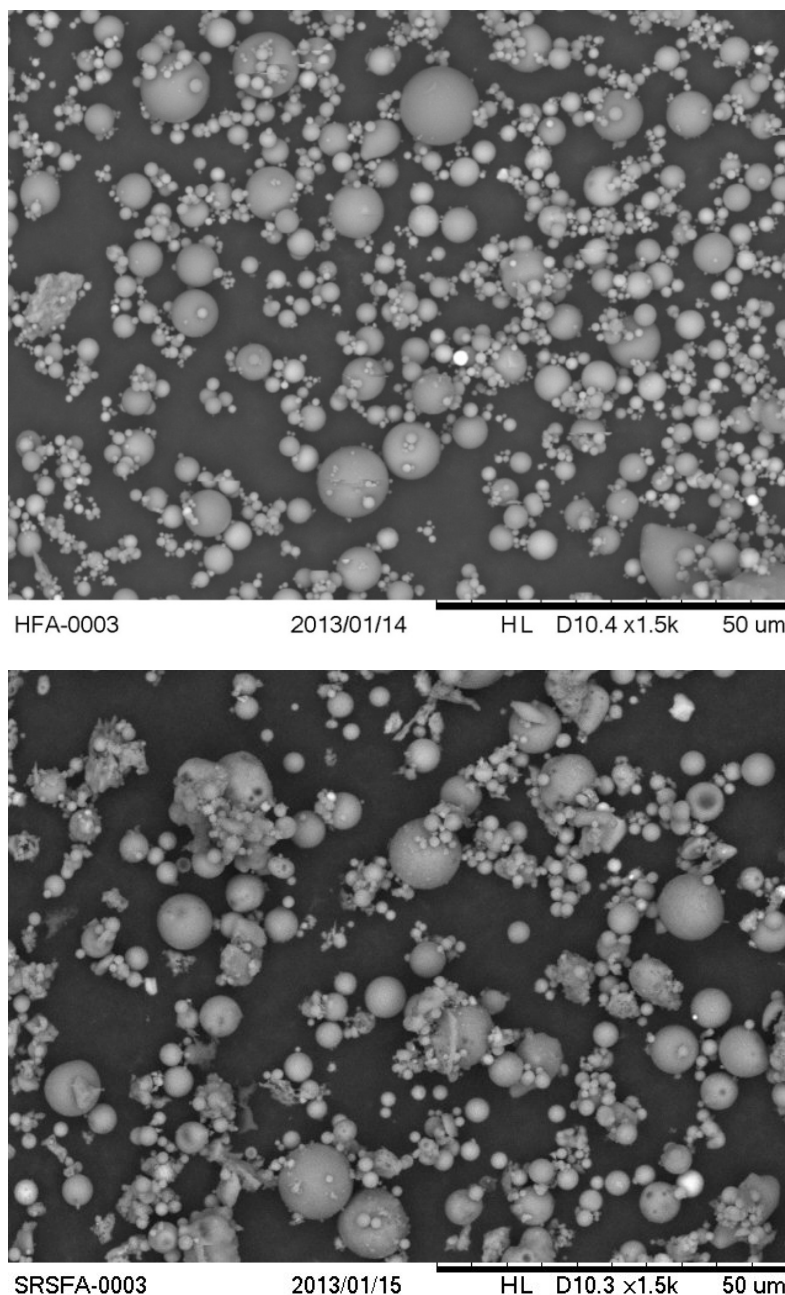


Figure B-6. Micrographs (1500x) Northwest (top) and Southeast (bottom) Fly Ashes

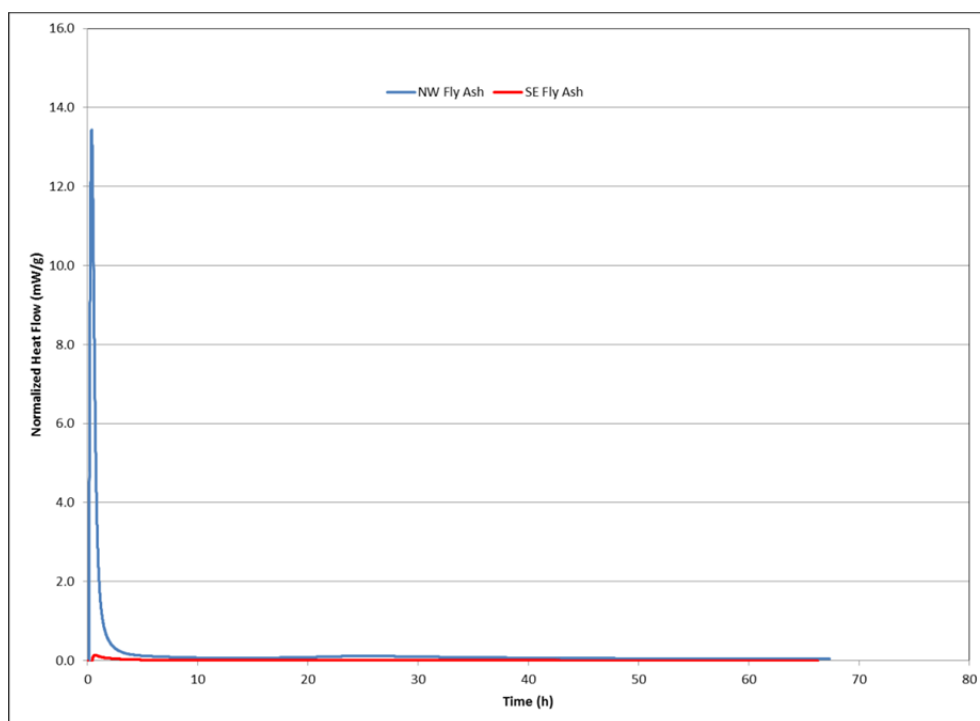


Figure B-7. Normalized Heat Flow of Fly Ashes Mixed with Water (1:0.4 FA:Water)

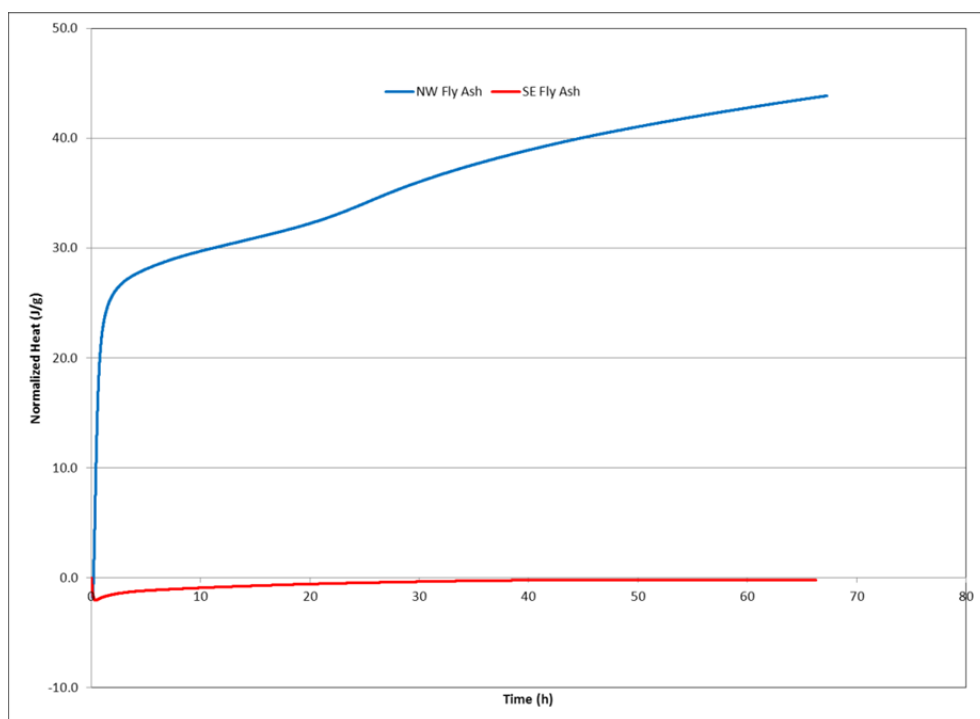


Figure B-8. Normalized Total Heat Generated by Fly Ashes with Water (1:0.4 FA:Water)

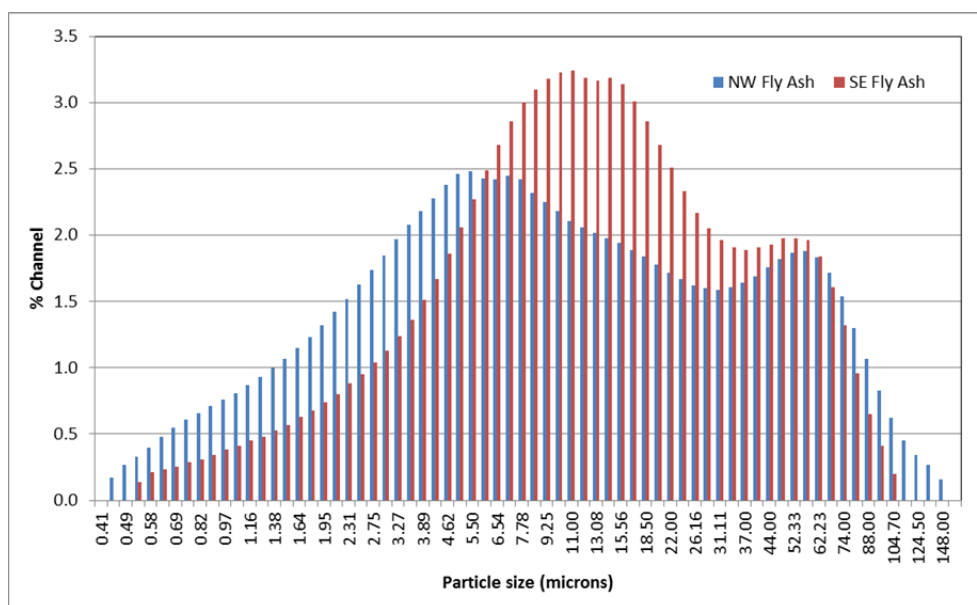


Figure B-9. Volumetric Particle Size Distribution of Fly Ashes

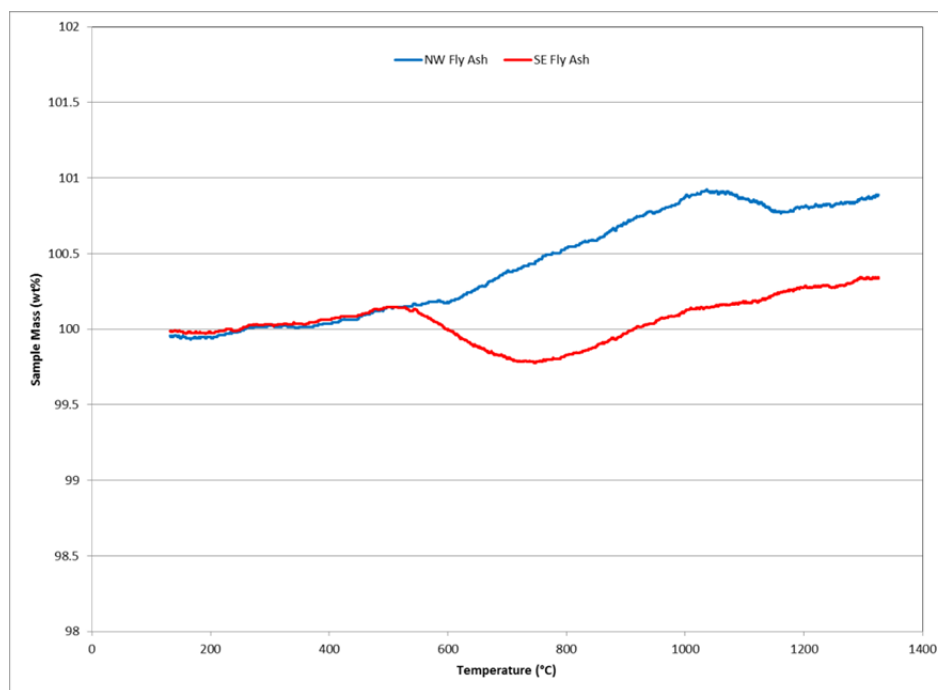


Figure B-10. Thermal Gravimetric Analysis of the Fly Ashes

Table B-2. Density and Hausner Ratio of Fly Ashes

Fly Ash Source	True Density (g/ml)	Aerated Density (g/ml)	Tap Density (g/ml)	Hausner Ratio
Northwest	2.588	0.98	1.59	1.63
Southeast	2.432	0.74	1.29	1.75

B.3 Ordinary Portland Cement Properties

Figure B-11 is an SEM micrograph of the NW sourced ordinary portland cement (OPC). The normalized heat flow and total heat of a 0.4 water-to-cement mix is shown in Figure B-12. The values are expected for mixes of this ratio with OPC. Figure B-13 and Figure B-14 show the volumetric particle size distribution and thermal gravimetric analyses of the OPC. The surface area of the cement as measured by BET is $1.57 \text{ m}^2/\text{g}$. The true density and the aerated and tap densities for the cement are listed in Table B-3.

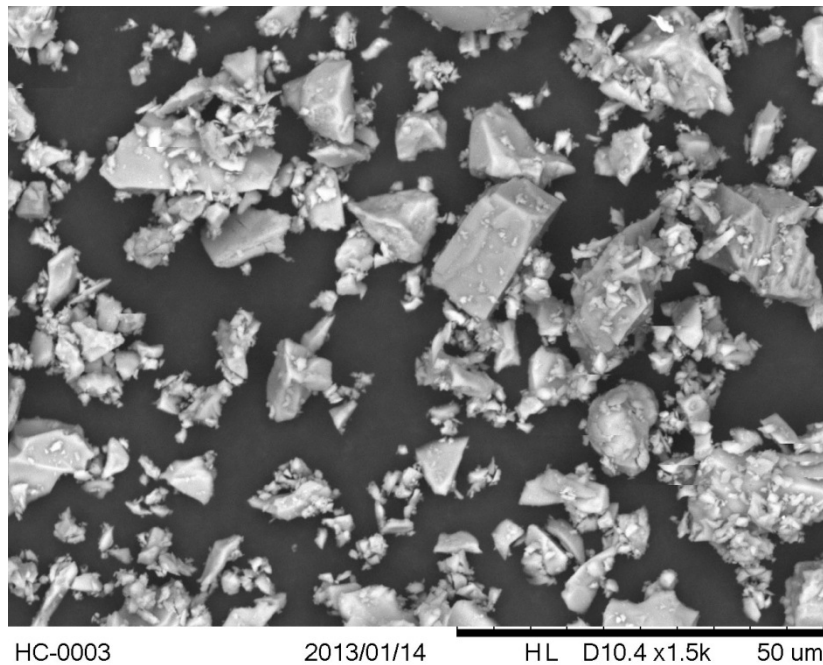


Figure B-11. Micrograph of Ordinary Portland Cement (1500x)

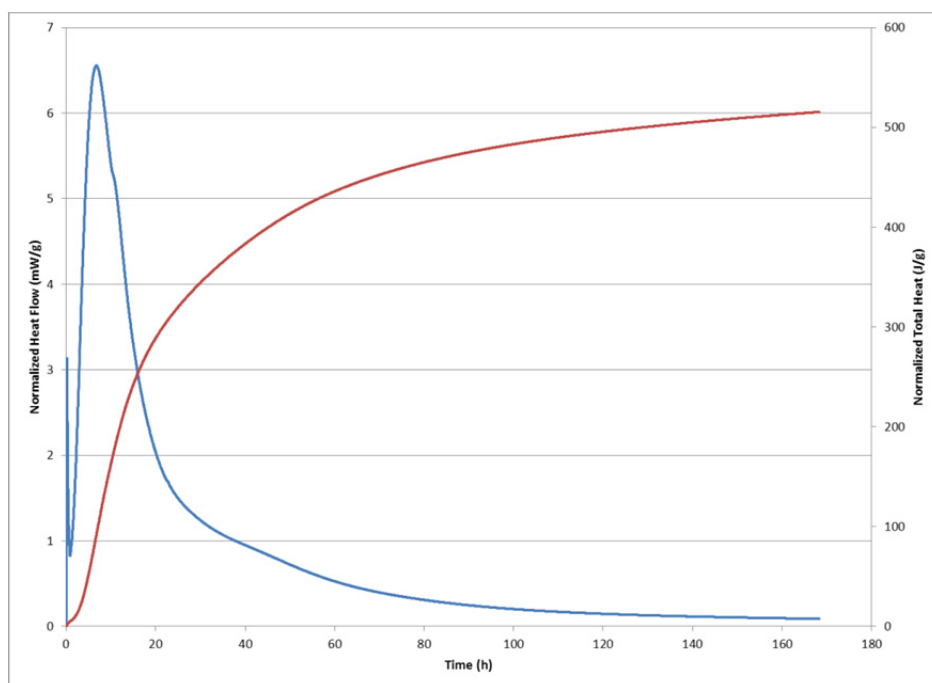


Figure B-12. Normalized Heat Flow and Total Heat from Ordinary Portland Cement with Water (1:0.4 OPC:Water)

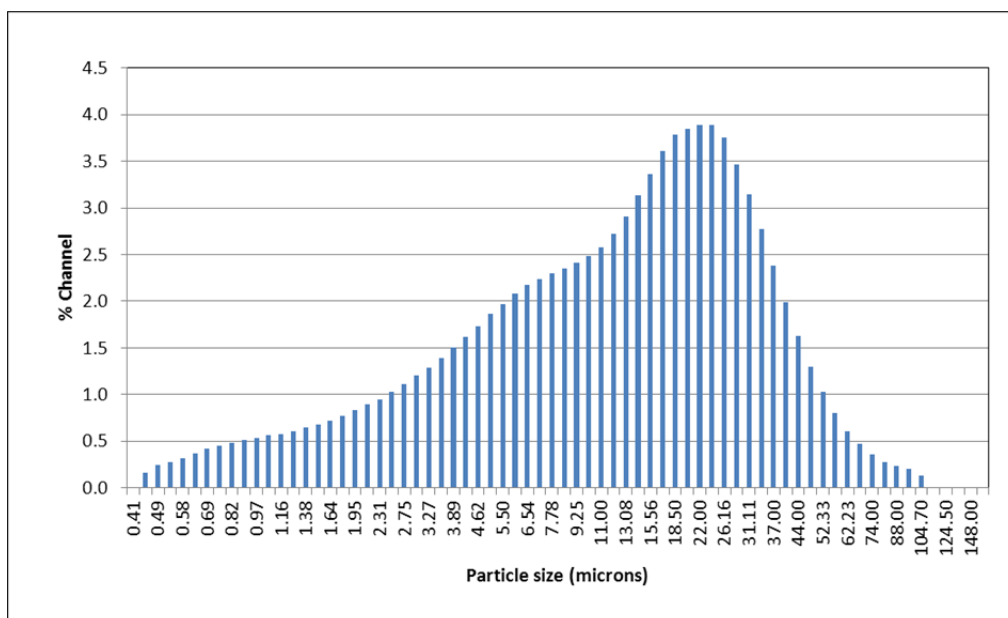


Figure B-13. Volumetric Particle Size Distribution of Ordinary Portland Cement

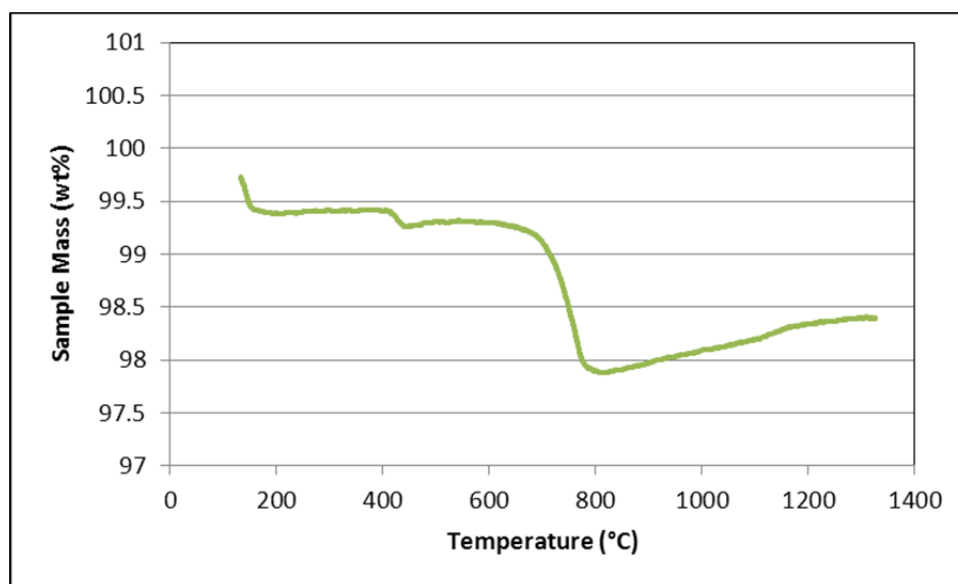


Figure B-14. Thermal Gravimetric Analysis of the Cement

Table B-3. Density and Hausner Ratio of Ordinary Portland Cement

Cement Source	True Density (g/ml)	Aerated Density (g/ml)	Tap Density (g/ml)	Hausner Ratio
Northwest	3.204	1.00	1.56	1.56

B.4 Final Dry Blend Mix Properties

Figure B-15 shows the volumetric particle size distributions of the four dry blends calculated from the measured particle size distribution of the individual components and the dry blend ratio. Table B-4 is the calculated density, measured aerated and tap densities, and calculated Hausner ratio of the dry blends. The flowability of a powder with a Hausner ratio >1.60 is considered very, very poor (Amidon et al. 2009).

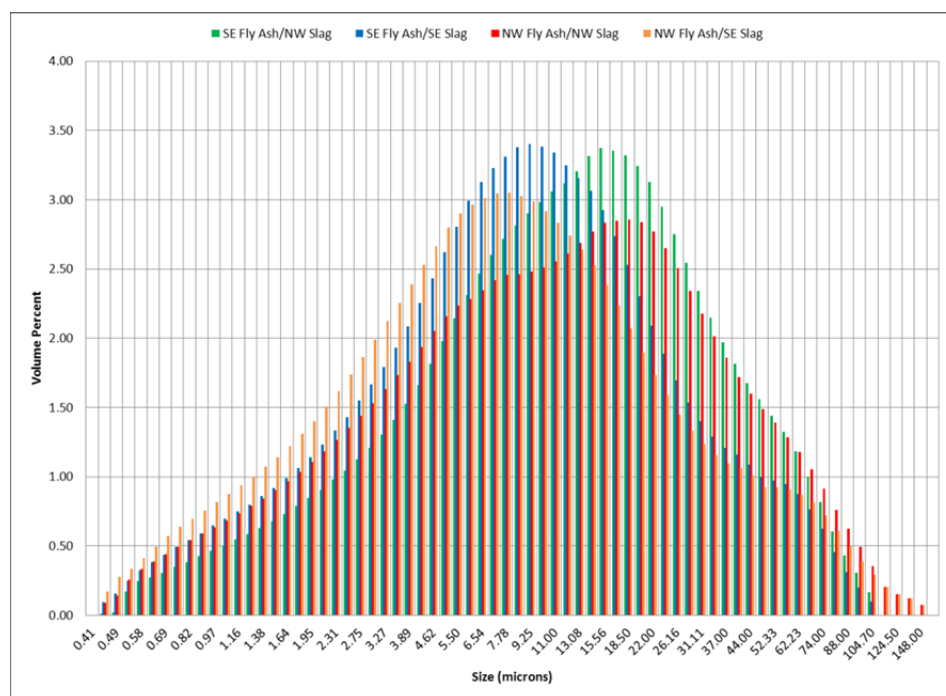


Figure B-15. Calculated Particle Size Distribution of the Dry Blends

Table B-4. Density and Hausner Ratio of Dry Blends

Source		True Density (g/ml) ^(a)	Aerated Density (g/ml)	Tap Density (g/ml)	Hausner Ratio (unitless)
Fly Ash	Slag				
Southeast	Northwest	2.6802	0.80	1.35	1.68
Southeast	Southeast	2.6907	0.72	1.19	1.65
Northwest	Northwest	2.7628	0.88	1.44	1.63
Northwest	Southeast	2.7740	0.74	1.36	1.61
(a) Calculated from measured density of individual components and blend ratios.					

B.5 Chemical Analysis Methods

This section describes the analytical methods used to characterize the chemical compositions of the waste simulants, dry blend components, Cast Stone specimens, and leachates from the EPA Draft Method 1315 (2012) leach tests.

B.5.1 Sample Preparation- Microwave Digestion

The three dry materials used to make Cast Stone were individually chemically digested at PNNL to determine their elemental composition. Microwave-assisted strong acid digestions were conducted using a combination of 16 M HNO₃ (~17 wt%), 12 M HCl (7 wt%), 32 M HF (3.3 wt%), 0.5 grams of H₃BO₃ (1.5 wt%), and deionized water (DIW; 71.2 wt %). The dry materials were first pulverized to powders and digested following the PNNL-AGG-MARS-001¹

¹ Lindberg MJ. 2011. "Operation of the MARS 5 Microwave Accelerated Reaction System." AGG-MARS-001 (Rev. 3), unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

method, which was modified from EPA Draft Method 3052 (EPA 1996). The dry materials solid-acid mixture ratio (0.1 g/ 35 mL) was typically reacted for 1 hour at $90 \pm 5^\circ\text{C}$. Upon complete dissolution of the sample, the resulting solution was centrifuged, filtered through a 0.45- μm membrane, and analyzed for trace metals using inductively coupled plasma-mass spectroscopy (ICP-MS) and major cations using inductively coupled plasma-optical emission spectroscopy (ICP-OES). The results of the elemental composition measurements were converted to elemental oxide composition to estimate the mass balance of each solid.

B.5.2 Sample Preparation, Cations- Lithium Tetraborate Fusion

The dry materials used to make Cast Stone were chemically digested to determine the elemental composition. The powdered samples were prepared following the Savannah River National Laboratory (SRNL) L29, ITS-0070 method (SRNL 2010). Samples were pulverized using a Wig L Bug grinding mill with agate grinding media. The samples were ground to a fine powder to increase the surface area of the sample that comes in contact with the flux material. The $\text{Li}_2\text{B}_4\text{O}_7$ flux is prepared in 50-ml platinum crucibles by adding 0.1 g of sample to the crucible and 0.5 g of $\text{Li}_2\text{B}_4\text{O}_7$. The sample is mixed and another 0.5 g of sample is added on top of the mixed sample. The crucible is placed in a furnace at 1000°C for 30 minutes and then taken out to cool. A stir bar is added to the crucible after cooling and 20 ml of 4% nitric acid and 2 ml of concentrated HCl are added to the crucible. The sample is placed on a heater/stir plate set at 90°C . After the sample has dissolved, the contents are poured into a 100-ml volumetric flask and the volume is brought to 100 ml with DIW.

B.5.3 Sample Preparation, Anions- Potassium Hydroxide Fusion

For anion analyses, a 1-g sample of the pulverized sample is added to a 50-ml platinum crucible. Three grams of KOH are added to the crucible. The crucible is then placed in a heating block set at 360°C . The heating block is placed on a gyratory shaker set at 125 rpm. The flux is heated and shaken for at least 1 hour or until the sample solidifies. The crucible is then removed, allowed to cool and a stir bar is added. Ten milliliters of DIW are added to the crucible and the sample is stirred for 30 minutes at 90°C . The sample digestion is then transferred to a 50-ml volumetric flask and brought to volume with DIW.

B.5.4 Percent Solids

The percent solids of dry blend materials and cured Cast Stone was determined gravimetrically by drying a 10-g aliquot of the material in an oven at 105°C . The sample is dried to a constant weight and the percent solid is calculated by the mass change before and after drying.

B.5.5 Major Cation Analysis

At Pacific Northwest National Laboratory (PNNL), major cation analysis (including Al, Si, Ca, Mg, Na, K, Fe, and Mn) for both solids digestates and solution leachates were performed using a Perkin Elmer OPTIMA 3300 DV (Waltham, MA) inductively coupled plasma-atomic emission spectrometer (ICP-AES or ICP-OES) using procedure PNNL-AGG-ICP-AES, Determination of Element by ICP-AES,¹ which is similar to Test Methods for Evaluating Solid Wastes: Physical/Chemical Methods SW-846 6010C (EPA 2007a). High-purity calibration standards

¹ Baum SR. 2008. "Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES) Analysis." PNNL-AGG-ICP-AES (Rev. 2), unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

were used to generate calibration curves and verify continuing calibration during the analytical run. A serial dilution was made of select samples to investigate and correct for matrix interferences.

At SRNL, elements were analyzed using Agilent's Varian 730-ES® Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES). The instrument is calibrated using matrix matched standards at 1, 5 and 10 ppm. A glass cyclonic spray chamber and a glass Meinhard C3® concentric nebulizer are used. Gas flow rates consist of a plasma gas at 16.5 L/min, auxiliary gas at 1.5 L/min, and the nebulizer gas at 1.2 L/min. The radio frequency generator is set at 1.2 kW. The sample digestion is diluted by 10X in a Li₂B₄O₇ matrix blank for elements greater than 1 wt% and the sample digestion is also run straight for elements that are less than 1 wt%. Elemental concentrations are reported as a wt% element and also wt% elemental oxide.

B.6 Anion Analysis

Anions are run on a Dionex ICS-5000 capillary ion chromatography (IC) system at both national laboratories. A gradient run using 0.2 mM to 35 mM of KOH is used. The anions run on the KOH digestion are nitrite, nitrate, fluoride, chloride, and oxalate. The instrument is calibrated using 1, 5 and 10 ppm of each anion in water. The flux is filtered and a 100X dilution from the filtrate in water is made to run through the instrument for analyses.

B.7 Trace Metals Analysis

Trace metals for both solids digestates and solution leachates were measured at PNNL using a Perkin Elmer ELAN DRC II (Waltham, MA) inductively coupled plasma-mass spectrometer (ICP-MS) following procedure PNNL-AGG-415, "Inductively Coupled Plasma Mass Spectrophotometry (ICP-MS) Analysis,"¹ which is similar to Test Methods for Evaluating Solid Wastes: Physical/Chemical Methods (SW-846 6020A; EPA 2007b). High-purity calibration standards were used to generate calibration curves and verify continuing calibration during the analytical run. A serial dilution was made of select samples to investigate and correct for matrix interferences. Typical instrument detection limits for trace metals using this ICP-MS are in the parts per trillion range.

B.8 Uranium, Technetium, and Iodine Analysis

Uranium, Tc, and iodine in the Cast Stone (after microwave digestion for U and Tc and fusion for iodine) and EPA Draft Method 1315 leachates were also measured at PNNL using a Perkin Elmer ELAN DRC II (Waltham, MA) ICP-MS following procedure PNNL-AGG-415, "Inductively Coupled Plasma Mass Spectrophotometry (ICP-MS) Analysis,"¹ which is similar to Test Methods for Evaluating Solid Wastes: Physical/Chemical Methods (SW-846 6020A; EPA 2007b).

B.9 Total Carbon Analysis

The total carbon content of the dry materials used to make the dry blend was determined at PNNL directly using a Shimadzu Carbon analyzer Model TOC-V CSN with a SSM-5000A following procedure AGG-TOC-001 Operating of Carbon Analyzer² (TOC-V + SSM-5000A + ASI (Shimadzu)), which is similar to ASTM Method E1915-01 *Standard Test Methods for Analysis of*

¹ Clayton E. 2008. "Inductively Coupled Plasma Mass Spectrometric (ICP-MS) Analysis." PNNL-AGG-415 (Rev. 2), unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

² Kutnyakov IK. 2004. "Operating of Carbon Analyzer (TOC-V + SSM-500A +ASI (Shimadzu))." AGG-TOC-001 (Rev. 0), unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

Metal Bearing Ores and Related Materials by Combustion Infrared Absorption Spectrometry (ASTM E1915 2001). The sample is combusted at 900°C with an oxidative catalyst. The carbon is converted to CO₂ and measured using a non-dispersive infrared gas analyzer.

B.10 Powder X-ray Diffraction

At PNNL, X-ray diffraction was used to characterize a portion of the dry material. X-ray diffractograms were collected using a Rigaku Ultima IV X-ray diffractometer with Cu-K α radiation X-ray tube and a graphite monochromator. Data were collected in the 2 θ range: 2–65° with a scanning step size of 0.05° 2 θ and a 4-second dwell time. The data were analyzed with the computer program JADE (MDI, Livermore, CA combined with the Joint Committee on Powder Diffraction Standards, International Center for Diffraction Data (ICDD) Newtown Square, PA database).

At SRNL, samples were ground in an agate mortar and pestle to reduce the particle size and to homogenize the samples. The ground powder was placed on a glass slide. A few drops of a 10% Amyl Acetate/Collodion solution was added to the ground powder to fix the powder to the glass slide.

At SRNL, X-ray diffraction data were collected on a Bruker D8 X-ray diffractometer by step scanning over the 2 θ ranges of 5–70° with a step size of 0.02° and a dwell time of 1 second. All the instrument parameters are listed in the Table B-5 below. Search-match identification of all the phases was performed using Jade software (Version 9.0) from Materials Data Inc. combined with the ICDD's PDF-4 database.

Table B-5. Instrument Parameters

Radiation Source	CuK α X-ray
Source Power	45 kV, 40 mA
Wavelength	1.5405982 Å
Goniometer	Bruker D8
Divergence Slit	1°
Divergence Soller Slit	None
Divergence Antiscatter	1°
Specimen Rotation	No
Diffracted Beam Antiscatter	1°
Diffracted Beam Soller Slit	2°
Secondary Monochromator	Curved pyrolytic graphite
Receiving Slit	0.15°
Detector	NaI Scintillation
2 θ Range	5°–70°
Step Interval	0.02° (2 θ)
Fixed Counting Time	1 s/step

B.11 Surface Area

The BET method of surface area measurement is based on the phenomenon of physical adsorption. At liquid nitrogen temperatures, nitrogen gas will adsorb to the surface of a solid due to physical forces of attraction between the adsorbent (solid) and the adsorbate (gas). These forces, which are essentially electrostatic in nature, are often called Van der Waals forces. The

quantity, or number of moles, of gas adsorbed at some equilibrium point is a function of the equilibrium vapor pressure of nitrogen in the system. The so-called BET method (1938) quantifies the amount of gas adsorbed at various equilibrium pressures in terms of the deposition of a monomolecular layer of gas followed by successive additional layers as the equilibrium pressure is progressively increased upwards to saturation—approximately 760 torr for nitrogen at liquid nitrogen temperature. Adsorption isotherms are measured as the quantity (moles) of gas adsorbed versus relative pressure, defined as the equilibrium pressure divided by the saturated vapor pressure of nitrogen.

During a surface area measurement, the sample is heated under vacuum to remove volatiles, usually water, from the material. The sample tube containing the sample is then pumped to high vacuum and the free volume of the sample/sample tube system is measured with helium. The sample is again evacuated to high vacuum and the saturated vapor pressure of nitrogen is measured in a separate tube before the adsorption measurement begins.

An adsorption isotherm is measured as follows. Nitrogen gas is dosed into the initially evacuated sample tube in measured quantities. At equilibrium, the amount adsorbed is simply the total quantity of gas that has been dosed into the system minus the quantity remaining in the free volume. This process is repeated for a number of equilibrium points from about 0.05 relative pressure up to 0.3. The data are plotted according to the BET equation, and $1/(\text{slope} + \text{intercept})$ gives the number of moles of gas in a monolayer. The total surface area is then calculated using 16.2 Angstroms squared as the area occupied by one nitrogen molecule, and the specific surface area in meters squared per gram (m^2/g) is obtained by dividing the total area by the sample mass.

B.12 Particle Size Distribution

The Microtrac X-100 particle size analyzer uses a wet sample delivery controller (recirculator) to disperse the sample uniformly in a fluid and deliver the sample to the analyzer. This wet sample delivery controller in its basic form consists of a reservoir where the sample is introduced, a fluid pump, a valve to the drain system, and the necessary tubing connections to the analyzer. The flow through the analyzer sample cell is always from the bottom to the top. A laser beam is projected through a transparent quartz cell containing a stream of moving particles suspended in DIW. Light from the laser strikes the particles and is scattered through various angles. The scattering angles and intensities of the scattered light are measured by two photodiode arrays producing electronic signals proportional to the measured light flux. The Microtrac proprietary mathematical software processes the signals to obtain a particle size distribution. Upon completion of the analysis, the Microtrac generates a report containing the tabular data, a histogram plot of the data, and various instrument parameters.

B.13 Particle Density

The density of the dry blend particles was measured using a Quantachrome Multipycnometer (SRNL Manual L29, ITS-0168). The density is determined by measuring the pressure difference when a known quantity of helium under pressure is allowed to flow from a known volume into a sample cell of known volume to which the solid or powdered material has been added. The density is then calculated by dividing the mass of the sample analyzed by the measured volume.

B.14 Fill and Tap Density

A Quantachrome Autotap was used to measure the tap density of dry blend components. Samples were placed in 250-ml graduated cylinders and mounted on a universal tap platform. After noting the initial volume and weight of the material, the sample was tapped 300 times. When

the tapping was completed the powder volume was read. Automatic rotation of cylinders during tapping promoted a flat powder interface.

B.15 Simultaneous Thermal Analysis

A Netzsch STA 409 Luxx, which couples differential scanning calorimetry (DSC) with thermal gravimetric analysis (TGA), was used at SRNL for determining the thermal response of the dry blend components. After loading the sample, the chamber was purged with nitrogen at 60 ml/min prior to heating. Samples were heated at 5°C/min up to 650°C in a flowing nitrogen atmosphere of 60 ml/min.

Appendix C. Cast Stone Processing Properties Data

This appendix contains data related to Cast Stone flow properties (flow diameter), gel time, slurry/paste rheology properties (yield stress and plastic viscosity), heat-of-hydration properties (heat generation at 300 hours, time to peak heat generation, and heat generation at peak), and bleed water measurements. All of these property measurements were made at Savannah River National Laboratory (SRNL) using samples of the Cast Stone mixes prepared by SRNL. However, 12 of the original 26 mixes in Table 2-1 were mistakenly made with the wrong source of blast furnace slag (BFS), as shown in Table 6-1. Additional samples of the 12 originally intended mixes (i.e., with the intended sources of BFS) were made by SRNL and all processing properties were measured for these 12 mixes. Hence, for all processing properties, there were 38 Cast Stone formulations prepared and property values measured. Table C-1 lists the parameter combinations for the 38 Cast Stone mixes. Table C-2 shows these mixes in a matrix display.

Table C-1. Test Matrix Used by SRNL for Cast Stone Processing and Curing Properties

Mix Number ^(a)	Random Run Order ^(b)	Block	Simulant	Sodium Molarity	Fly Ash Class F	Blast Furnace Slag	Dry Blend Mix ^(c)	Water-to-Dry Blend Solids Ratio
1	7	1	High SO ₄	5.0	SE Low Ca	NW	8:45:47	0.40
2	@	11	Average	5.0	SE Low Ca	SE	8:45:47	0.60
3	\$	27	Average	7.8	NW High Ca	SE	8:45:47	0.60
4		28	High Al	5.0	NW High Ca	SE	8:45:47	0.40
5		30	Average	7.8	NW High Ca	NW	8:45:47	0.40
6		12	Average	7.8	SE Low Ca	NW	8:45:47	0.40
7	#	14	High SO ₄	7.8	SE Low Ca	SE	8:45:47	0.40
8		1	SST Blend	5.0	SE Low Ca	NW	8:45:47	0.40
9		31	Hi Al	7.8	NW High Ca	SE	8:45:47	0.60
10		32	High SO ₄	5.0	NW High Ca	NW	8:45:47	0.60
11		6	High Al	7.8	SE Low Ca	NW	8:45:47	0.60
12		29	SST Blend	5.0	NW High Ca	SE	8:45:47	0.60
13	@	4	Average	5.0	SE Low Ca	SE	8:45:47	0.60
14	#	2	High SO ₄	7.8	SE Low Ca	SE	8:45:47	0.40
15	%	33	High SO ₄	7.8	NW High Ca	NW	8:45:47	0.40
16		36	SST Blend	7.8	NW High Ca	NW	8:45:47	0.40
17		25	High Al	5.0	SE Low Ca	SE	8:45:47	0.60
18		16	SST Blend	7.8	SE Low Ca	SE	8:45:47	0.40
19		15	High Al	7.8	SE Low Ca	NW	8:45:47	0.40
20		37	Average	5.0	NW High Ca	SE	8:45:47	0.40
21	&	23	High SO ₄	7.8	SE Low Ca	NW	8:45:47	0.60
22	\$	34	Average	7.8	NW High Ca	SE	8:45:47	0.60
23		20	SST Blend	7.8	SE Low Ca	NW	8:45:47	0.60
24		38	High Al	5.0	NW High Ca	NW	8:45:47	0.60
25	%	35	High SO ₄	7.8	NW High Ca	NW	8:45:47	0.40
26	&	18	High SO ₄	7.8	SE Low Ca	NW	8:45:47	0.60
27 (3a)	^	3	Average	7.8	NW High Ca	NW	8:45:47	0.60
28 (4a)		5	High Al	5.0	NW High Ca	NW	8:45:47	0.40
29 (5a)		9	Average	7.8	NW High Ca	SE	8:45:47	0.40
30 (9a)		10	Hi Al	7.8	NW High Ca	NW	8:45:47	0.60
31 (10a)		13	High SO ₄	5.0	NW High Ca	SE	8:45:47	0.60
32 (12a)		8	SST Blend	5.0	NW High Ca	NW	8:45:47	0.60
33 (15a)	*	17	High SO ₄	7.8	NW High Ca	SE	8:45:47	0.40
34 (16a)		22	SST Blend	7.8	NW High Ca	SE	8:45:47	0.40
35 (20a)		24	Average	5.0	NW High Ca	NW	8:45:47	0.40
36 (22a)	^	19	Average	7.8	NW High Ca	NW	8:45:47	0.60
37 (24a)		26	High Al	5.0	NW High Ca	SE	8:45:47	0.60
38 (25a)	*	21	High SO ₄	7.8	NW High Ca	SE	8:45:47	0.40

(a) Mix numbers with the same symbols are replicates. Mixes 3 and 22 were inadvertent replicates, due to an accidental switch in the simulant for Test 3 (from SST Blend to Average) during the final step of developing the test matrix in Table 2-1.

(b) The run order was randomized separately within Block 1 and within Block 2. The random run order for the Block 3 tests (retests of mis-batched mixes) was the same order as used for Mixes 27–38 (3a, 4a, ..., 25a).

(c) 8 wt% cement: 45 wt% fly ash, 47 wt% blast furnace slag, where cement is Type I/II Portland cement.

Table C-2. Test Matrix for Cast Stone Processing Properties of 38 Cast Stone Mixes

Waste Composition	Fly Ash Source/Blast Furnace Slag Source (Northwest or Southeast USA)							
	NW/NW	NW/SE	SE/NW	SE/SE	NW/NW	NW/SE	SE/NW	SE/SE
Average 5M	35 (20a)	20						13 2
High SO ₄ 5M			1		10	31 (10a)		
High Al 5M	28 (4a)	4			24	37 (24a)		17
SST Blend 5M			8		32 (12a)	12		
Average 7.8M	5	29 (5a)	6		27 (3a) 36 (22a)	3 22		
High SO ₄ 7.8M	15 25	33 (15a) 38 (25a)		14 7			26 21	
High Al 7.8M			19		30 (9a)	9	11	
SST Blend 7.8M	16	34 (16a)		18			23	
Mix Ratio (w/dry mix)	0.4	0.4	0.4	0.4	0.6	0.6	0.6	0.6

Original Mix No.

Mix No. (intended Mix)

Test Combination

Replicate

C.1 Average Flow Diameters

Table C-3 lists the average flow diameters for the 38 Cast Stone mixes.

Table C-3. Average Flow Diameter of 38 Cast Stone Mixes

Mix Number ^(a)	Random Run Number	Average Flow Diameter (mm) ^(b)	Water-to-Dry-Blend Ratio
1	7	93.5	0.40
2 @	11	228.1	0.60
3 \$	27	260.7	0.60
4	28	131.8	0.40
5	30	125.1	0.40
6	12	119.6	0.40
7 #	14	138.9	0.40
8	1	108.0	0.40
9	31	255.5	0.60
10	32	238.0	0.60
11	6	230.8	0.60
12	29	298.3	0.60
13 @	4	203.5	0.60
14 #	2	139.9	0.40
15 %	33	160.4	0.40
16	36	156.5	0.40
17	25	248.6	0.60
18	16	127.3	0.40
19	15	137.7	0.40
20	37	121.2	0.40
21 &	23	240.4	0.60
22 \$	34	255.5	0.60
23	20	193.3	0.60
24	38	245.2	0.60
25 %	35	158.6	0.40
26 &	18	232.8	0.60
27 (3a) ^	3	234.0	0.60
28 (4a)	5	109.0	0.40
29 (5a)	9	158.7	0.40
30 (9a)	10	248.9	0.60
31 (10a)	13	229.4	0.60
32 (12a)	8	242.0	0.60
33 (15a) *	17	163.7	0.40
34 (16a)	22	140.6	0.40
35 (20a)	24	111.5	0.40
36 (22a) ^	19	203.4	0.60
37 (24a)	26	234.0	0.60
38 (25a) *	21	165.0	0.40
(a) Test numbers with the same symbols are replicates.			
(b) Values were rounded to the number of decimal places shown.			

C.2 Gel Times

Table C-4 lists the measured gel times for the 38 Cast Stone mixes.

Table C-4. Gel Times and Water-to-Dry-Blend Ratios for 38 Cast Stone Mixes

Mix Number ^(a)	Random Run Number	Gel Time (min) ^(b)	Water-to-Dry-Blend Ratio
1	7	1	0.40
2 @	11	20	0.60
3 \$	27	55	0.60
4	28	2	0.40
5	30	2	0.40
6	12	6.5	0.40
7 #	14	15	0.40
8	1	15	0.40
9	31	60	0.60
10	32	13	0.60
11	6	38	0.60
12	29	15	0.60
13 @	4	20	0.60
14 #	2	15	0.40
15 %	33	6	0.40
16	36	1	0.40
17	25	35	0.60
18	16	7	0.40
19	15	10	0.40
20	37	1	0.40
21 &	23	31.5	0.60
22 \$	34	> 26 ^(c)	0.60
23	20	121	0.60
24	38	12	0.60
25 %	35	3	0.40
26 &	18	30	0.60
27 (3a) ^	3	35	0.60
28 (4a)	5	2	0.40
29 (5a)	9	10	0.40
30 (9a)	10	45	0.60
31 (10a)	13	10	0.60
32 (12a)	8	115	0.60
33 (15a) *	17	9	0.40
34 (16a)	22	5	0.40
35 (20a)	24	1	0.40
36 (22a) ^	19	22	0.60
37 (24a)	26	12	0.60
38 (25a) *	21	9	0.40
(a) Test numbers with the same symbols are replicates.			
(b) Values were rounded to the number of decimal places shown.			
(c) There was insufficient material to complete the test.			

C.3 Set or Hardening Time

Figure C-1 displays plots of the ultrasonic pulse velocity (UPV) measurements described in Section 6.3. Also on the plots are the Vicat results when it was noted that the sonic velocity was increasing. The two points plotted show the penetration of the needle fully extending to 35 mm and then not penetrating, indicating “set.” See Section 6.3 for further discussion of these data and how they were generated.

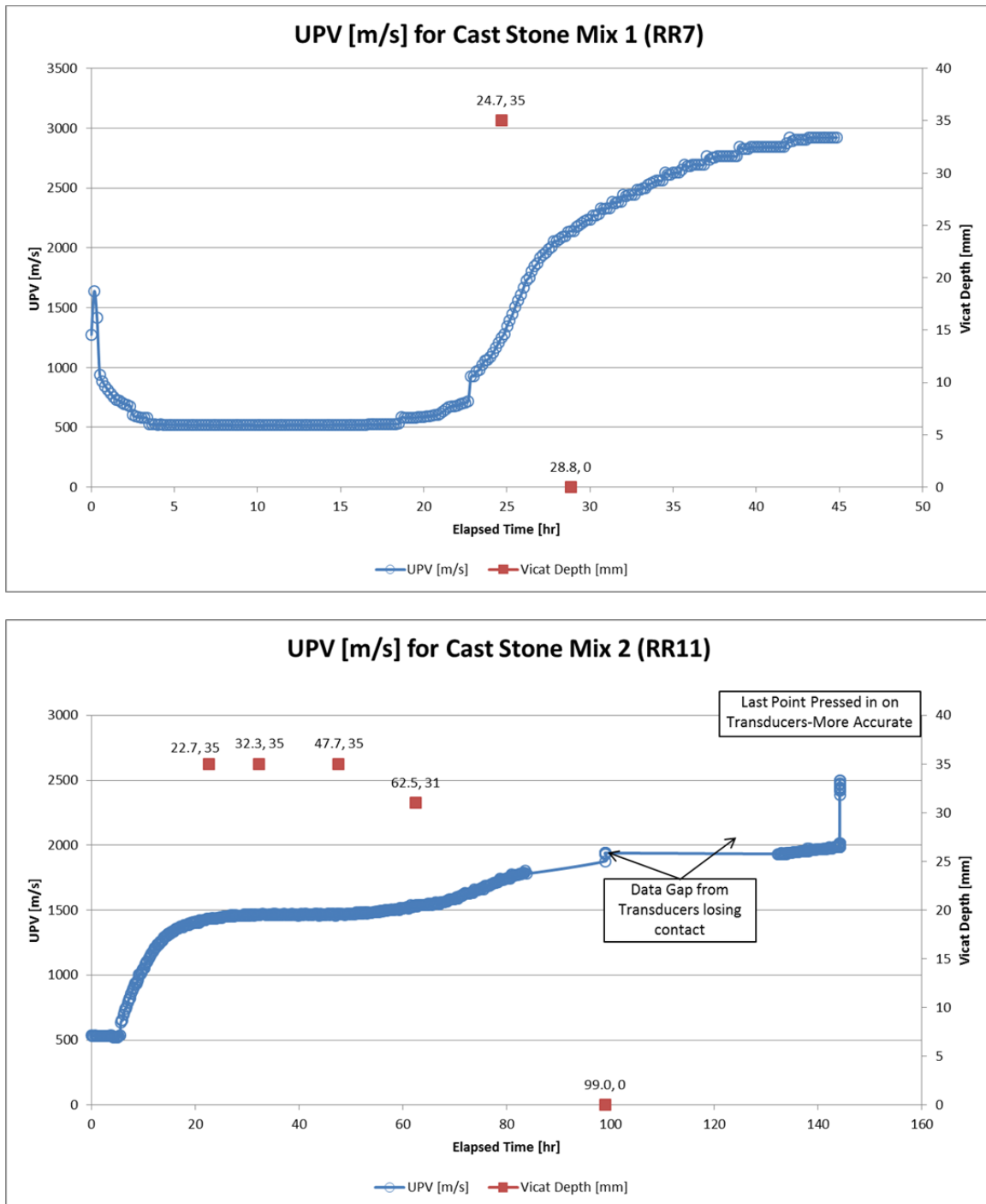


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time

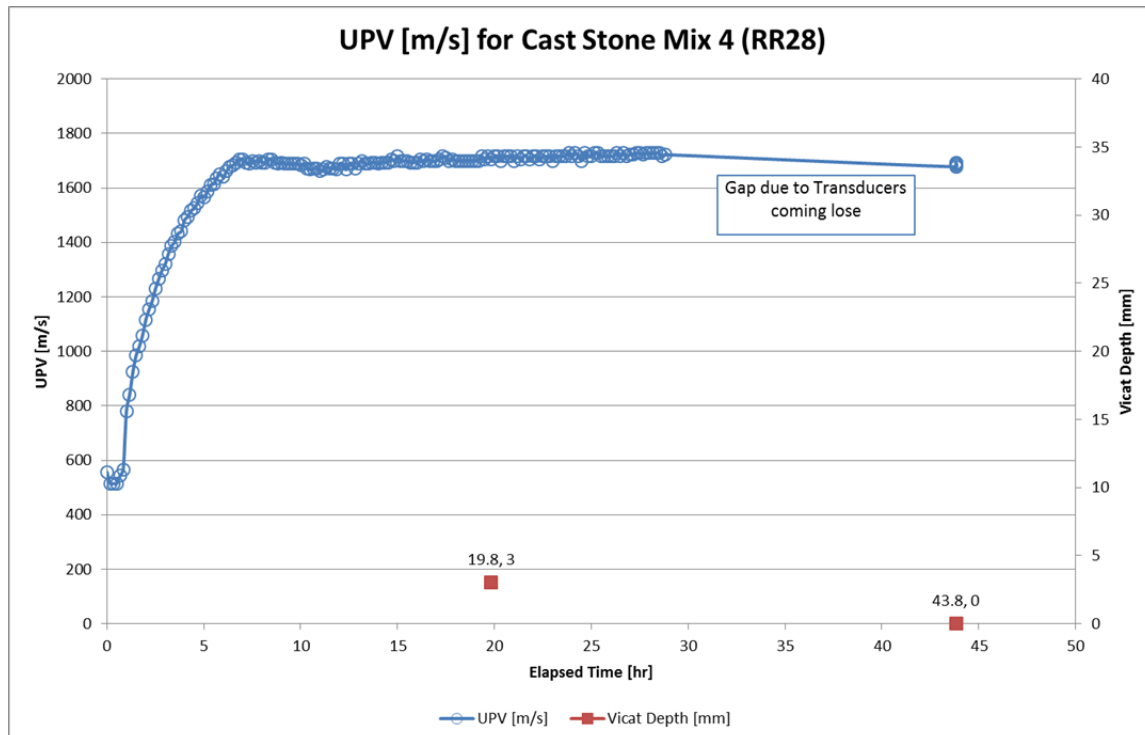
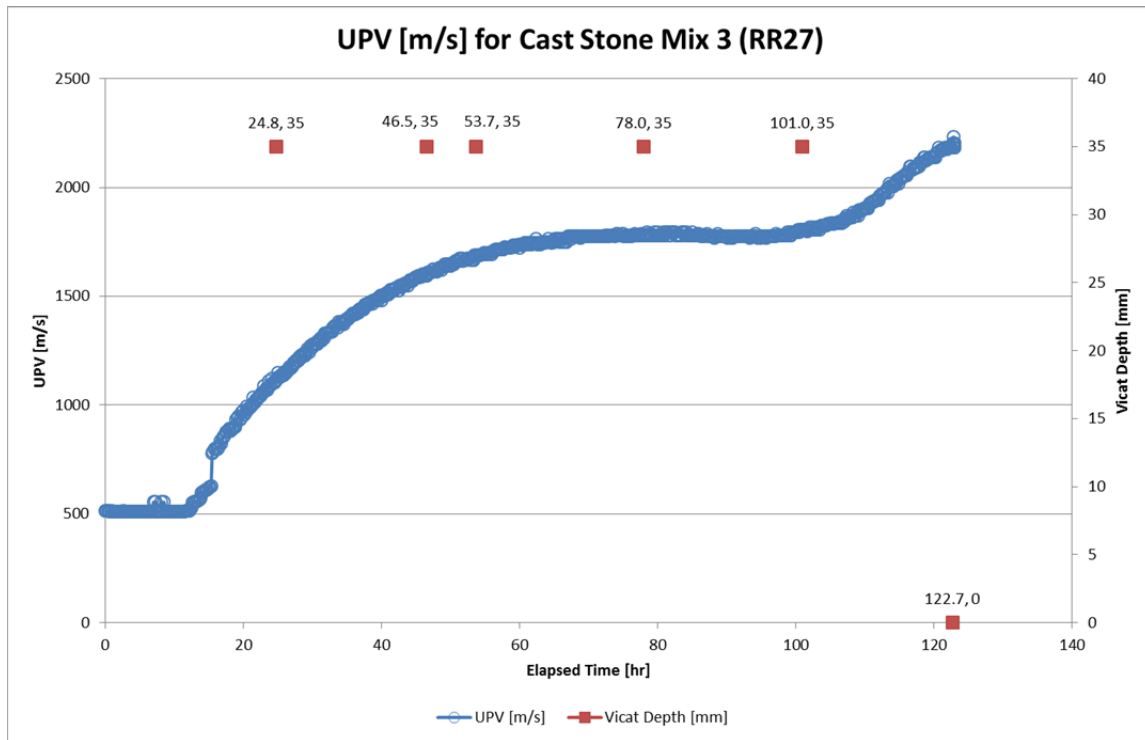


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

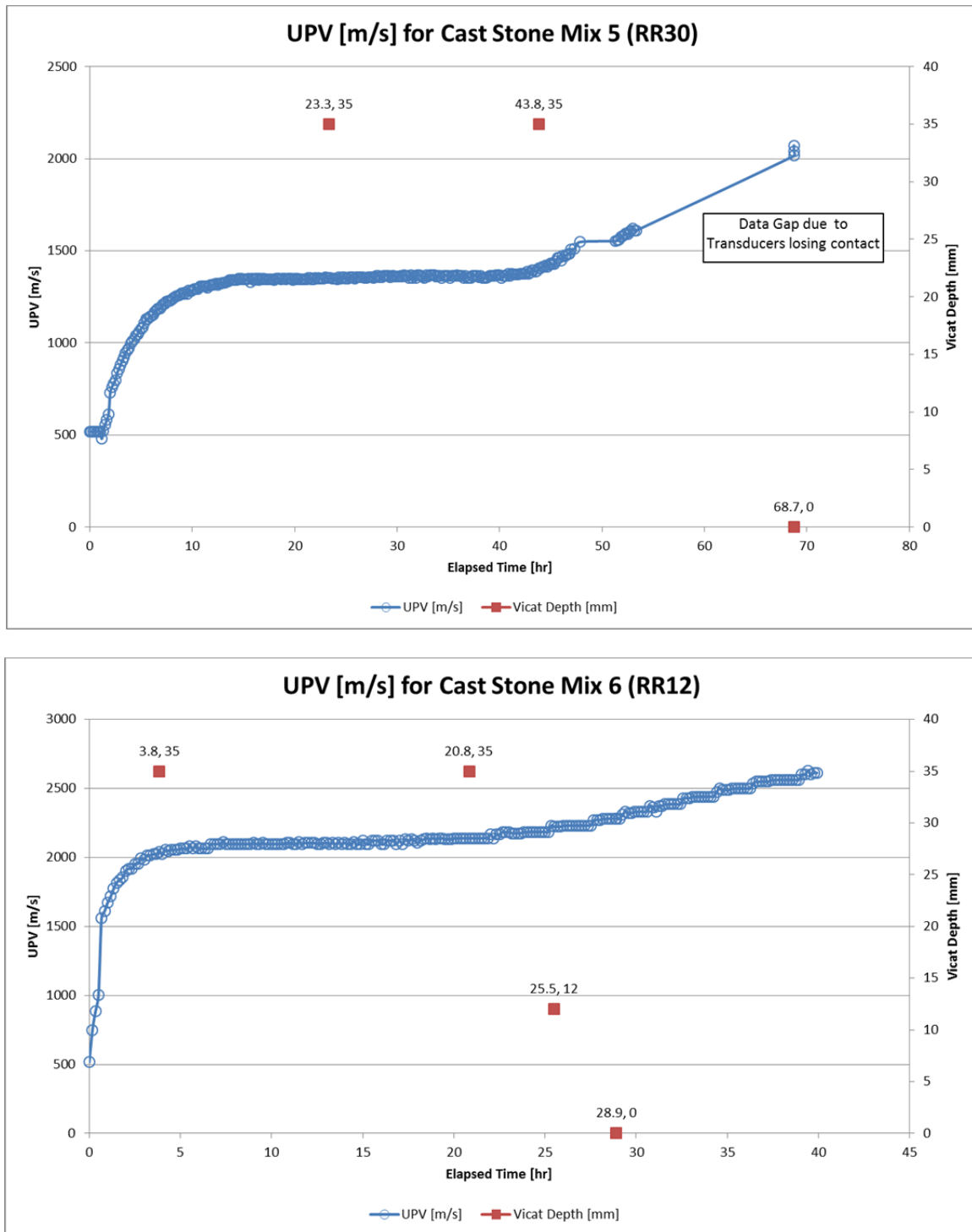


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

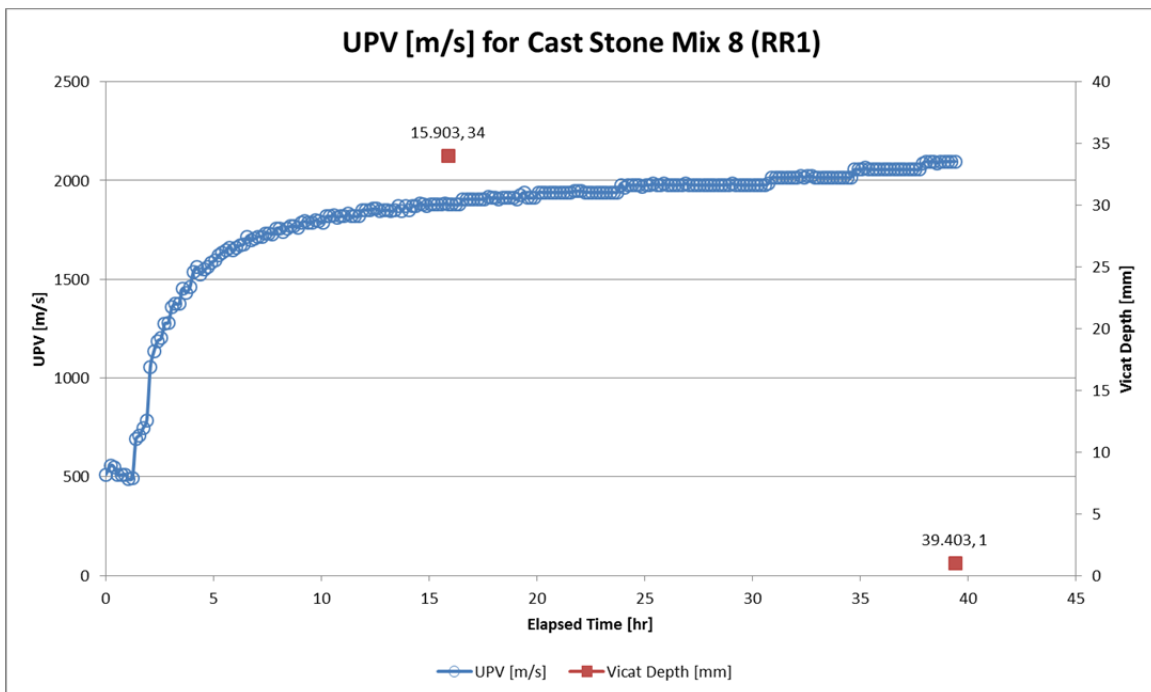
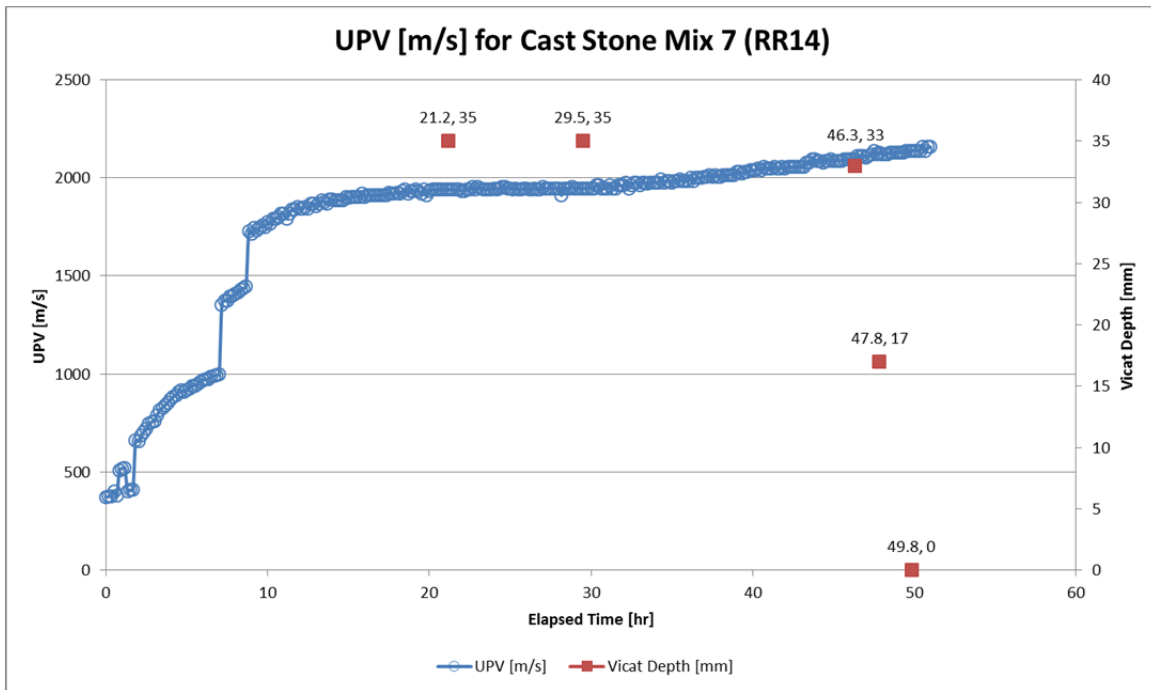


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

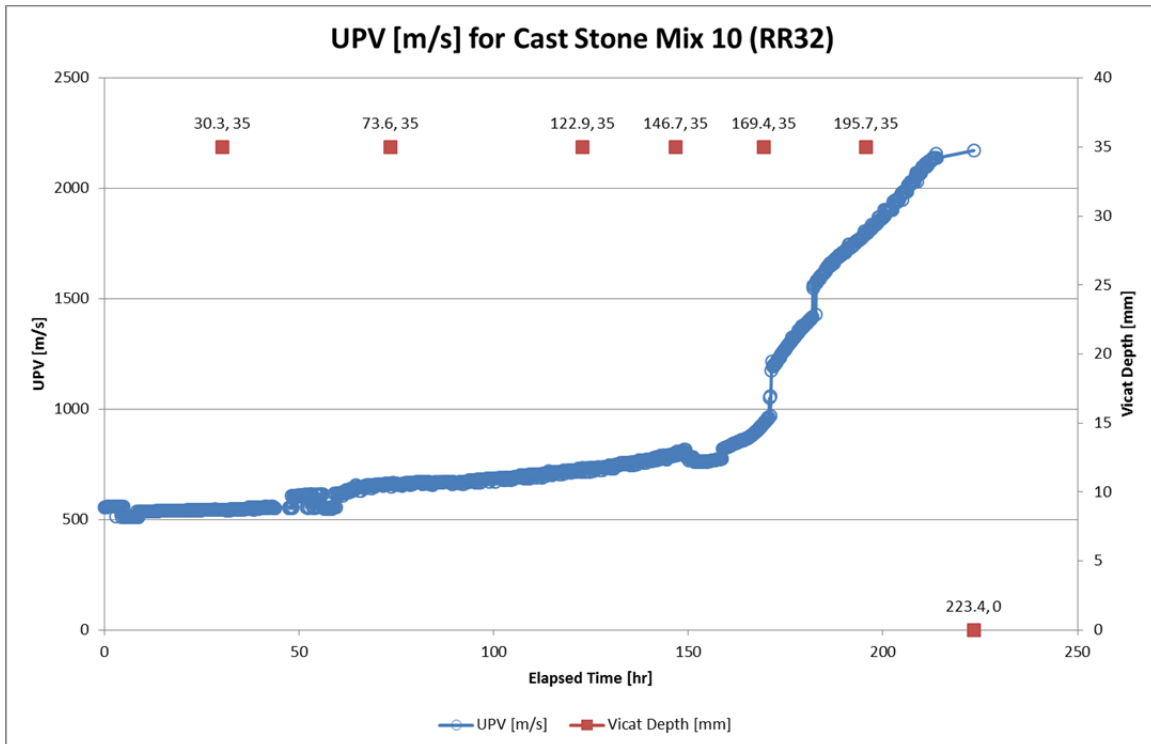
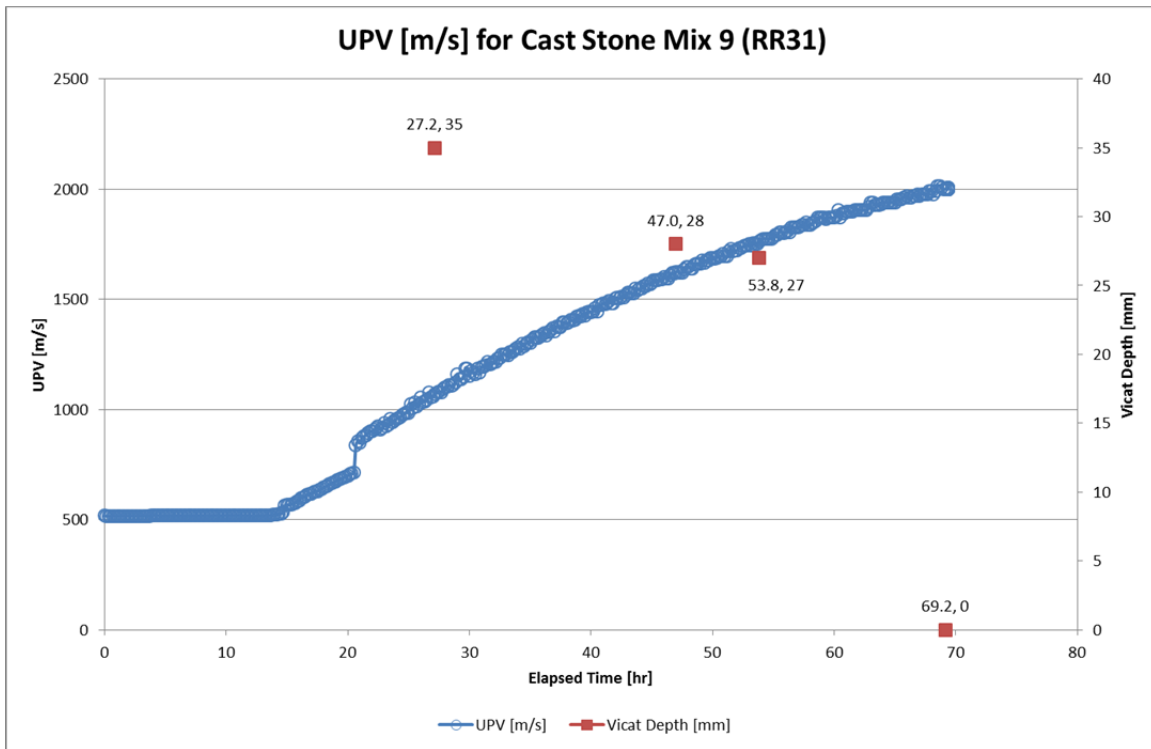


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

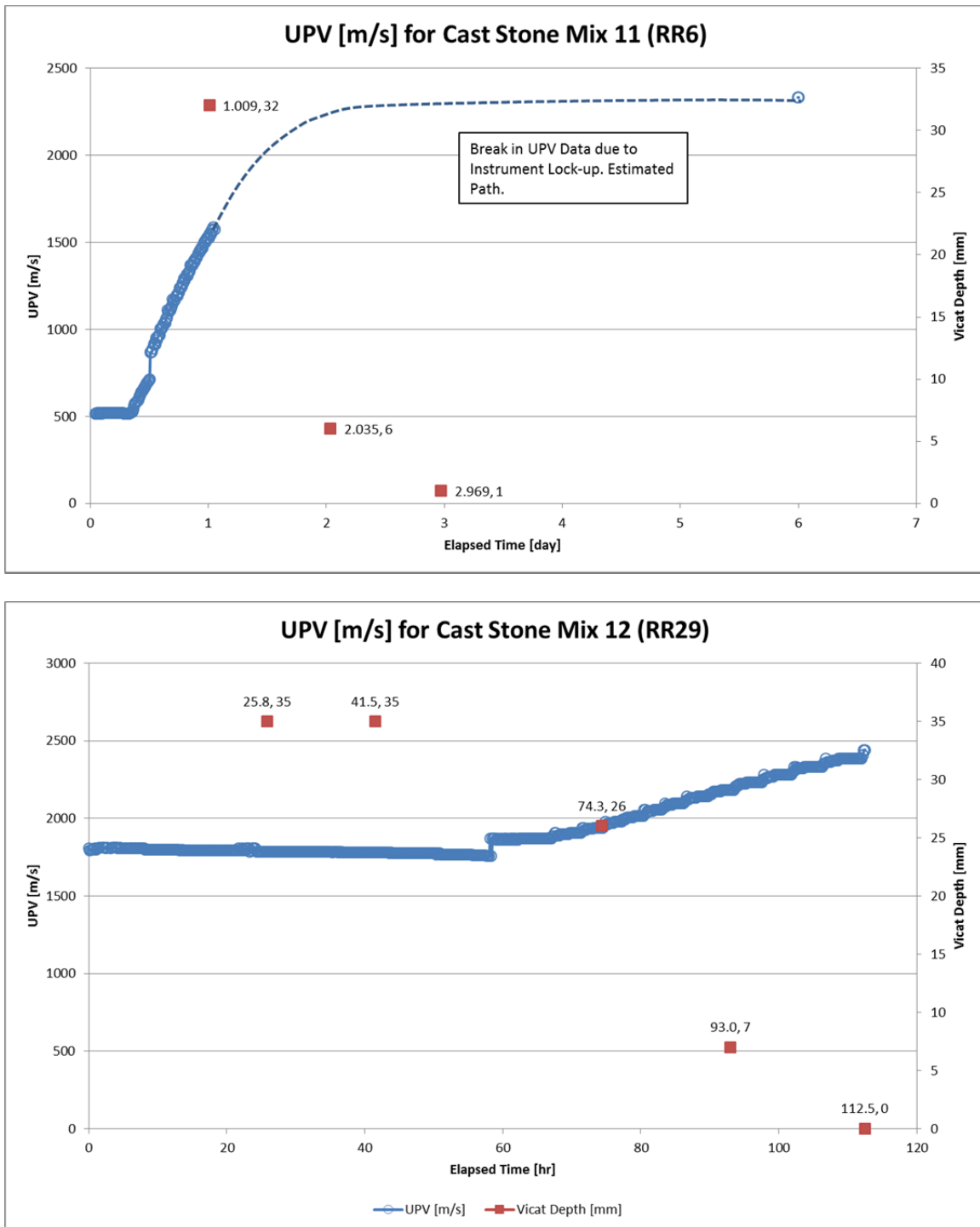


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

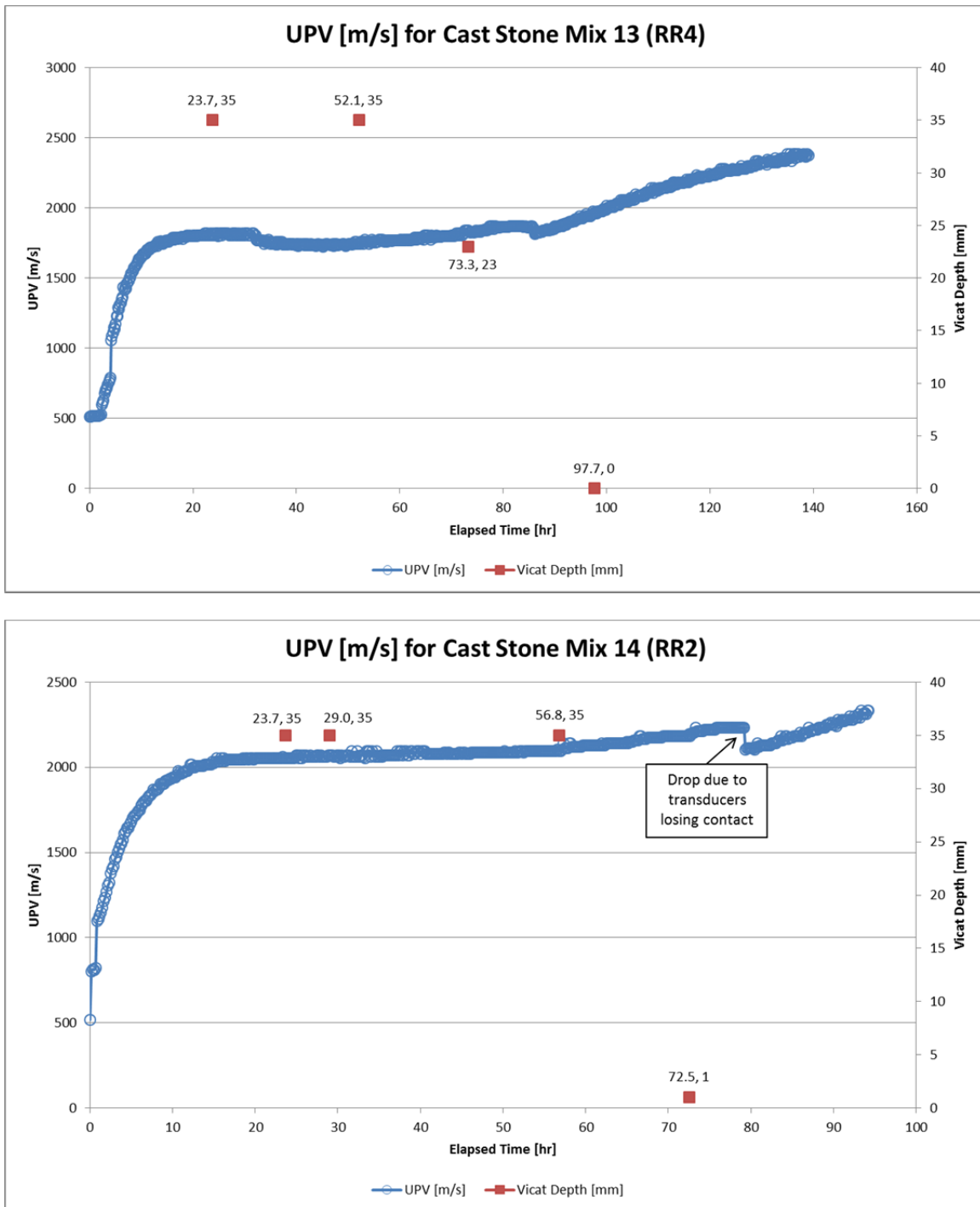


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

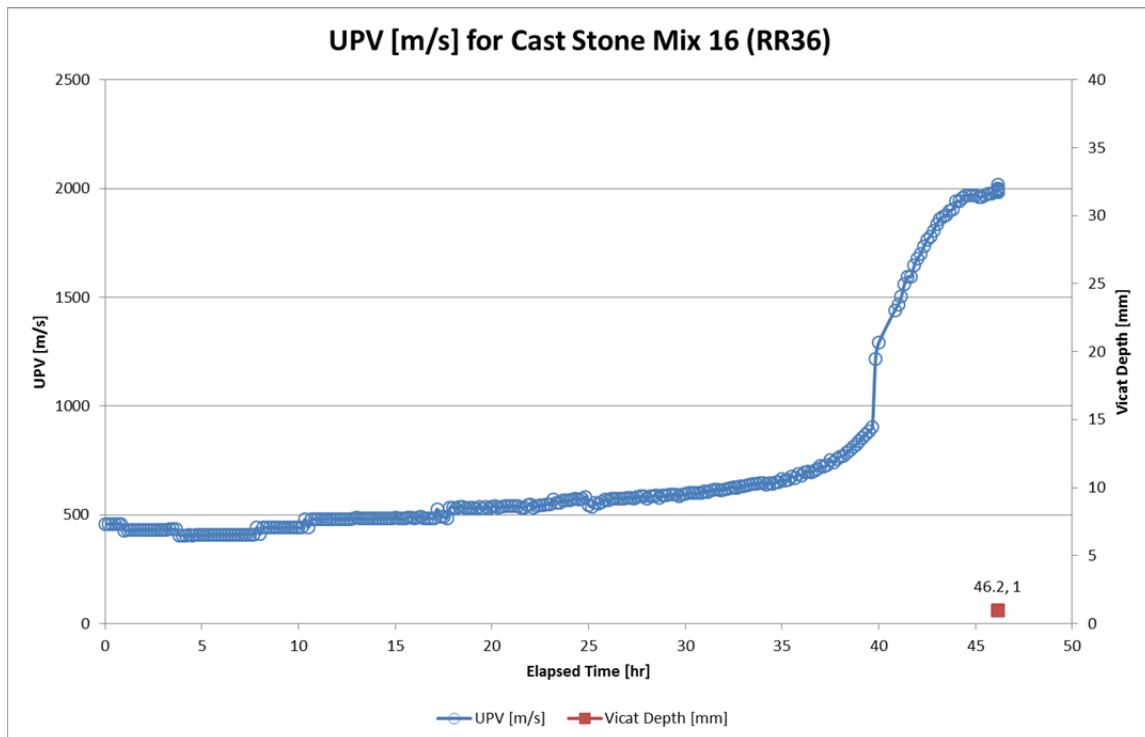
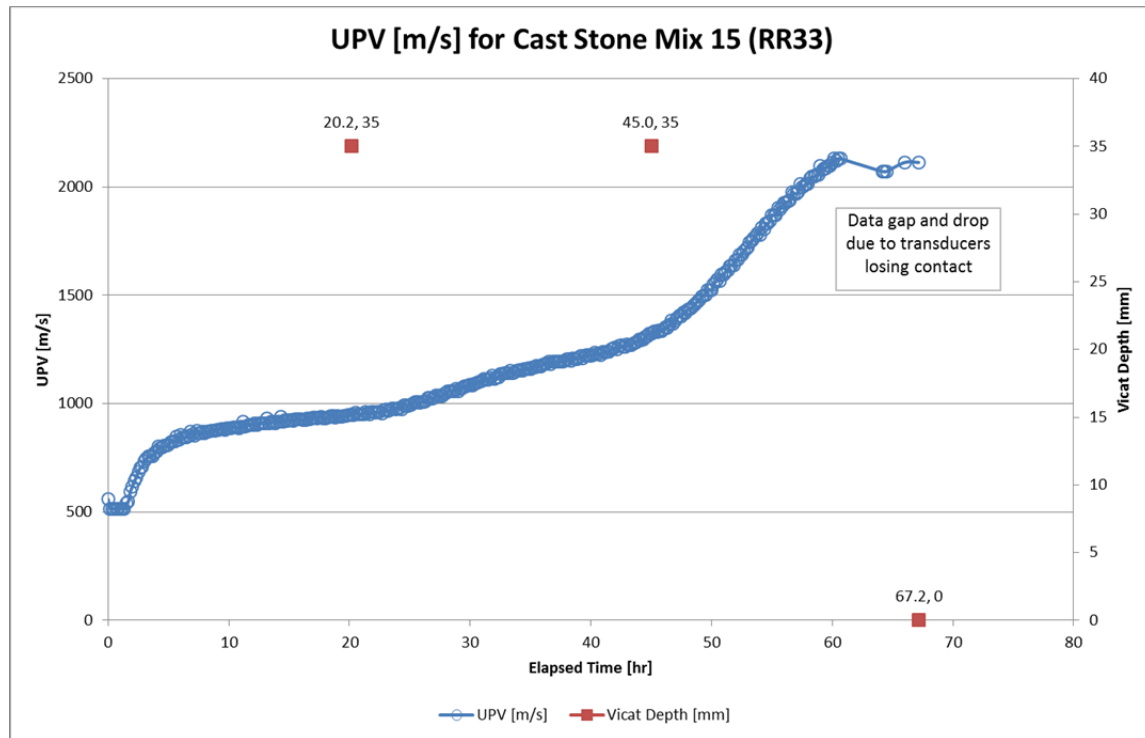


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

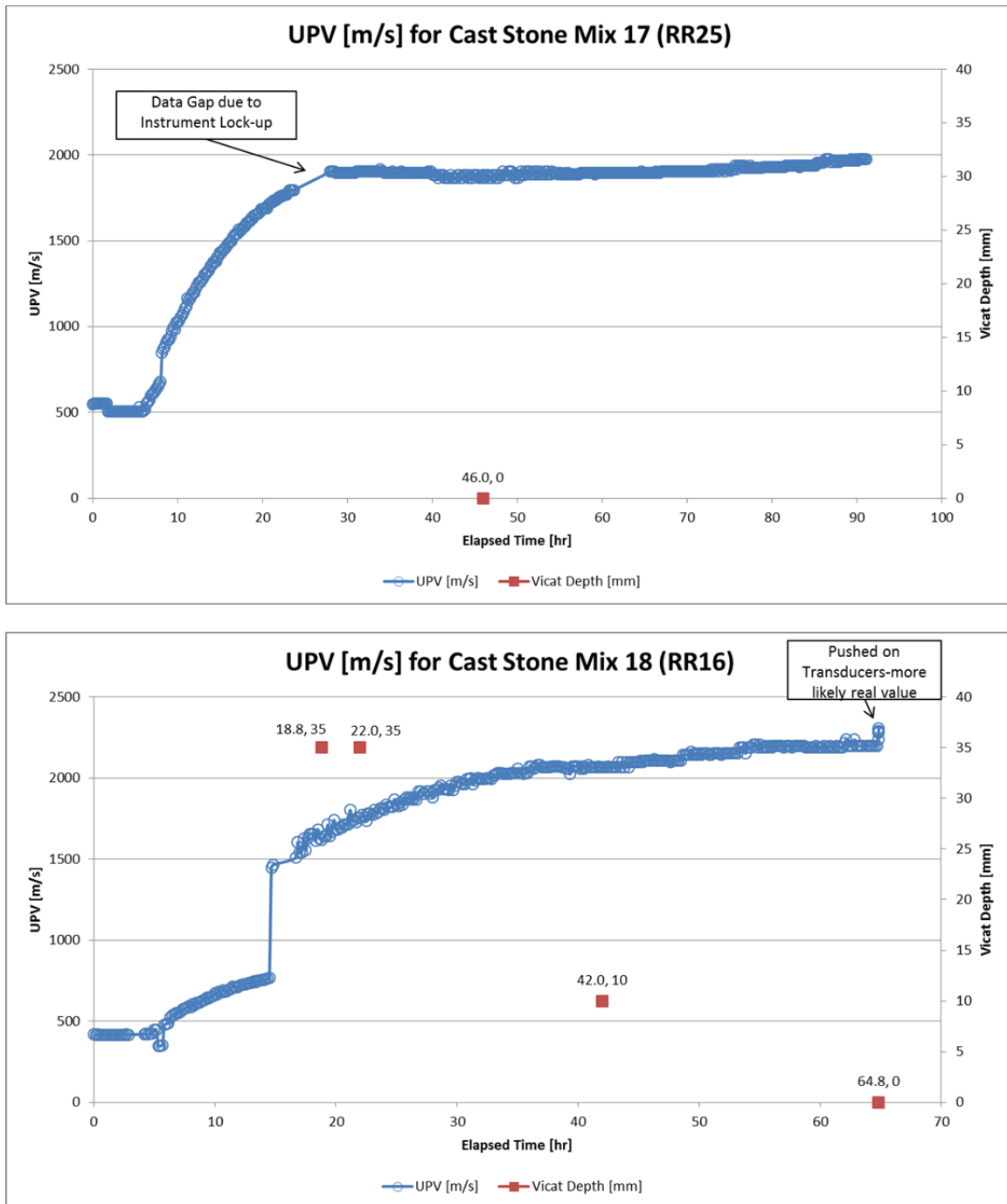


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

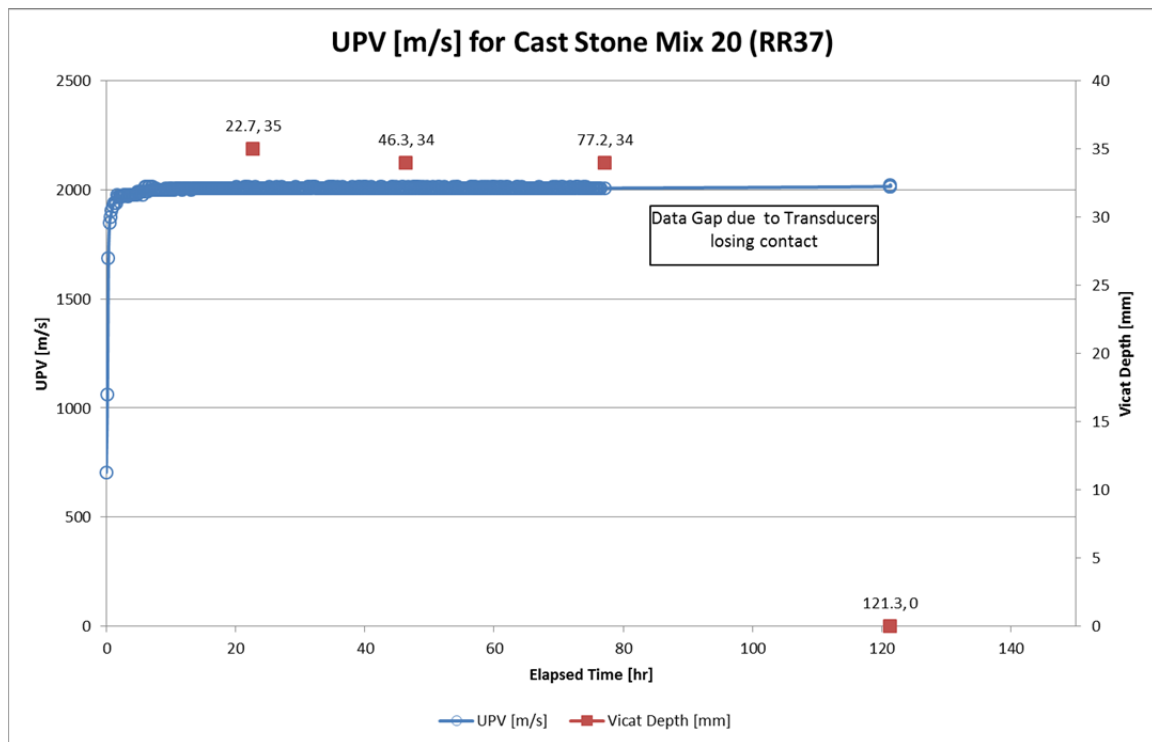
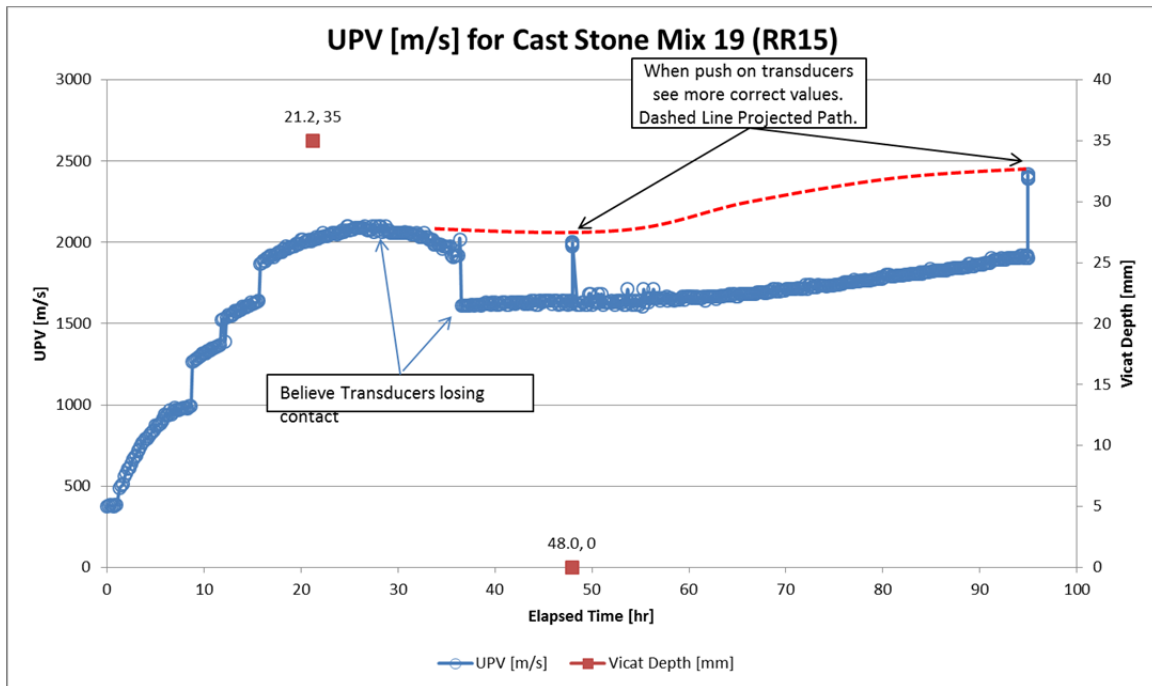


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

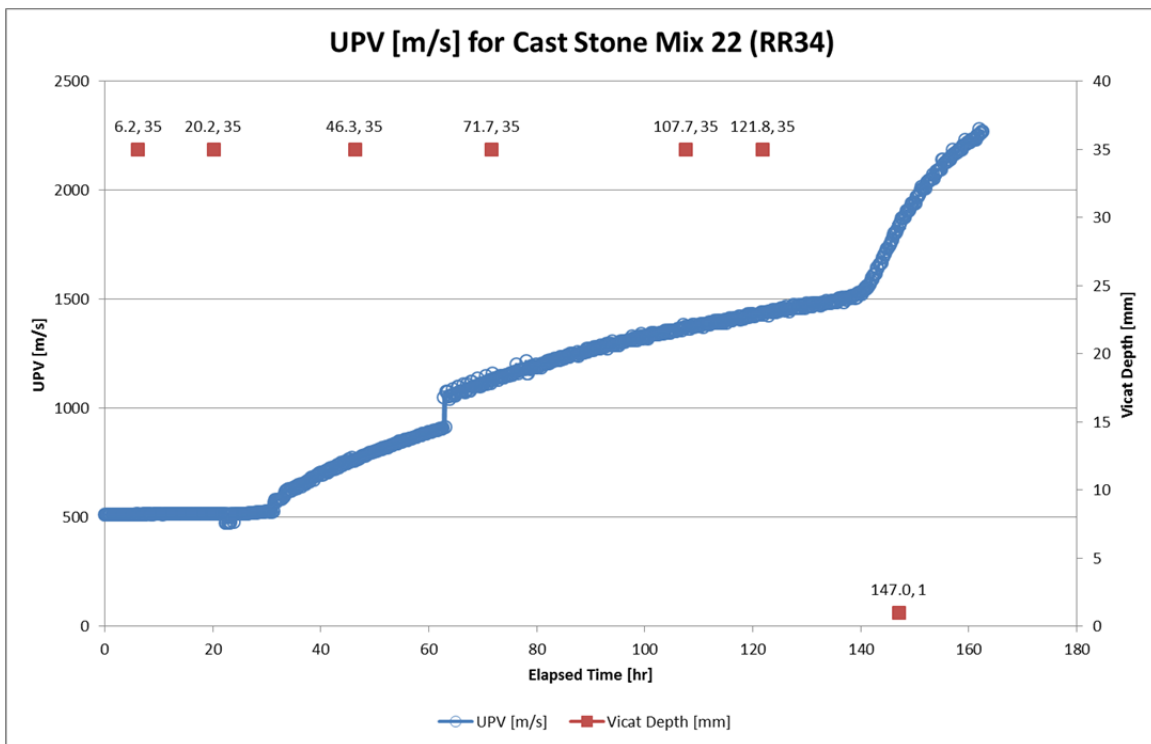
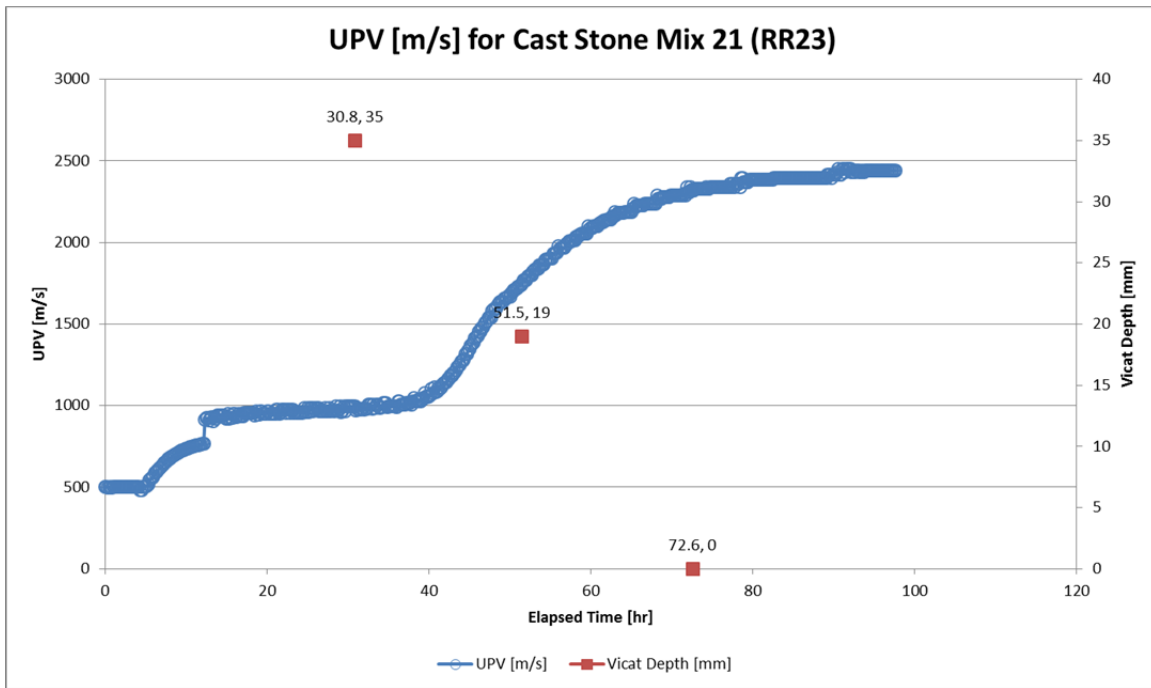


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

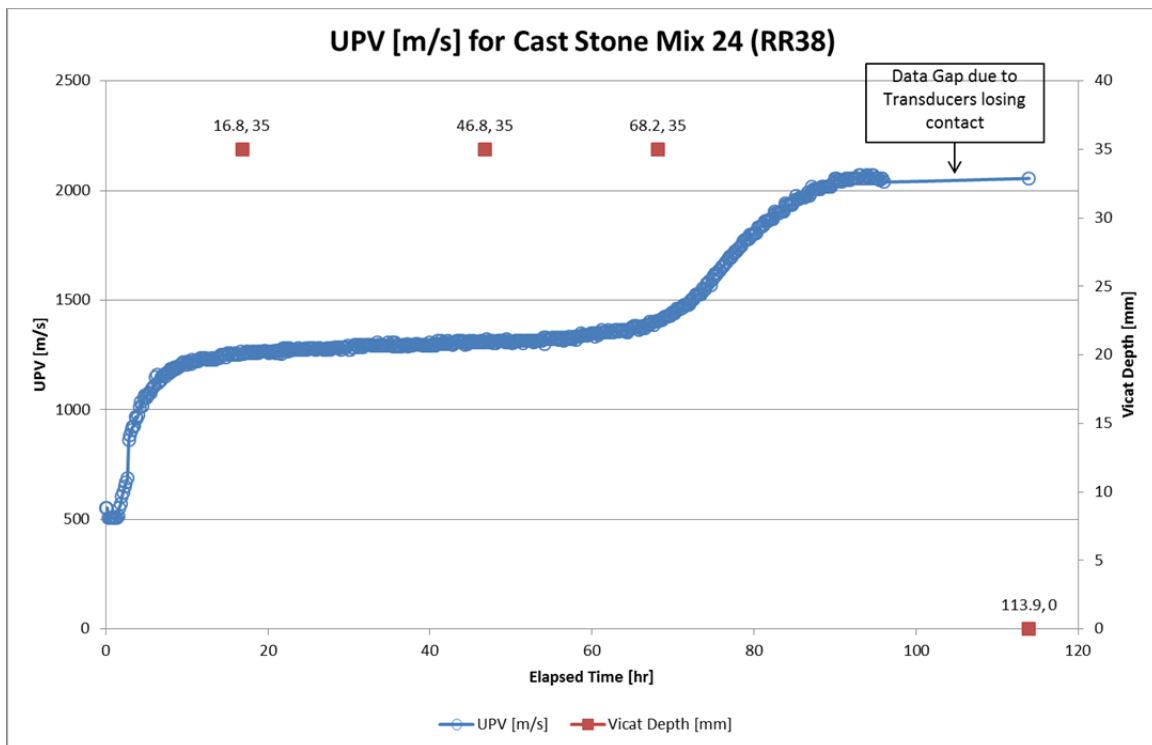
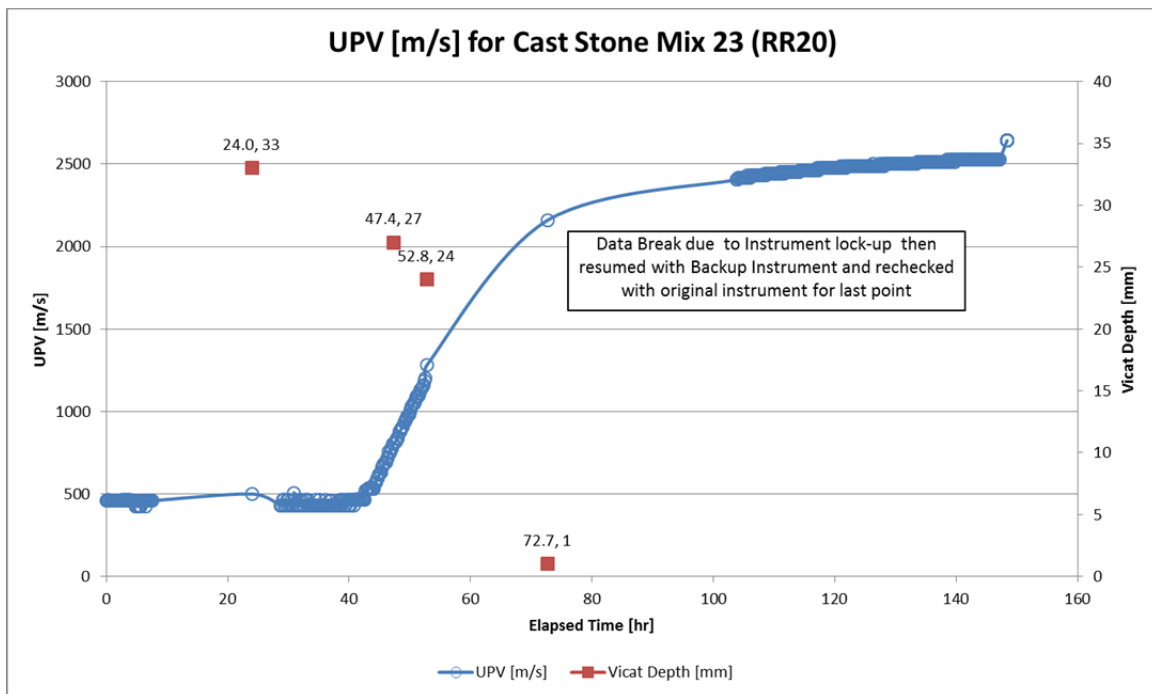


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

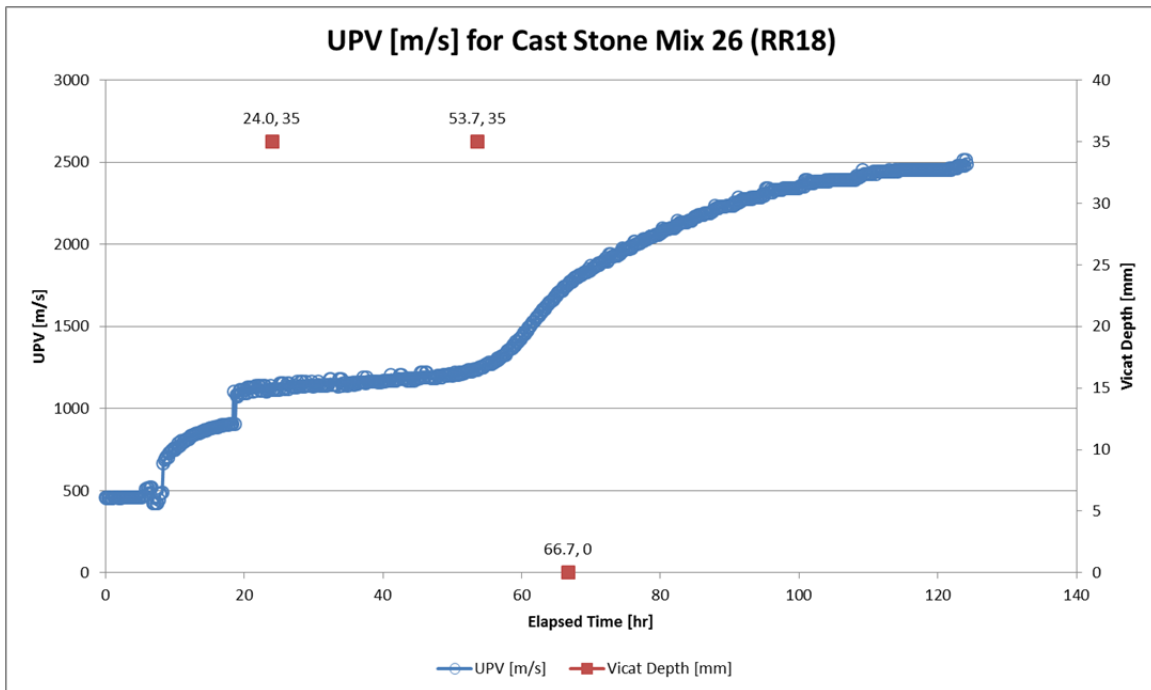
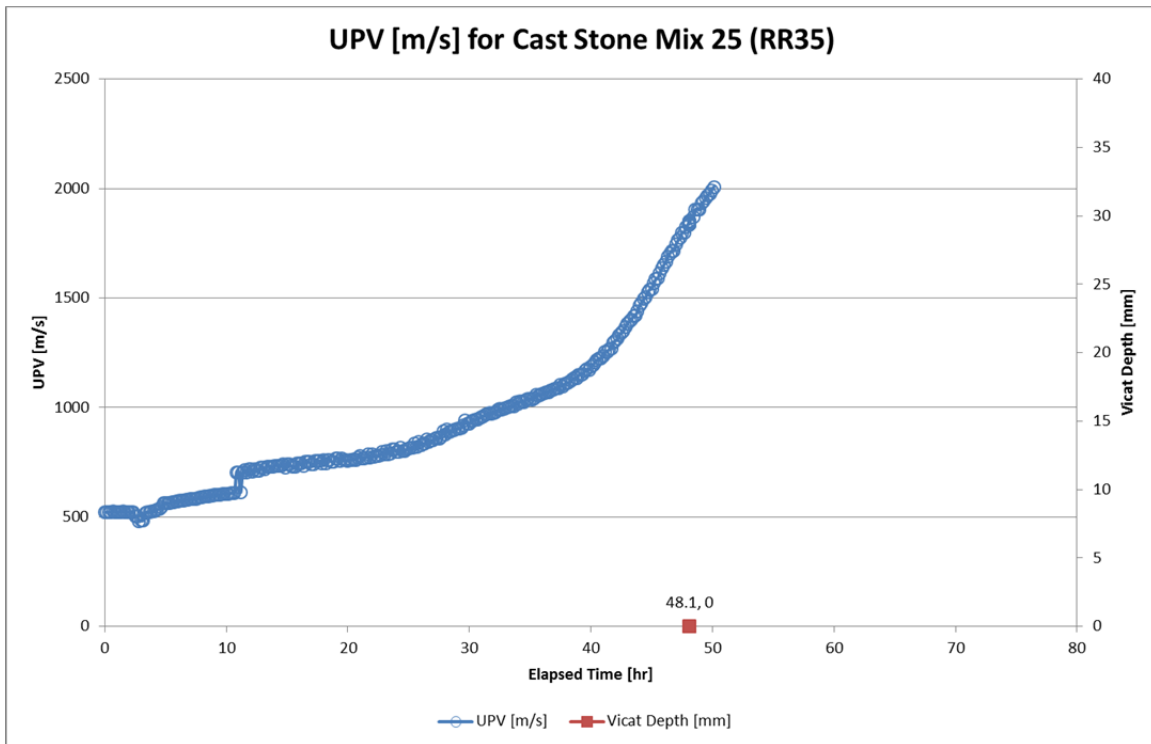


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

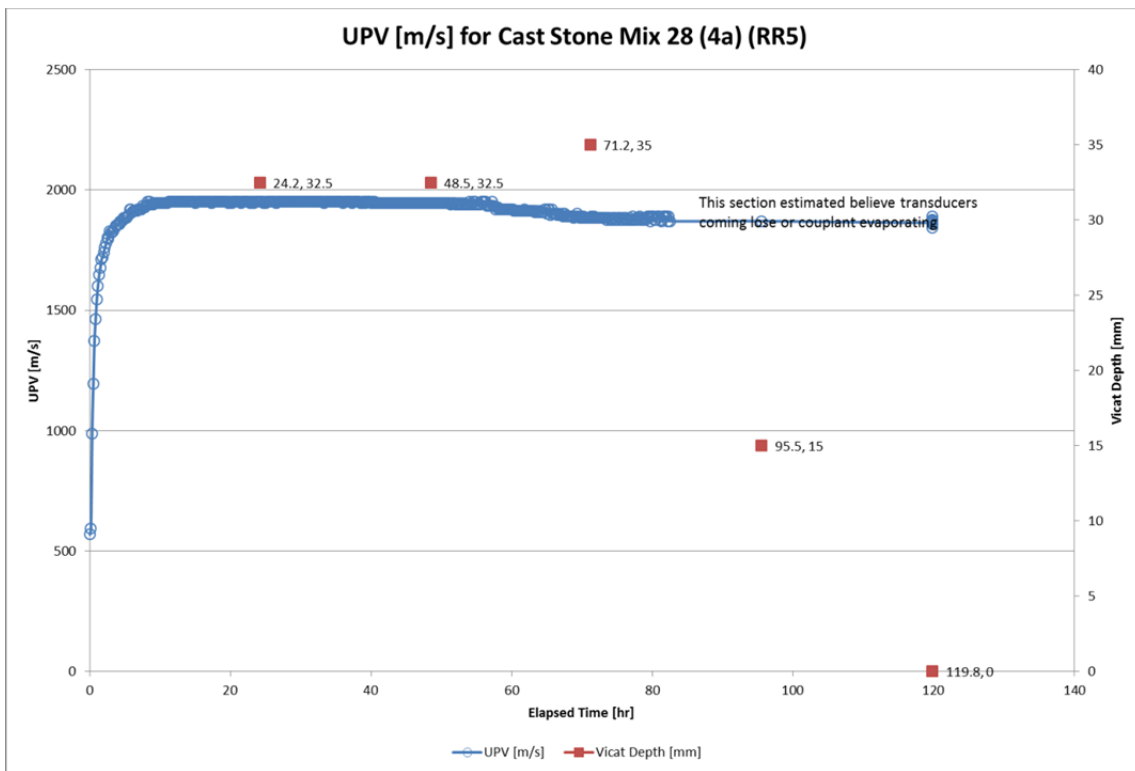
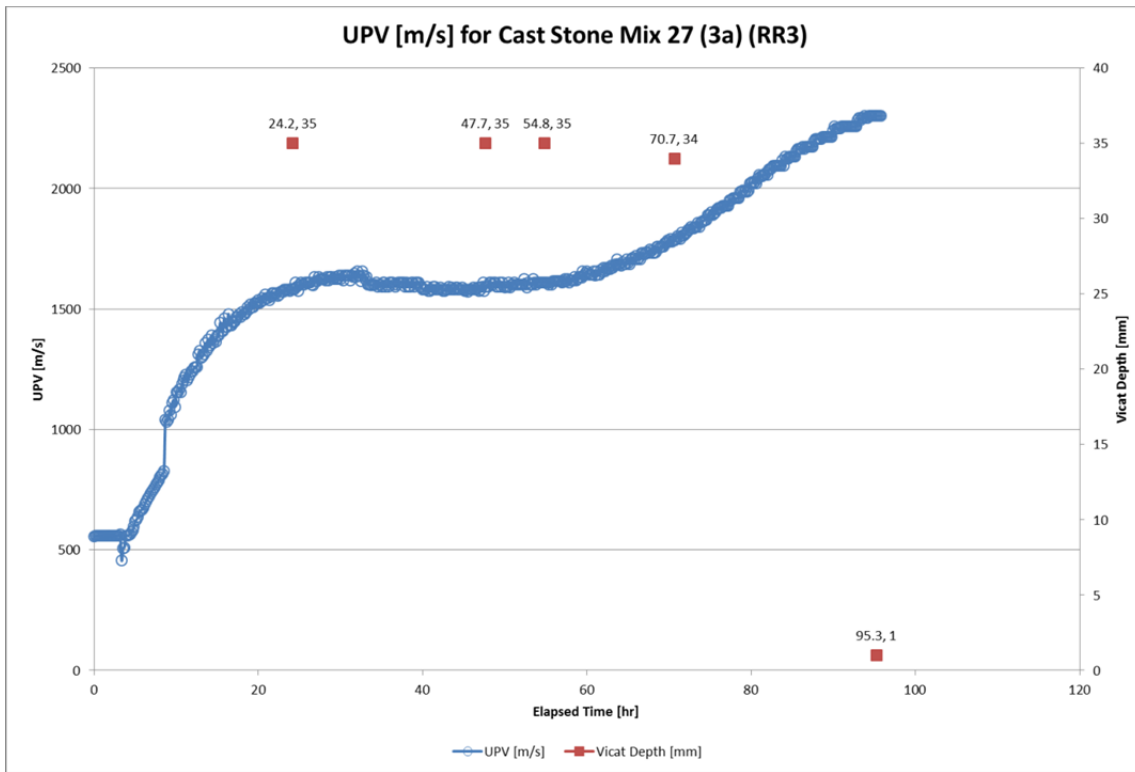


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

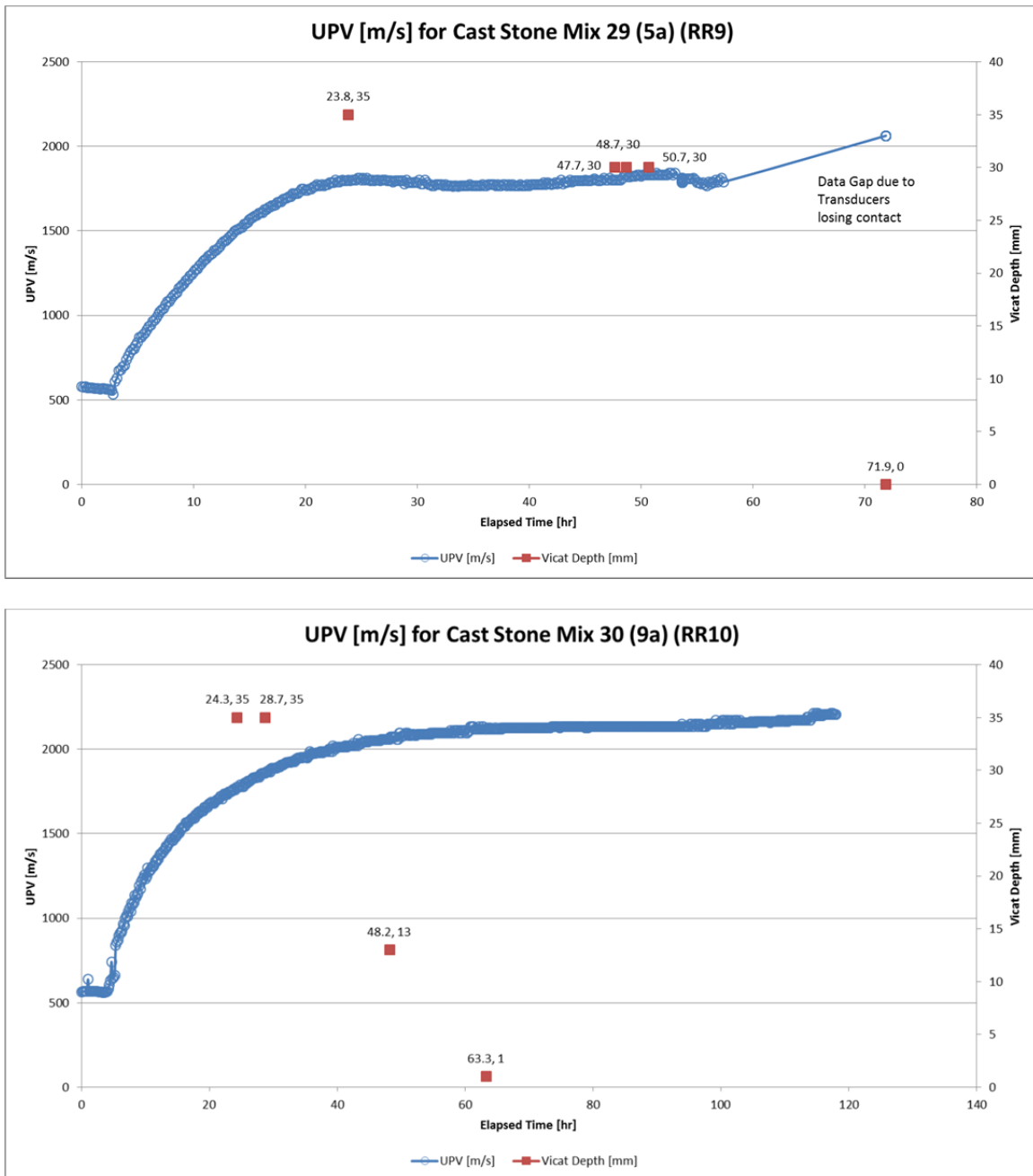


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

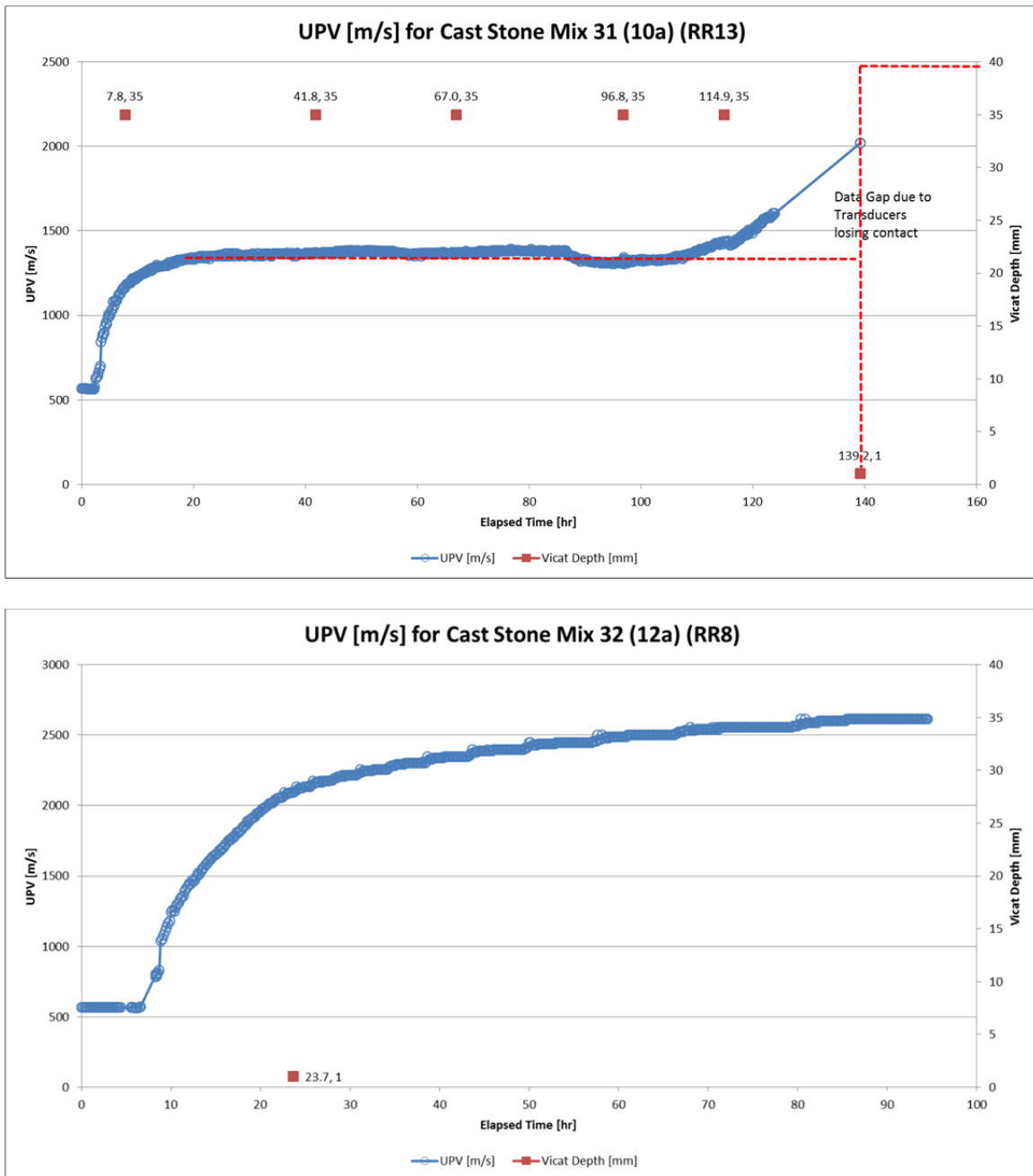


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

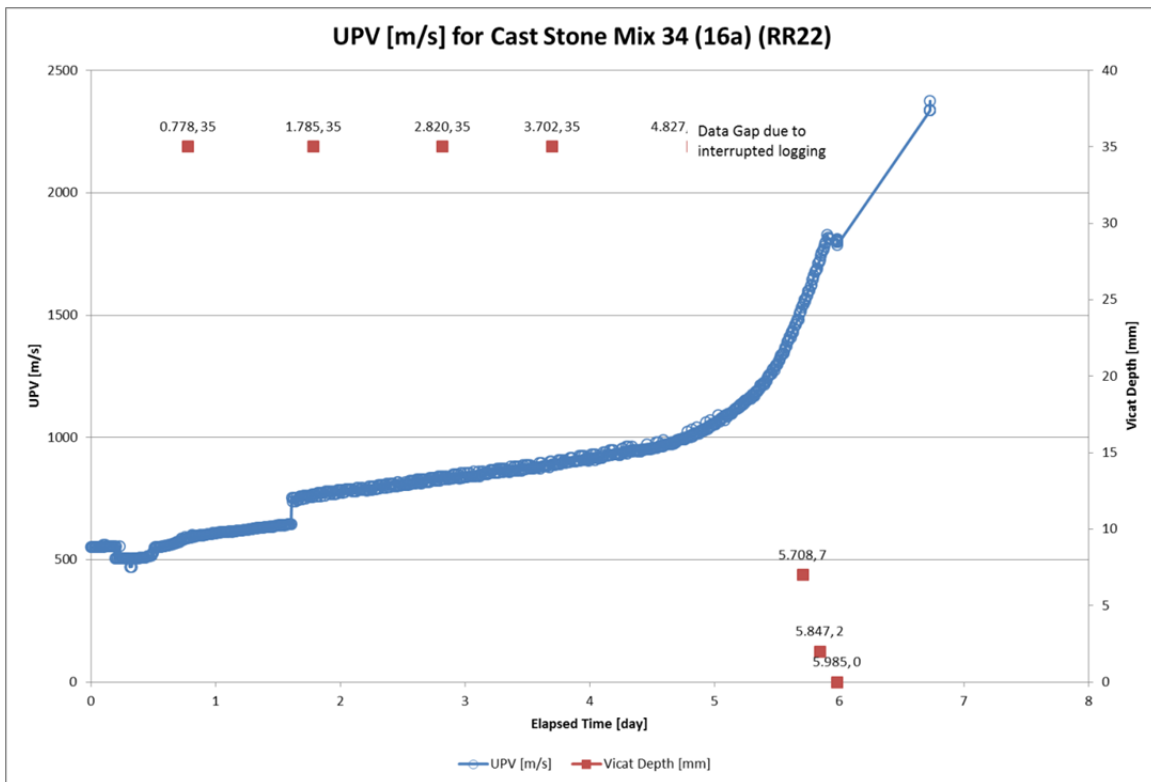
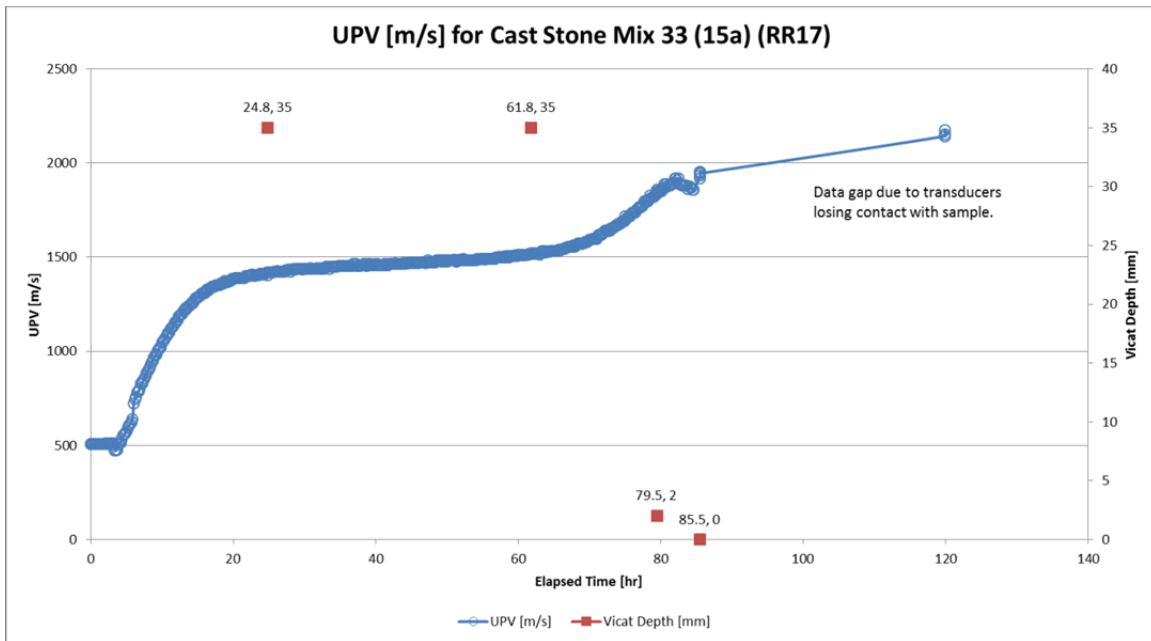


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

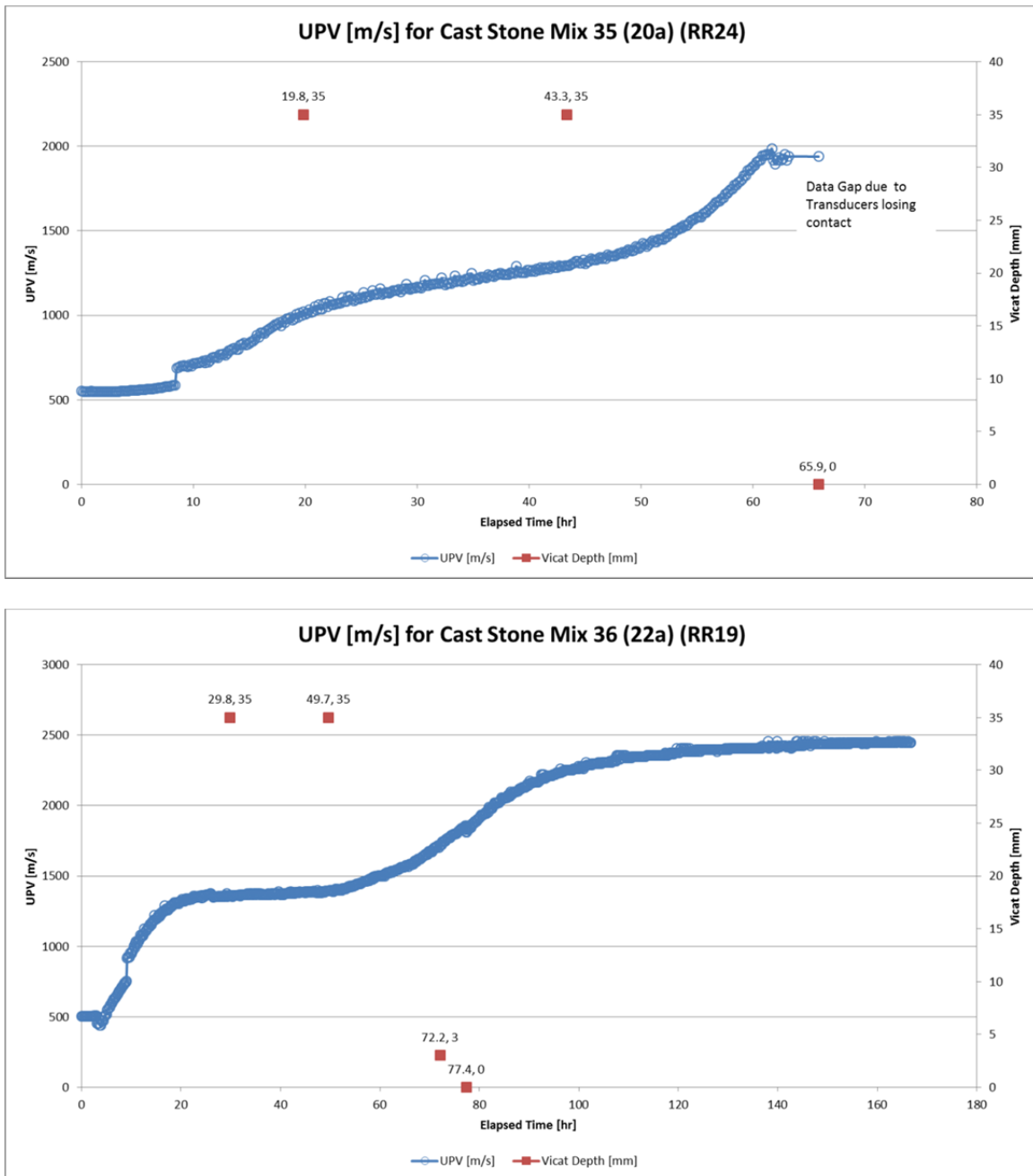


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

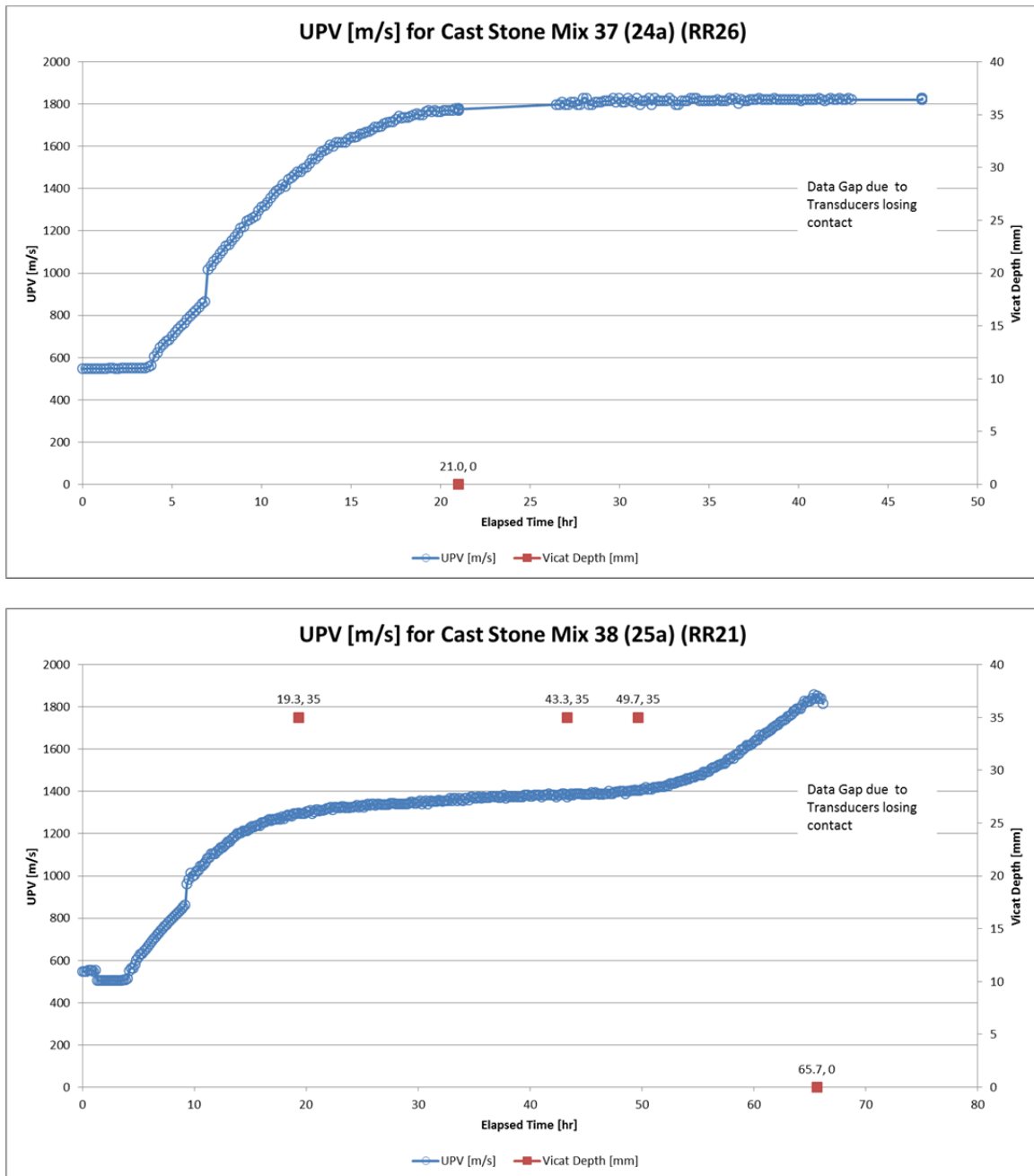


Figure C-1. UPV Data for Cast Stone Mixes to Estimate Set Time (contd)

C.4 Slurry Rheology Properties

Table C-5 contains the measured values for two flow properties: yield stress (Pa) and plastic viscosity (cP). These properties were measured on only one sample per Cast Stone mix, so there are no repeat results with which to calculate means and standard deviations (SDs). The calculated Bingham plastic viscosities and yield stress values over the linear range, including the linear regression R^2 values are shown in Table C-6. Table C-7 shows the Bingham plastic viscosities and yield stress values for the replicate pairs for tests, including averages, SDs, and percent relative standard deviation. The results are similar between the replicate pairs, with %RSD being less than 10% (Table C-7).

C.5 Heat Generation

Table C-8 contains the measured values for three heat generation properties of Cast Stone: heat generation (energy per gram) at 300 hours (J/g), time to peak energy (hr) (referred to time to peak heat generation in main text), and energy per gram at peak (mW/g) (referred to heat generation at peak in main text). These properties were measured on only one sample per Cast Stone mix, so there are no repeat results with which to calculate means and SDs. See Section 6.5 for further discussion of how these property data were obtained.

Table C-5. Slurry Rheological Properties for 38 Cast Stone Mixes

Mix/ Number ^(a)	Random Run Order	Yield Stress ^(b) (Pa)	Plastic Viscosity ^(b) (cP)
1	7	(c)	(c)
2 @	11	9.0	86
3 \$	27	3.1	80
4	28	48.2	329
5	30	67.3	421
6	12	58.7	580
7 #	14	38.1	558
8	1	39.6	520
9	31	2.1	82
10	32	6.8	93
11	6	3.0	92
12	29	3.1	54
13 @	4	9.1	92
14 #	2	36.7	602
15 %	33	36.5	278
16	36	31.1	296
17	25	5.9	67
18	16	43.7	518
19	15	31.9	521
20	37	71.9	322
21 &	23	4.7	93
22 \$	34	3.5	91
23	20	7.9	165
24	38	5.8	98
25 %	35	37.9	287
26 &	18	4.8	91
27 (3a) ^	3	4.2	88
28 (4a)	5	64.6	267
29 (5a)	9	23.7	240
30 (9a)	10	2.7	72
31 (10a)	13	10.0	68
32 (12a)	8	3.9	71
33 (15a) *	17	21.9	202
34 (16a)	22	50.6	338
35 (20a)	24	92.4	275
36 (22a) ^	19	5.1	85
37 (24a)	26	6.7	80
38 (25a) *	21	28.8	236
(a) Test numbers with the same symbols are replicates.			
(b) Values were calculated to several decimal places but rounded to the number of decimal places shown for reporting in this table and statistical data analyses.			
(c) Slurry was too thick to measure rheological properties.			

Table C-6. Bingham Plastic Results

Mix Number	Water-to-Dry Blend Ratio	Up Curve			Down Curve			Fitted Range (s ⁻¹)
		Plastic Viscosity (cP)	Yield Stress (Pa)	R ²	Plastic Viscosity (cP)	Yield Stress (Pa)	R ²	
1	0.4	285	64.3	0.9947	(a)	(a)	(a)	25-300
2	0.6	90.2	7.7	0.9976	86.0	9.0	0.9965	10-300
3	0.6	90.2	4.6	0.9978	88.1	4.2	0.9994	10-300
4	0.4	285	64.3	0.9947	267	64.6	0.9996	25-300
5	0.4	433	67.2	0.9968	421	67.3	0.9994	10-300
6	0.4	597	57.6	0.9994	580	58.7	0.9999	25-300
7	0.4	571	33.9	0.9994	558	38.1	0.9992	10-300
8	0.4	469	61.1	0.9953	520	39.6	0.9991	25-300
9	0.6	82.2	2.7	0.9998	82.0	2.1	0.9999	10-300
10	0.6	95.3	7.3	0.9974	93.1	6.8	0.9982	10-300
11	0.6	92.7	3.3	0.9992	92.0	3.0	0.9997	10-300
12	0.6	49.3	4.0	0.9981	51.2	3.4	0.9978	10-300
13	0.6	96.2	7.8	0.9978	92.2	9.1	0.9964	10-300
14	0.4	617	33.3	0.9997	602	36.7	0.9994	10-300
15	0.4	275	40.3	0.9975	278	36.5	0.9998	10-300
16	0.4	271	37.9	0.9938	296	31.1	0.9999	10-300
17	0.6	68.3	5.1	0.9991	67.4	5.9	0.9962	10-300
18	0.4	525	47.0	0.9989	518	43.7	0.9996	25-300
19	0.4	522	35.9	0.9994	521	31.9	0.9999	25-300
20	0.4	308	79.5	0.9957	322	71.9	0.9955	10-300
21	0.6	94.1	4.7	0.9987	92.6	4.7	0.9993	10-300
22	0.6	91.6	4.1	0.9997	91.0	3.5	0.9998	10-300
23	0.6	161	10.8	0.9985	165	7.9	0.9996	10-300
24	0.6	99.9	6.5	0.9982	97.8	5.8	0.9987	10-300
25	0.4	273	44.8	0.9997	287	37.9	0.9998	10-300
26	0.6	92.3	4.9	0.9989	90.9	4.8	0.9993	10-300
(a) Slurry was too thick to measure rheological properties.								

Table C-7. Bingham Plastic Results for Replicate Pairs of Tests

Mix Number	Water-to-Dry Blend Ratio	Up Curve		Down Curve	
		Plastic Viscosity (cP)	Yield Stress (Pa)	Plastic Viscosity (cP)	Yield Stress (Pa)
2	0.4	617	33.3	602	36.7
14	0.4	571	33.9	558	38.1
	Avg.	594	33.6	580	37.4
	SD	32	0.4	31	1.0
	%RSD	5.4	1.3	5.4	2.7
3	0.6	79.7	3.6	80.0	3.1
19	0.6	91.6	4.1	91.0	3.5
	Avg.	85.7	3.9	85.5	3.3
	SD	8.4	0.3	7.8	0.3
	%RSD	9.8	9.0	9.1	8.8
4	0.6	96.2	7.8	92.2	9.1
11	0.6	90.2	7.7	86.0	9.0
	Avg.	93.2	7.7	89.1	9.0
	SD	4.3	0.1	4.4	0.1
	%RSD	4.6	1.1	4.9	1.2
17	0.4	275	40.3	278	36.5
21	0.4	273	44.8	287	37.9
	Avg.	274	42.5	282	37.2
	SD	1	3.2	6	1.0
	%RSD	0.4	7.6	2.1	2.6
18	0.6	92.3	4.9	90.9	4.8
23	0.6	94.1	4.7	92.6	4.7
	Avg.	93.2	4.8	91.8	4.7
	SD	1.3	0.1	1.2	0.1
	%RSD	1.4	2.1	1.3	1.8

Table C-8. Cast Stone Heat Generation Properties for 38 Cast Stone Mixes

Mix/ Test # ^(a)	Random Run Order	Energy at 300 hr ^(b) (J/g)	Time to Peak Energy ^(c) (hr)	Energy at Peak ^(a) (mW/g)
1	7	230.0	27.30	4259
2 @	11	382.3	73.87	2157
3 \$	27	394.0	86.23	2798
4	28	328.0	96.82	2028
5	30	292.0	37.80	1502
6	12	250.3	35.12	1614
7 #	14	334.6	47.27	2280
8	1	150.4	26.55	892
9	31	434.0	96.20	2557
10	32	363.0	94.97	2939
11	6	287.6	81.75	1452
12	29	405.0	56.02	1873
13 @	4	351.8	71.93	2152
14 #	2	340.9	45.92	2275
15 %	33	314.0	43.88	1838
16	36	294.0	59.53	2280
17	25	394.0	97.20	2115
18	16	306.2	35.93	1725
19	15	253.3	70.10	1106
20	37	326.0	96.35	2095
21 &	23	271.0	51.78	1609
22 \$	34	397.0	84.65	2806
23	20	270.0	48.02	2434
24	38	371.0	91.57	2173
25 %	35	291.0	36.88	1972
26 &	18	272.0	50.72	1675
27 (3a) ^	3	325.2	53.48	1744
28 (4a)	5	288.3	90.00	1635
29 (5a)	9	330.9	60.80	2622
30 (9a)	10	342.3	76.32	1625
31 (10a)	13	394.7	94.97	2589
32(12a)	8	316.9	15.97	3775
33 (15a) *	17	318.0	52.93	2346
34 (16a)	22	297.0	164.07	2200
35 (20a)	24	272.0	93.85	923
36 (22a) ^	19	327.0	55.70	1748
37 (24a)	26	403.0	100.90	1877
38 (25a) *	21	323.0	53.25	2393
(a) Test numbers with the same symbols are replicates.				
(b) Values were reported to the number of decimal places shown and did not require rounding.				
(c) Values were recorded in an hh:mm format, and were converted to values of hours in decimal format with two decimal places.				

C.6 Fresh Densities

The measured densities of freshly prepared Cast Stone mixes closely approximated the densities calculated using the measured densities of the salt solutions and the dry blend materials using the equation

$$\frac{1}{\rho} = \sum_{i=1}^n \frac{x_i}{\rho_i} \quad (\text{C-1})$$

where ρ = density of the mixture, g/cm³
 ρ_i = density of each component, g/cm³
 x_i = mass fraction of each component.

Although reactions occur and there are phase changes, the measured density is not significantly different from the density calculated from the mass fraction and density of the individual components. The results are presented in Table C-9.

Table C-9. Fresh Density Results for 38 Cast Stone Mixes

Mix Number ^(a)	Random Run Number	Density ^(b)		
		Measured (g/cm ³)	Calculated (g/cm ³)	Difference
1	7	1.870	1.893	-1.19%
2 @	11	1.730	1.757	-1.49%
3 \$	27	1.796	1.821	-1.36%
4	28	1.915	1.913	0.08%
5	30	1.960	1.953	0.36%
6	12	1.913	1.928	-0.78%
7 #	14	1.914	1.928	-0.70%
8	1	1.872	1.889	-0.92%
9	31	1.780	1.819	-2.13%
10	32	1.757	1.771	-0.76%
11	6	1.770	1.798	-1.60%
12	29	1.765	1.771	-0.30%
13 @	4	1.755	1.757	-0.12%
14 #	2	1.914	1.928	-0.72%
15 %	33	1.937	1.950	-0.65%
16	36	1.947	1.946	0.04%
17	25	1.732	1.745	-0.74%
18	16	1.913	1.924	-0.57%
19	15	1.909	1.926	-0.87%
20	37	1.950	1.925	1.28%
21 &	23	1.774	1.796	-1.26%
22 \$	34	1.792	1.821	-1.57%
23	20	1.786	1.795	-0.53%
24	38	1.742	1.761	-1.09%
25 %	35	1.942	1.950	-0.38%
26 &	18	1.777	1.796	-1.06%
27 (3a) ^	3	1.807	1.818	-0.63%
28 (4a)	5	1.921	1.910	0.58%
29 (5a)	9	1.956	1.957	-0.07%
30 (9a)	10	1.795	1.817	-1.20%
31 (10a)	13	1.757	1.774	-0.95%
32 (12a)	8	1.769	1.768	0.06%
33 (15a) *	17	1.938	1.953	-0.77%
34 (16a)	22	1.951	1.949	0.09%
35 (20a)	24	1.928	1.921	0.34%
36 (22a) ^	19	1.855	1.818	2.01%
37 (24a)	26	1.749	1.764	-0.82%
38 (25a) *	21	1.952	1.953	-0.06%
(a) Test numbers with the same symbols are replicates.				
(b) Values were rounded to the number of decimal places shown.				

C.7 Free Liquids

Table C-10 contains the results of free liquids (i.e., bleed water) measurements (vol%) from duplicate samples after 1 and 3 days. All except three mixes (# 12, 23, and 32) yielded zero bleed

water after 3 days. Because nearly all results are zero, the means and SDs of the duplicate results were not calculated. See Section 6.7 for further discussion of these data and how they were generated.

Table C-10. Free Water (vol %) for 38 Cast Stone Mixes

Mix/Test # ^(a)	Random Run Order	Day 1 ^(b)		Day 3 ^(b)	
		Duplicate 1	Duplicate 2	Duplicate 1	Duplicate 2
1	7	0.00	0.00	0.00	0.00
2 @	11	0.00	0.00	0.00	0.00
3 \$	27	0.00	0.00	0.00	0.00
4	28	0.00	0.00	0.00	0.00
5	30	0.00	0.00	0.00	0.00
6	12	0.00	0.00	0.00	0.00
7 #	14	0.00	0.00	0.00	0.00
8	1	0.00	0.00	0.00	0.00
9	31	0.45	0.40	0.00	0.00
10	32	0.00	0.00	0.00	0.00
11	6	0.00	0.00	0.00	0.00
12	29	1.09	1.11	0.00	0.00
13 @	4	0.00	0.00	0.00	0.00
14 #	2	0.00	0.00	0.00	0.00
15 %	33	0.00	0.00	0.00	0.00
16	36	0.84	0.68	0.00	0.00
17	25	0.52	0.45	0.28	0.21
18	16	0.00	0.00	0.00	0.00
19	15	0.00	0.00	0.00	0.00
20	37	0.00	0.00	0.00	0.00
21 &	23	0.00	0.00	0.00	0.00
22 \$	34	0.00	0.00	0.00	0.00
23	20	2.33	2.39	2.43	2.20
24	38	0.00	0.00	0.00	0.00
25 %	35	0.00	0.00	0.00	0.00
26 &	18	0.00	0.00	0.00	0.00
27 (3a) ^	3	0.00	0.00	0.00	0.00
28 (4a)	5	0.00	0.00	0.00	0.00
29 (5a)	9	0.00	0.00	0.00	0.00
30 (9a)	10	0.00	0.00	0.00	0.00
31 (10a)	13	0.00	0.00	0.00	0.00
32(12a)	8	5.84	5.91	4.47	4.46
33 (15a) *	17	0.00	0.00	0.00	0.00
34 (16a)	22	0.00	0.00	0.00	0.00
35 (20a)	24	0.00	0.00	0.00	0.00
36 (22a) ^	19	0.00	0.00	0.00	0.00
37 (24a)	26	0.00	0.00	0.00	0.00
38 (25a) *	21	0.00	0.00	0.00	0.00
(a) Test numbers with the same symbols are replicates.					
(b) Values were rounded to the number of decimal places shown.					

Appendix D. Cured Cast Stone Properties Data

This appendix contains data from compressive strength, cured density, and EPA Draft Method 1315 (2012) leach test measurements (effective diffusivity and leachability index) for Cast Stone Mixes 1–26 prepared by PNNL. Compressive strength and cured density results measured by SRNL for a subset of these mixes are also presented.

D.1 Chemical Composition

Table D-1 lists the chemical compositions of Cast Stone Mixes 1–26 (see Table 2-1) measured at PNNL for the specimens spiked with Tc and U for the EPA Draft Method 1315 leach tests. Note that the mixes are labeled as “tests” in Table D-1. The analytical methods are described in Appendix B.

Table D-1. Total Cation Composition of the Monoliths Used for Leach Tests

Analyte	Units	Test 1	Test 2	Test 3	Test 4
		T1LCS1-5HIS	T2LCS2-5AVG	T3HCS2-7.8AVG	CT4HCS2-5HIA
Aluminum	µg/g	83900	71600	54700	42500
Antimony	µg/g	<22.6	<23.7	<23.7	<21.9
Arsenic	µg/g	<35.2	<36.9	<36.9	<34.1
Barium	µg/g	792	712	2260	1710
Cadmium	µg/g	<5.82	<6.09	<6.10	<5.63
Calcium	µg/g	161000	128000	153000	115000
Cesium	µg/g	5.23	4.68	2.24	2.64
Chromium	µg/g	487	626	976	462
Copper	µg/g	<161	<168	<168	<155
Iron	µg/g	25600	21800	18600	13900
Lead	µg/g	64.9	58.8	50.1	33.7
Magnesium	µg/g	12000	29100	36200	27500
Manganese	µg/g	784	1360	1570	1170
Mercury	µg/g	11.6	<11.2	<11.3	<10.4
Molybdenum	µg/g	<38.7	<40.5	<40.6	<37.4
Nickel	µg/g	<602	<630	<631	<583
Phosphorus	µg/g	<3840	<4020	<4020	<3710
Potassium	µg/g	11500	<11600	<11600	<10800
Selenium	µg/g	<68.1	<71.3	<71.4	<65.9
Silicon	µg/g	172000	163000	169000	129000
Silver	µg/g	8.19	3.69	<2.31	<2.13
Sodium	µg/g	48100	63700	53000	86500
Strontium	µg/g	855	714	1330	1000
Titanium	µg/g	4740	4030	3060	2300
Zinc	µg/g	<473	<496	<496	<458
Zirconium	µg/g	<348	<364	<364	<336
Technetium-99	µg/g	4.53	5.93	8.01	4.35
Iodine 127	µg/g	149	76	119	79.7
Uranium 238	µg/g	62.8	45.6	105	60.7
% Dry Solids	wt%	78.4	73.2	75.0	80.9

Table D-1. Total Composition of the Monoliths Used for Leach Tests (contd)

Analyte	Units	Test 5	Test 6	Test 7	Test 8
		T5HCS1-7.8AVG	T6LCS1-7.8AVG	T7LCS2-7.8HIS	T8LCS1-5RAS
Aluminum	µg/g	53600	72700	63400	73800
Antimony	µg/g	<21.8	<22.1	<21.2	<22.5
Arsenic	µg/g	<34.0	<34.4	<33.0	<35.1
Barium	µg/g	2020	644	636	721
Cadmium	µg/g	<5.62	<5.68	5.5	<5.79
Calcium	µg/g	153000	132000	116000	148000
Cesium	µg/g	2.66	4.94	4.68	5.27
Chromium	µg/g	689	618	574	246
Copper	µg/g	<155	<157	<150	<160
Iron	µg/g	17400	21300	19600	22600
Lead	µg/g	50.3	73.1	62.5	59.9
Magnesium	µg/g	15700	10100	26300	10900
Manganese	µg/g	777	649	1240	707
Mercury	µg/g	<10.4	<10.5	<10.1	<10.7
Molybdenum	µg/g	<37.4	<37.8	<36.3	<38.5
Nickel	µg/g	<581	<588	<564	<599
Phosphorus	µg/g	<3710	<3750	<3600	<3820
Potassium	µg/g	<10700	<10900	<10400	<11100
Selenium	µg/g	<65.8	<66.5	<63.8	<67.8
Silicon	µg/g	137000	141000	154000	156000
Silver	µg/g	<2.13	<2.15	<2.07	<2.19
Sodium	µg/g	72100	63600	58700	43100
Strontium	µg/g	1250	698	645	779
Titanium	µg/g	2960	3980	3660	4290
Zinc	µg/g	<457	<462	<444	<471
Zirconium	µg/g	<336	<340	<326	<346
Technetium-99	µg/g	6.64	6.55	7.17	4.26
Iodine 127	µg/g	137	56.6	48	31.5
Uranium 238	µg/g	100	122	130	87.9
% Dry Solids	wt%	79.8	78.8	82.5	78.8

Table D-1. Total Composition of the Monoliths Used for Leach Tests (contd)

Analyte	Units	Test 9	Test 10	Test 11	Test 12
		T9HCS2-7.8HIA	T10HCS1-5HIS	T11LCS1-7.8HIA	T12HCS2-5RAS
Aluminum	µg/g	47900	54400	70400	45300
Antimony	µg/g	<24.0	<24.7	<24.8	<23.0
Arsenic	µg/g	<37.4	<38.5	<38.6	<35.8
Barium	µg/g	1750	2130	590	2130
Cadmium	µg/g	12.7	9.61	11.7	<5.91
Calcium	µg/g	119000	161000	120000	146000
Cesium	µg/g	2.22	2.75	4.37	2.41
Chromium	µg/g	833	592	752	289
Copper	µg/g	<171	<176	<176	<163
Iron	µg/g	13900	17800	18400	16600
Lead	µg/g	52.7	49.1	78.2	36.8
Magnesium	µg/g	27500	16300	8860	32600
Manganese	µg/g	1150	807	560	1350
Mercury	µg/g	<11.4	<11.7	<11.8	<10.9
Molybdenum	µg/g	<41.1	<42.3	<42.5	<39.3
Nickel	µg/g	<639	<658	<661	<611
Phosphorus	µg/g	<4080	<4200	<4210	<3900
Potassium	µg/g	<11800	<12200	<12200	<11300
Selenium	µg/g	<72.3	<74.5	<74.7	<69.1
Silicon	µg/g	129000	142000	127000	152000
Silver	µg/g	<2.34	<2.41	<2.42	<2.24
Sodium	µg/g	95600	66100	89600	71000
Strontium	µg/g	1030	1310	632	1220
Titanium	µg/g	2320	3050	3500	2730
Zinc	µg/g	<503	<518	<519	<481
Zirconium	µg/g	<369	<380	<382	<353
Technetium-99	µg/g	9.26	6	9.16	5.96
Iodine 127	µg/g	62.1	36.6	78.5	47.5
Uranium 238	µg/g	113	86.4	128	64.9
% Dry Solids	wt%	75.8	72.4	74.2	74.0

Table D-1. Total Composition of the Monoliths Used for Leach Tests (contd)

Analyte	Units	Test 13	Test 14	Test 15	Test 16
		T13LCS2-5AVG	T14LCS2-7.8HIS	T15HCS1-7.8HIS	T16HCS1-7.8RAS
Aluminum	µg/g	68500	68600	57500	53600
Antimony	µg/g	<24.7	<22.3	<22.7	<22.2
Arsenic	µg/g	<38.5	<34.8	<35.4	<34.5
Barium	µg/g	715	675	2180	2110
Cadmium	µg/g	<6.35	<5.74	<5.84	<5.70
Calcium	µg/g	130000	124000	167000	163000
Cesium	µg/g	4.74	4.75	2.69	2.64
Chromium	µg/g	541	576	620	346
Copper	µg/g	<175	<158	<161	<157
Iron	µg/g	21300	21200	18700	18300
Lead	µg/g	56.3	64.2	50.2	44.4
Magnesium	µg/g	27700	28000	16900	16600
Manganese	µg/g	1300	1320	820	797
Mercury	µg/g	<11.7	<10.6	<10.8	<10.5
Molybdenum	µg/g	<42.2	<38.2	<38.9	<37.9
Nickel	µg/g	<657	<594	<605	<590
Phosphorus	µg/g	<4190	<3790	<3850	<3760
Potassium	µg/g	<12100	<11000	<11200	<10900
Selenium	µg/g	<74.3	<67.2	<68.4	<66.8
Silicon	µg/g	164000	164000	145000	142000
Silver	µg/g	<2.41	<2.18	<2.21	<2.16
Sodium	µg/g	64500	67400	78800	80000
Strontium	µg/g	684	655	1300	1270
Titanium	µg/g	3870	3890	3180	3120
Zinc	µg/g	<517	<467	<476	<464
Zirconium	µg/g	<380	<343	<349	<341
Technetium-99	µg/g	6.44	7.1	7.11	7.49
Iodine 127	µg/g	49.4	56	44.3	58.1
Uranium 238	µg/g	79.6	137	110	108
% Dry Solids	wt%	73.1	81.1	80.0	81.0

Table D-1. Total Composition of the Monoliths Used for Leach Tests (contd)

Analyte	Units	Test 17	Test 18	Test 19	Test 20
		T17LCS2-5HIA	T18LCS2-7.8RAS	T19-LCS1-7.8 HIA	T20HCS2-5AVG
Aluminum	µg/g	72200	68600	81300	52800
Antimony	µg/g	<24.1	<22.0	<22.8	<22.9
Arsenic	µg/g	<37.6	<34.3	<35.5	<35.7
Barium	µg/g	677	675	713	2280
Cadmium	µg/g	<6.20	<5.67	10.7	<5.89
Calcium	µg/g	127000	123000	147000	156000
Cesium	µg/g	4.56	4.52	4.73	2.74
Chromium	µg/g	514	336	518	408
Copper	µg/g	<171	<156	<162	<163
Iron	µg/g	20900	21600	22600	18600
Lead	µg/g	59.7	60.3	70.5	28.8
Magnesium	µg/g	28200	29200	10500	36600
Manganese	µg/g	1290	1360	675	1570
Mercury	µg/g	<11.5	<10.5	<10.8	<10.9
Molybdenum	µg/g	<41.3	<37.7	<39.0	<39.2
Nickel	µg/g	<642	<586	<607	<610
Phosphorus	µg/g	<4090	<3740	<3870	<3890
Potassium	µg/g	13000	<10800	13200	<11300
Selenium	µg/g	<72.6	<66.3	<68.7	<69.0
Silicon	µg/g	160000	171000	151000	167000
Silver	µg/g	<2.35	<2.15	<2.22	<2.23
Sodium	µg/g	63800	70100	75200	54300
Strontium	µg/g	674	654	745	1300
Titanium	µg/g	3850	4030	4210	3080
Zinc	µg/g	<505	<461	<478	<479
Zirconium	µg/g	<371	<339	<351	<352
Technetium-99	µg/g	6.41	7.26	6.78	4.51
Iodine 127	µg/g	50.6	57.7	55.2	24.8
Uranium 238	µg/g	106	128	127	71.4
% Dry Solids	wt%	72.9	80.8	80.2	80.2

Table D-1. Total Composition of the Monoliths Used for Leach Tests (contd)

Analyte	Units	Test 21	Test 22	Test 23	Test 24
		T21LCS1-7.8HIS	T22HCS2-7.8AVG	T23LCS1-7.8RAS	T24HCS1-5HIA
Aluminum	µg/g	73300	49100	67700	63500
Antimony	µg/g	<24.4	<23.4	<24.6	<25.2
Arsenic	µg/g	<38.0	<36.4	<38.3	<39.2
Barium	µg/g	637	1920	575	2240
Cadmium	µg/g	<6.28	<6.02	<6.32	<6.48
Calcium	µg/g	131000	131000	115000	164000
Cesium	µg/g	4.5	2.32	4.34	2.69
Chromium	µg/g	756	828	409	534
Copper	µg/g	<173	<166	<174	<179
Iron	µg/g	21300	15800	19800	20200
Lead	µg/g	79.6	52.7	76.9	45.8
Magnesium	µg/g	10000	31500	9570	18600
Manganese	µg/g	640	1340	465	777
Mercury	µg/g	<11.6	<11.1	<11.7	<12.0
Molybdenum	µg/g	<41.8	<40.0	<42.0	<43.1
Nickel	µg/g	<650	<623	<654	<671
Phosphorus	µg/g	<4140	<3970	<4170	<4270
Potassium	µg/g	<12000	<11500	<12100	<12400
Selenium	µg/g	<73.6	<70.5	<74.0	<75.9
Silicon	µg/g	144000	142000	133000	157000
Silver	µg/g	<2.38	<2.28	<2.39	<2.45
Sodium	µg/g	93200	101000	86000	76700
Strontium	µg/g	661	1090	577	1280
Titanium	µg/g	3990	2620	3690	3380
Zinc	µg/g	<511	<490	<514	<527
Zirconium	µg/g	<376	<360	<378	<387
Technetium-99	µg/g	9.68	9.7	9.9	6.67
Iodine 127	µg/g	87.3	84.5	92.3	51.2
Uranium 238	µg/g	194	1050	193	101
% Dry Solids	wt%	73.6	74.2	74.0	72.5

Table D-1. Total Composition of the Monoliths Used for Leach Tests (contd)

Analyte	Units	Test 25	Test 26
		T25HCS1-7.8HIS	T26LCS1-7.8HIS
Aluminum	μg/g	55600	55400
Antimony	μg/g	<22.9	<24.6
Arsenic	μg/g	<35.7	<38.3
Barium	μg/g	1890	1950
Cadmium	μg/g	<5.89	<6.33
Calcium	μg/g	137000	143000
Cesium	μg/g	2.71	2.45
Chromium	μg/g	578	807
Copper	μg/g	<163	<175
Iron	μg/g	16600	17000
Lead	μg/g	50	59.6
Magnesium	μg/g	15100	15600
Manganese	μg/g	613	636
Mercury	μg/g	<10.9	<11.7
Molybdenum	μg/g	<39.2	<42.1
Nickel	μg/g	<610	<655
Phosphorus	μg/g	<3880	<4180
Potassium	μg/g	<11300	<12100
Selenium	μg/g	<68.9	<74.1
Silicon	μg/g	128000	132000
Silver	μg/g	<2.23	<2.40
Sodium	μg/g	60200	96100
Strontium	μg/g	1070	1110
Titanium	μg/g	2750	2820
Zinc	μg/g	<479	<515
Zirconium	μg/g	<352	<378
Technetium-99	μg/g	7	9.69
Iodine 127	μg/g	60.6	77.9
Uranium 238	μg/g	134	86.5
% Dry Solids	wt%	80.5	74.3

D.2 Compressive Strength

Compressive strength measurements for three cylindrical samples (referred to as *repeats*) of each of the Cast Stone mixes are presented in Table D-2 for the PNNL results and Table D-3 for the SRNL results. SRNL did not re-batch and test the 12 mixes that were mis-batched, so there are no SRNL data for those mixes (nor any PNNL data for the mixes mis-batched by SRNL). The tables also show the means and standard deviations (SDs) for the three repeat results. The “Repeat SDs” represent the variation in making three cylindrical samples from each mix and measuring compressive strength. The results are presented in units of pounds per square inch (psi) and megapascals (MPa). The mean values in psi units from the PNNL data (Table D-2) were used as the compressive strength response values for plots and statistical analyses of the data from the 26 mixes. Note that only the PNNL compressive strength data were statistically analyzed.

Table D-2. PNNL Compressive Strength Results for 26 Cast Stone Mixes

Test/Mix # ^(a)	Random Run Order	Compressive Strength (psi) ^(b)					Compressive Strength (MPa) ^(c)				
		Repeats			Summary		Repeats			Summary	
		1	2	3	Mean	SD	1	2	3	Mean	SD
1	7	4633	4424	4173	4410	230.3	31.94	30.50	28.77	30.40	1.587
2 @	11	4809	4844	4823	4825	17.6	33.16	33.40	33.25	33.27	0.121
3 \$	3	2522	2550	2428	2500	63.9	17.39	17.58	16.74	17.24	0.440
4	5	8988	7237	7878	8034	885.9	61.97	49.90	54.32	55.40	6.107
5	9	5123	4991	5153	5089	86.2	35.32	34.41	35.53	35.09	0.595
6	12	3198	2937	3244	3126	165.6	22.05	20.25	22.37	21.56	1.143
7 #	14	6931	6537	5865	6444	539.0	47.79	45.07	40.44	44.43	3.716
8	1	1768	1774	1716	1753	31.9	12.19	12.23	11.83	12.08	0.220
9	10	1955	1926	1404	1762	310.1	13.48	13.28	9.68	12.15	2.139
10	13	3472	2608	1787	2622	842.6	23.94	17.98	12.32	18.08	5.811
11	6	847	875	848	857	15.9	5.84	6.03	5.85	5.91	0.107
12	8	6247	6467	6527	6414	147.4	43.07	44.59	45.00	44.22	1.017
13 @	4	4840	4872	4809	4840	31.5	33.37	33.59	33.16	33.37	0.215
14 #	2	6696	6454	6856	6669	202.4	46.17	44.50	47.27	45.98	1.395
15 %	17	4543	4570	5819	4977	729.0	31.32	31.51	40.12	34.32	5.027
16	22	5324	6039	6454	5939	571.6	36.71	41.64	44.50	40.95	3.941
17	25	3681	4209	4099	3996	278.6	25.38	29.02	28.26	27.55	1.920
18	16	7227	7298	7388	7304	80.7	49.83	50.32	50.94	50.36	0.556
19	15	2995	2554	2996	2848	254.9	20.65	17.61	20.66	19.64	1.758
20	24	7778	7929	7361	7689	294.2	53.63	54.67	50.75	53.02	2.031
21 &	23	1411	1336	1329	1359	45.5	9.73	9.21	9.16	9.37	0.316
22 \$	19	2316	2287	2587	2397	165.5	15.97	15.77	17.84	16.53	1.142
23	20	1578	1546	1803	1642	140.1	10.88	10.66	12.43	11.32	0.965
24	26	3588	3292	3610	3497	177.6	24.74	22.70	24.89	24.11	1.223
25 %	21	6032	6295	6209	6179	134.1	41.59	43.40	42.81	42.60	0.923
26 &	18	922	980	940	947	29.7	6.36	6.76	6.48	6.53	0.205
(a) Test numbers with the same symbols are replicates.											
(b) The repeat values were rounded to zero decimal places before calculating the mean and SD. After the mean and SD were calculated, they were rounded to zero and one decimal places, respectively.											
(c) The repeat values were rounded to two decimal places before calculating the mean and SD. After the mean and SD were calculated, they were rounded to two and three decimal places, respectively.											

Table D-3. SRNL Compressive Strength Results for 26 Cast Stone Mixes

Test/Mix # ^(a)	Random Run Order	Compressive Strength (psi) ^(b)					Compressive Strength (MPa) ^(c)					
		Repeats			Summary		Repeats			Summary		
		1	2	3	Mean	SD	1	2	3	Mean	SD	
1	7	3591	2940	3007	3179	358.1	24.76	20.27	20.73	21.92	2.470	
2	@	11	4623	4737	4302	4554	225.6	31.87	32.66	29.66	1.555	
3	— ^(e)	—	—	—	—	—	—	—	—	—	—	
4	—	—	—	—	—	—	—	—	—	—	—	
5	—	—	—	—	—	—	—	—	—	—	—	
6	12	2871	2925	2834	2877	45.8	19.79	20.17	19.54	19.83	0.317	
7	#	14	5575	5845	5594	5671	150.7	38.44	40.30	38.57	1.038	
8	1	1661	1660	1654	1658	3.8	11.45	11.45	11.40	11.43	0.029	
9	—	—	—	—	—	—	—	—	—	—	—	
10	—	—	—	—	—	—	—	—	—	—	—	
11	6	1065	1078	1070	1071	6.6	7.34	7.43	7.38	7.38	0.045	
12	—											
13	@	4	4671	4970	4555	4732	214.1	32.21	34.27	31.41	1.476	
14	#	2	5584	5557	5516	5552	34.2	38.50	38.31	38.03	0.236	
15	—	—	—	—	—	—	—	—	—	—	—	
16	—	—	—	—	—	—	—	—	—	—	—	
17	25	5131	4594	5050	4925	289.5	35.38	31.67	34.82	33.96	2.000	
18	16	6282	5930	6081	6098	176.6	43.31	40.89	41.93	42.04	1.214	
19	15	3164	3123	3106	3131	29.8	21.82	21.53	21.42	21.59	0.207	
20	—	—	—	—	—	—	—	—	—	—	—	
21	&	23	1011	1218	1057	1095	108.7	6.97	8.40	7.29	0.750	
22	—	—	—	—	—	—	—	—	—	—	—	
23	20	190 ^(d)	1683	1360	1522	228.4	1.31 ^(d)	11.60	9.38	10.49	1.570	
24	—	—	—	—	—	—	—	—	—	—	—	
25	—	—	—	—	—	—	—	—	—	—	—	
26	&	18	1166	1168	1137	1157	17.3	8.04	8.05	7.84	0.118	
27 (3a)	^	3	1329	1363	1380	1357	26.0	9.16	9.40	9.51	0.179	
28 (4a)		5	7683	7575	7807	7688	116.1	52.97	52.23	53.83	0.801	
29 (5a)		9	5074	5281	5124	5160	108.0	34.98	36.41	35.33	0.745	
30 (9a)		10	1332	1355	1385	1357	26.6	9.18	9.34	9.55	0.186	
31 (10a)		13	4381	4585	4713	4560	167.4	30.21	31.61	32.49	1.150	
32 (12a)		8	4189	4348	3930	4156	211.0	28.88	29.98	27.10	1.453	
33 (15a)	*	17	5023	4839	4338	4733	354.5	34.63	33.36	29.91	32.63	2.442
34 (16a)		22	7263	7238	6809	7103	255.2	50.08	49.90	46.95	48.98	1.757
35 (20a)		24	6928	6116	6836	6627	444.6	47.77	42.17	47.13	45.69	3.065
36 (22a)	^	19	1382	1381	1366	1376	9.0	9.53	9.52	9.42	0.061	
37 (24a)		26	5018	5085	5307	5137	151.3	34.60	35.06	36.59	35.42	1.042
38 (25a)	*	21	5134	4798	5088	5007	182.2	35.40	33.08	35.08	34.52	1.257
(a) Test numbers with the same symbols are replicates.												
(b) The repeat values were rounded to zero decimal places before calculating the Mean and SD. After the mean and SD were calculated, they were rounded to zero and one decimal places, respectively.												
(c) The repeat values were rounded to one decimal place before calculating the mean and SD. After the mean and SD were calculated, they were rounded to one and two decimal places, respectively.												
(d) This value was judged to be an outlier and was not used to calculate the mean and SD.												
(e) A dash (—) denotes mixes that were not re-made and tested after originally being mis-batched.												

D.3 Density, Surface Area, and Volume of PNNL Cast Stone Specimens

Table D-4 lists the cured monolith density, surface area, and volume of each of the six cylinders prepared from Cast Stone Mixes 1–26 made at PNNL. The average of three diameter measurements and the average of three overall length measurements were used to determine the volume of each monolith. Each monolith was weighed to determine its mass. The mass of the monolith was divided by the calculated volume resulting in the cured density. Cylinder surface areas and volumes were calculated from the average measured monolith dimensions.

Table D-4. Cast Stone Monolith Density, Surface Area, and Volume

Mix #1 Sample Name	Cast Stone Cylinder Density (g/cc)	Cast Stone Cylinder Surface Area (cm ²)	Cast Stone Cylinder Volume (cc)	Mix #2 Sample Name	Cast Stone Cylinder Density (g/cc)	Cast Stone Cylinder Surface Area (cm ²)	Cast Stone Cylinder Volume (cc)
CS-T1LCS1-5HIS-1	1.87	196	195	CS-T2LCS2-5AVG-1	1.74	193	190
CS-T1LCS1-5HIS-2	1.86	197	196	CS-T2LCS2-5AVG-2	1.72	196	194
CS-T1LCS1-5HIS-3	1.87	194	192	CS-T2LCS2-5AVG-3	1.71	196	195
CS-T1LCS1-5HIS-4	1.89	196	193	CS-T2LCS2-5AVG-4	1.72	196	194
CS-T1LCS1-5HIS-5	1.87	197	195	CS-T2LCS2-5AVG-5	1.74	194	191
CS-T1LCS1-5HIS-6	1.85	197	197	CS-T2LCS2-5AVG-6	1.73	196	193
Mix #3 Sample Name				Mix #4 Sample Name			
CS-T3HCS2-7.8AVG-1	1.77	196	195	CS-T4HCS2-5HIA-1	1.94	196	193
CS-T3HCS2-7.8AVG-2	1.78	196	194	CS-T4HCS2-5HIA-2	1.94	195	192
CS-T3HCS2-7.8AVG-3	1.76	196	194	CS-T4HCS2-5HIA-3	1.92	196	194
CS-T3HCS2-7.8AVG-4	1.76	197	196	CS-T4HCS2-5HIA-4	1.94	197	194
CS-T3HCS2-7.8AVG-5	1.77	195	193	CS-T4HCS2-5HIA-5	1.94	195	193
CS-T3HCS2-7.8AVG-6	1.76	197	197	CS-T4HCS2-5HIA-6	1.97	193	189
Mix #5 Sample Name				Mix #6 Sample Name			
CS-T5HCS1-7.8AVG-1	1.98	194	191	CS-T6LCS1-7.8AVG-1	1.92	196	193
CS-T5HCS1-7.8AVG-2	1.95	197	195	CS-T6LCS1-7.8AVG-2	1.90	195	193
CS-T5HCS1-7.8AVG-3	1.95	196	194	CS-T6LCS1-7.8AVG-3	1.91	197	196
CS-T5HCS1-7.8AVG-4	1.95	197	195	CS-T6LCS1-7.8AVG-4	1.92	194	192
CS-T5HCS1-7.8AVG-5	1.96	193	190	CS-T6LCS1-7.8AVG-5	1.90	196	194
CS-T5HCS1-7.8AVG-6	1.96	196	194	CS-T6LCS1-7.8AVG-6	1.90	195	193
Mix #7 Sample Name				Mix #8 Sample Name			
CS-T7LCS2-7.8HIS-1	1.95	195	193	CS-T8LCS1-5RAS-1	1.89	194	192
CS-T7LCS2-7.8HIS-2	1.94	195	193	CS-T8LCS1-5RAS-2	1.89	194	192
CS-T7LCS2-7.8HIS-3	1.96	195	193	CS-T8LCS1-5RAS-3	1.88	195	193
CS-T7LCS2-7.8HIS-4	1.94	195	193	CS-T8LCS1-5RAS-4	1.87	196	195
CS-T7LCS2-7.8HIS-5	1.92	198	197	CS-T8LCS1-5RAS-5	1.89	194	191
CS-T7LCS2-7.8HIS-6	1.94	196	194	CS-T8LCS1-5RAS-7	1.86	196	195

Table D-4. Cast Stone Monolith Density, Surface Area, and Volume (contd)

Mix #9 Sample Name	Cast Stone Cylinder Density (g/cc)	Cast Stone Cylinder Surface Area (cm ²)	Cast Stone Cylinder Volume (cc)	Mix #10 Sample Name	Cast Stone Cylinder Density (g/cc)	Cast Stone Cylinder Surface Area (cm ²)	Cast Stone Cylinder Volume (cc)
CS-T9HCS2-7.8HIA-1	1.79	195	193	CS-T10HCS1-5HIS-1	1.79	193	190
CS-T9HCS2-7.8HIA-2	1.78	196	195	CS-T10HCS1-5HIS-2	1.78	195	193
CS-T9HCS2-7.8HIA-3	1.80	195	192	CS-T10HCS1-5HIS-3	1.77	193	191
CS-T9HCS2-7.8HIA-4	1.78	196	194	CS-T10HCS1-5HIS-4	1.80	192	188
CS-T9HCS2-7.8HIA-5	1.82	196	193	CS-T10HCS1-5HIS-5	1.81	191	187
CS-T9HCS2-7.8HIA-6	1.80	195	192	CS-T10HCS1-5HIS-6	1.78	194	192
Mix #11 Sample Name				Mix #12 Sample Name			
CS-T11LCS1-7.8HIA-1	1.75	196	194	CS-T12HCS2-5RAS-1	1.82	190	186
CS-T11LCS1-7.8HIA-2	1.77	195	192	CS-T12HCS2-5RAS-2	1.81	192	189
CS-T11LCS1-7.8HIA-3	1.78	195	192	CS-T12HCS2-5RAS-3	1.79	194	191
CS-T11LCS1-7.8HIA-4	1.74	197	196	CS-T12HCS2-5RAS-4	1.80	194	192
CS-T11LCS1-7.8HIA-5	1.75	195	193	CS-T12HCS2-5RAS-5	1.78	196	195
CS-T11LCS1-7.8HIA-6	1.76	195	193	CS-T12HCS2-5RAS-6	1.80	193	190
Mix #13 Sample Name				Mix #14 Sample Name			
CS-T13LCS2-5AVG-1	1.68	197	195	CS-T14LCS2-7.8HIS-1	1.92	194	192
CS-T13LCS2-5AVG-2	1.71	196	195	CS-T14LCS2-7.8HIS-2	1.94	194	191
CS-T13LCS2-5AVG-3	1.71	196	194	CS-T14LCS2-7.8HIS-3	1.91	197	195
CS-T13LCS2-5AVG-4	1.71	196	194	CS-T14LCS2-7.8HIS-4	1.93	194	191
CS-T13LCS2-5AVG-5	1.73	194	192	CS-T14LCS2-7.8HIS-5	1.93	194	191
CS-T13LCS2-5AVG-6	1.71	196	194	CS-T14LCS2-7.8HIS-6	1.90	198	197
Mix #15 Sample Name				Mix #16 Sample Name			
CS-T15HCS1-7.8HIS-1	1.95	195	193	CS-T16HCS1-7.8RAS-1	1.97	194	192
CS-T15HCS1-7.8HIS-2	1.97	194	191	CS-T16HCS1-7.8RAS-2	1.97	195	193
CS-T15HCS1-7.8HIS-3	1.96	195	193	CS-T16HCS1-7.8RAS-3	1.98	194	191
CS-T15HCS1-7.8HIS-4	1.95	196	194	CS-T16HCS1-7.8RAS-4	1.97	194	192
CS-T15HCS1-7.8HIS-5	1.94	196	194	CS-T16HCS1-7.8RAS-5	1.98	193	190
CS-T15HCS1-7.8HIS-6	1.92	198	197	CS-T16HCS1-7.8RAS-6	2.04	191	186

Table D-4. Cast Stone Monolith Density, Surface Area, and Volume (contd)

Mix #17 Sample Name	Cast Stone Cylinder Density (g/cc)	Cast Stone Cylinder Surface Area (cm ²)	Cast Stone Cylinder Volume (cc)	Mix #18 Sample Name	Cast Stone Cylinder Density (g/cc)	Cast Stone Cylinder Surface Area (cm ²)	Cast Stone Cylinder Volume (cc)
CS-T17LCS2-5HIA-1	1.71	196	195	CS-T18LCS2-7.8RAS-1	1.90	196	195
CS-T17LCS2-5HIA-2	1.73	194	192	CS-T18LCS2-7.8RAS-2	1.90	195	193
CS-T17LCS2-5HIA-3	1.74	195	193	CS-T18LCS2-7.8RAS-3	1.93	194	192
CS-T17LCS2-5HIA-4	1.74	193	191	CS-T18LCS2-7.8RAS-4	1.92	194	192
CS-T17LCS2-5HIA-5	1.76	193	190	CS-T18LCS2-7.8RAS-5	1.90	197	195
CS-T17LCS2-5HIA-6	1.74	194	191	CS-T18LCS2-7.8RAS-6	1.92	196	194
Mix #19 Sample Name				Mix #20 Sample Name			
CS-T19LCS1-7.8HIA-1	1.91	195	193	CS-T20HCS2-5AVG-1	1.96	193	190
CS-T19LCS1-7.8HIA-2	1.91	196	194	CS-T20HCS2-5AVG-2	1.95	194	191
CS-T19LCS1-7.8HIA-3	1.91	196	194	CS-T20HCS2-5AVG-3	1.92	196	195
CS-T19LCS1-7.8HIA-4	1.92	197	195	CS-T20HCS2-5AVG-4	1.93	196	194
CS-T19LCS1-7.8HIA-5	1.93	193	190	CS-T20HCS2-5AVG-5	1.95	194	191
CS-T19LCS1-7.8HIA-6	1.91	195	192	CS-T20HCS2-5AVG-6	1.95	199	197
Mix #21 Sample Name				Mix #22 Sample Name			
CS-T21LCS1-7.8HIS-1	1.75	195	194	CS-T22HCS2-7.8AVG-1	1.80	195	192
CS-T21LCS1-7.8HIS-2	1.77	195	193	CS-T22HCS2-7.8AVG-2	1.77	197	195
CS-T21LCS1-7.8HIS-3	1.79	194	191	CS-T22HCS2-7.8AVG-3	1.79	195	193
CS-T21LCS1-7.8HIS-4	1.77	196	194	CS-T22HCS2-7.8AVG-4	1.77	197	195
CS-T21LCS1-7.8HIS-5	1.77	196	194	CS-T22HCS2-7.8AVG-5	1.79	195	193
CS-T21LCS1-7.8HIS-6	1.79	194	192	CS-T22HCS2-7.8AVG-6	1.78	196	195
Mix #23 Sample Name				Mix #24 Sample Name			
CS-T23LCS1-7.8RAS-1	1.81	192	189	CS-T24HCS1-5HIA-1	1.79	194	192
CS-T23LCS1-7.8RAS-2	1.79	194	192	CS-T24HCS1-5HIA-2	1.76	196	195
CS-T23LCS1-7.8RAS-3	1.79	195	193	CS-T24HCS1-5HIA-3	1.74	198	198
CS-T23LCS1-7.8RAS-4	1.81	193	190	CS-T24HCS1-5HIA-4	1.77	195	193
CS-T23LCS1-7.8RAS-5	1.81	194	192	CS-T24HCS1-5HIA-5	1.79	194	192
CS-T23LCS1-7.8RAS-6	1.79	194	192	CS-T24HCS1-5HIA-6	1.78	195	193

Table D-4. Cast Stone Monolith Density, Surface Area, and Volume (contd)

Mix #25 Sample Name	Cast Stone Cylinder Density (g/cc)	Cast Stone Cylinder Surface Area (cm ²)	Cast Stone Cylinder Volume (cc)	Mix #26 Sample Name	Cast Stone Cylinder Density (g/cc)	Cast Stone Cylinder Surface Area (cm ²)	Cast Stone Cylinder Volume (cc)
CS-T25HCS1-7.8HIS-1	1.91	198	197	CS-T26LCS1-7.8HIS-1	1.79	195	193
CS-T25HCS1-7.8HIS-2	1.94	198	196	CS-T26LCS1-7.8HIS-2	1.76	197	196
CS-T25HCS1-7.8HIS-3	1.95	196	194	CS-T26LCS1-7.8HIS-3	1.79	195	193
CS-T25HCS1-7.8HIS-4	1.98	196	193	CS-T26LCS1-7.8HIS-4	1.78	197	196
CS-T25HCS1-7.8HIS-5	1.97	194	191	CS-T26LCS1-7.8HIS-5	1.79	196	194
CS-T25HCS1-7.8HIS-6	1.95	196	194	CS-T26LCS1-7.8HIS-6	1.78	196	194

Table D-5 lists the cured density values for triplicate samples prepared from 26 Cast Stone mixes at SRNL. SRNL did not re-batch and test the 12 mixes that were mis-batched, so there are no data for those mixes. Table D-5 also shows the means and SDs for cured- density measurements made using the pycnometer method on three “repeat” monoliths made from each mix. The “Repeat SDs” represent the variation in preparing three samples from each mix and measuring the cured density for each sample.

Table D-5. Cast Stone Cured Densities (g/cm³) Measured by Pycnometer at SRNL

Test/Mix # ^(a)	Random Run Order	Repeats ^(b)			Mean ^(b)	SD ^(b)
		1	2	3		
1	7	1.898	1.898	1.897	1.898	0.0006
2 @	11	1.770	1.762	1.785	1.772	0.0117
3	— ^(c)	—	—	—	—	—
4	—	—	—	—	—	—
5	—	—	—	—	—	—
6	12	1.938	1.936	1.939	1.938	0.0015
7 #	14	1.938	1.937	1.940	1.938	0.0015
8	1	1.890	1.891	1.883	1.888	0.0044
9	—	—	—	—	—	—
10	—	—	—	—	—	—
11	6	1.780	1.783	1.788	1.784	0.0040
12	—	—	—	—	—	—
13 @	4	1.768	1.757	1.763	1.763	0.0055
14 #	2	1.932	1.935	1.942	1.936	0.0051
15	—	—	—	—	—	—
16	—	—	—	—	—	—
17	25	1.776	1.771	1.771	1.773	0.0029
18	16	1.930	1.920	1.918	1.923	0.0064
19	15	1.929	1.927	1.929	1.928	0.0012
20	—	—	—	—	—	—
21 &	23	1.794	1.799	1.790	1.794	0.0045
22	—					
23	20	1.809	1.810	1.801	1.807	0.0049
24	—	—	—	—	—	—
25	—	—	—	—	—	—
26 &	18	1.810	1.799	1.802	1.804	0.0057
27 (3a) ^	3	1.803	1.804	1.801	1.803	0.0015
28 (4a)	5	1.952	1.950	1.956	1.953	0.0031
29 (5a)	9	1.976	1.969	1.968	1.971	0.0044
30 (9a)	10	1.808	1.809	1.808	1.808	0.0006
31 (10a)	13	1.808	1.806	1.797	1.804	0.0059
32(12a)	8	1.801	1.808	1.804	1.804	0.0035
33 (15a) *	17	1.967	1.968	1.968	1.968	0.0006
34 (16a)	22	1.987	1.984	1.978	1.983	0.0046
35 (20a)	24	1.973	1.979	1.975	1.976	0.0031
36 (22a) ^	19	1.825	1.820	1.821	1.822	0.0026
37 (24a)	26	1.802	1.797	1.806	1.802	0.0045
38 (25a) *	21	1.981	1.975	1.982	1.979	0.0038
(a) Test numbers with the same symbols are replicates.						
(b) The repeat density values were rounded to three decimal places before calculating the mean and SD. After the mean and SD were calculated, they were rounded to three and four decimal places, respectively.						
(c) A dash (–) denotes mixes that were not re-made and tested after originally being mis-batched.						

D.4 Porosity

Table D-6 lists the porosity values for triplicate samples prepared from 26 Cast Stone mixes made at SRNL. SRNL did not re-batch and test the 12 mixes that were mis-batched, so there are no data for those mixes. Table D-6 also shows the means and SDs for the three results per mix. The “Repeat SDs” represent the variation in preparing three samples from each mix and measuring the porosity of each sample.

The porosity was determined by the mass loss upon heating samples to 105 °C in a laboratory oven. The samples were measured repeatedly until the mass change on consecutive days was <5%. The Cast Stone pieces that were used to measure the porosity were taken from the Cast Stone specimens by breaking the cylinders and removing pieces from the center region. The total porosity is defined here as the percentage of total volume occupied by the pore solution for the saturated case. It is assumed that the density of the pore solution is equivalent to the density of the salt solution from which the sample was prepared. The porosity is then calculated by dividing the volume of the pore solution by the overall volume of the Cast Stone. The volume of the pore solution is determined by assuming the mass loss is due entirely to water and dividing by the density of water and the volume fraction of water in the original salt solution.

Table D-6. Cast Stone Porosities (%) Measured at SRNL

Test/Mix # ^(a)	Random Run Order	Repeats ^(b)			Mean ^(b)	SD ^(b)
		1	2	3		
1		47.9	48.7	48.1	48.2	0.42
2	@	55.1	55.4	54.9	55.1	0.25
3	— ^(c)	—	—	—	—	—
4	—	—	—	—	—	—
5	—	—	—	—	—	—
6		51.3	50.7	51.1	51.0	0.31
7	#	47.3	47.3	47.5	47.4	0.12
8		51.1	50.4	50.5	50.7	0.38
9	—	—	—	—	—	—
10	—	—	—	—	—	—
11		60.4	61.2	60.9	60.8	0.40
12	—	—	—	—	—	—
13	@	55.0	54.8	55.2	55.0	0.20
14	#	47.8	47.2	47.8	47.6	0.35
15	—	—	—	—	—	—
16	—	—	—	—	—	—
17		57.1	57.1	57.0	57.1	0.06
18		48.7	48.5	48.7	48.6	0.12
19		51.0	56.7	51.4	53.0	3.18
20	—	—	—	—	—	—
21	&	62.5	61.9	62.3	62.2	0.31
22	—	—	—	—	—	—
23		61.0	61.4	64.9	62.4	2.15
24	—	—	—	—	—	—
25	—	—	—	—	—	—
26	&	61.8	61.3	61.3	61.5	0.29
27 (3a)	^	58.2	57.7	58.8	58.2	0.55
28 (4a)		47.0	47.9	47.9	47.6	0.52
29 (5a)		46.8	46.9	46.9	46.9	0.06
30 (9a)		58.4	58.0	58.2	58.2	0.20
31 (10a)		55.8	55.8	55.8	55.8	0.00
32 (12a)		58.2	55.1	54.4	55.9	2.02
33 (15a)	*	49.0	48.2	48.0	48.4	0.53
34 (16a)		48.3	48.6	48.7	48.5	0.21
35 (20a)		48.6	48.3	49.1	48.7	0.40
36 (22a)	^	60.0	59.7	59.5	59.7	0.25
37 (24a)		57.5	57.6	57.5	57.5	0.06
38 (25a)	*	48.3	48.0	48.9	48.4	0.46

(a) Test numbers with the same symbols are replicates.
(b) The repeat porosity values were rounded to one decimal place before calculating the mean and SD. After the mean and SD were calculated, they were rounded to one and two decimal places, respectively.
(c) A dash (—) denotes mixes that were not re-made and tested after originally being mis-batched.

D.5 EPA Draft Method 1315 Leach Test Results

Table D-7 through Table D-13 provide the measured effective diffusivities of Na, nitrate (NO₃), nitrite (NO₂), I, Tc, Cr, and U based on performing EPA Draft Method 1315 on duplicate (i.e., replicate) samples from each of the 26 Cast Stone mixes prepared at PNNL (Mixes 1–26). The leach tests were conducted for a total of 91 days and leachate exchanges occurred at 0.08, 1, 2, 7, 14, 28, 42, 49, 63, 77, and 91 cumulative days. The 77- and 91-day intervals provide longer-term data beyond the 63 days in EPA Draft Method 1315, but they were not included in the statistical analyses for the screening tests. The results are provided for both monoliths of each of the 26 mixes.

For purposes of statistical analyses, the average diffusivity values were calculated for the total durations of 28, 42, 49, and 63 days for each of the two duplicate leach tests for each of the 26 Cast Stone mixes. Also, leachability index (LI) values were calculated using these averages, according to the formula

$$LI = -\log_{10}\left(\frac{Diff_{28} + Diff_{42} + Diff_{49} + Diff_{63}}{4}\right) \quad (D-1)$$

for each of the duplicate samples for each of the 26 Cast Stone mixes. Table D-14 and Table D-15 present the average effective diffusivity and LI values for Na, NO₃, NO₂, I, Tc, Cr, and U for each of the duplicate leach tests for the 26 Cast Stone samples. Also included are the mean and SD values for the duplicate (repeat) values. The mean values of LI (for the 28-, 42-, 49-, and 63-day periods, and for the duplicate samples) were chosen for statistical analyses because LI values satisfy the “constant variance” assumption of ordinary least squares fitting methods used in the statistical analyses better than the diffusivity values.

Table D-7. Sodium Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	1.53E-09	1.83E-07	3.84E-07	3.81E-07	3.45E-07	1.19E-07	7.57E-07	6.66E-08	8.69E-08	1.82E-07
0.92	1	2.26E-08	1.58E-08	7.63E-08	8.97E-08	9.86E-08	9.11E-08	1.69E-08	3.04E-08	3.60E-08	3.54E-08
1	2	4.32E-09	4.16E-09	3.22E-08	3.09E-08	2.85E-08	2.91E-08	1.86E-08	1.66E-08	3.16E-08	2.96E-08
5	7	1.97E-09	2.17E-09	2.01E-08	1.99E-08	1.37E-08	1.74E-08	5.17E-08	2.80E-08	1.17E-08	2.18E-08
7	14	1.97E-09	1.61E-09	1.36E-08	1.27E-08	9.96E-09	1.01E-08	1.52E-08	1.23E-08	1.37E-08	1.30E-08
14	28	1.42E-09	1.51E-09	8.60E-09	8.43E-09	7.01E-09	6.63E-09	2.31E-08	9.71E-09	3.99E-09	9.95E-09
14	42	1.22E-09	1.21E-09	5.72E-09	1.39E-08	2.40E-09	5.46E-09	7.66E-09	7.09E-09	6.16E-09	6.49E-09
7	49	1.16E-09	1.11E-09	6.01E-09	5.46E-09	5.33E-09	5.22E-09	5.72E-09	7.95E-09	5.16E-09	4.87E-09
14	63	1.00E-09	1.06E-09	3.67E-09	3.55E-09	3.96E-09	3.95E-09	6.19E-09	5.92E-09	3.79E-09	4.11E-09
14	77	1.09E-09	1.02E-09	2.15E-09	3.09E-09	3.81E-09	2.73E-09	4.34E-09	4.51E-09	3.40E-09	2.07E-09
14	91	9.22E-10	9.61E-10	2.51E-09	1.67E-09	3.50E-09	3.04E-09	4.28E-09	4.69E-09	1.86E-09	2.36E-09

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	3.04E-07	2.84E-07	2.05E-08	6.16E-08	1.21E-07	6.20E-08	5.23E-08	3.51E-07	4.01E-07	2.52E-07
0.92	1	5.58E-08	5.43E-08	2.22E-08	2.05E-08	1.17E-08	1.44E-08	3.55E-08	3.54E-08	3.69E-08	4.18E-08
1	2	4.13E-08	4.08E-08	1.87E-08	1.96E-08	6.03E-09	6.30E-09	2.66E-08	2.39E-08	3.00E-08	3.64E-08
5	7	1.73E-08	2.60E-08	1.00E-08	1.07E-08	4.22E-09	3.85E-09	1.33E-08	1.11E-08	1.71E-08	2.18E-08
7	14	1.39E-08	1.43E-08	8.66E-09	8.32E-09	3.91E-09	3.55E-09	6.04E-09	7.81E-09	2.53E-08	1.69E-08
14	28	1.00E-08	1.07E-08	4.93E-09	6.07E-09	2.80E-09	2.79E-09	5.85E-09	5.73E-09	1.02E-08	1.07E-08
14	42	6.81E-09	6.62E-09	3.38E-09	3.74E-09	2.53E-09	2.26E-09	4.55E-09	4.59E-09	7.56E-09	7.56E-09
7	49	4.93E-09	5.70E-09	2.77E-09	3.13E-09	2.35E-09	2.33E-09	4.44E-09	4.43E-09	6.44E-09	6.98E-09
14	63	3.50E-09	3.63E-09	1.97E-09	2.14E-09	2.11E-09	2.05E-09	3.76E-09	3.71E-09	5.35E-09	5.42E-09
14	77	1.42E-09	1.99E-09	9.43E-10	1.23E-09	1.65E-09	1.95E-09	2.52E-09	1.84E-09	4.07E-09	3.70E-09
14	91	1.15E-09	1.70E-09	1.08E-09	1.44E-09	1.06E-09	1.38E-09	2.50E-09	2.81E-09	3.32E-09	3.37E-09

Table D-7. Sodium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	7.50E-07	1.73E-07	5.50E-08	2.55E-08	7.17E-08	2.57E-07	1.10E-07	3.12E-07	3.97E-08	3.81E-08
0.92	1	4.91E-08	4.87E-08	7.87E-09	8.15E-09	4.02E-08	3.62E-08	2.80E-08	2.92E-08	3.38E-08	3.01E-08
1	2	3.36E-08	3.49E-08	6.30E-09	6.81E-09	3.28E-08	3.14E-08	2.38E-08	2.65E-08	2.83E-08	2.70E-08
5	7	1.88E-08	1.84E-08	5.46E-09	4.56E-09	2.04E-08	1.89E-08	1.34E-08	1.49E-08	1.40E-08	1.54E-08
7	14	1.10E-08	1.26E-08	4.38E-09	4.86E-09	1.37E-08	1.27E-08	8.89E-09	1.00E-08	1.24E-08	1.33E-08
14	28	8.05E-09	7.20E-09	4.08E-09	3.73E-09	8.86E-09	8.81E-09	5.65E-09	6.00E-09	9.50E-09	9.10E-09
14	42	6.35E-09	6.53E-09	4.21E-09	4.15E-09	7.04E-09	6.25E-09	4.82E-09	4.64E-09	8.22E-09	7.20E-09
7	49	5.55E-09	5.92E-09	4.02E-09	4.04E-09	6.18E-09	6.10E-09	4.06E-09	4.31E-09	7.45E-09	6.14E-09
14	63	4.43E-09	5.29E-09	4.09E-09	3.77E-09	4.16E-09	4.13E-09	3.04E-09	3.06E-09	5.88E-09	5.18E-09
14	77	2.26E-09	3.43E-09	3.31E-09	4.08E-09	3.59E-09	3.09E-09	2.42E-09	2.17E-09	3.07E-09	3.90E-09
14	91	1.64E-09	2.31E-09	3.57E-09	2.23E-09	2.90E-09	2.38E-09	1.55E-09	1.56E-09	3.03E-09	2.16E-09

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		16a	16b	17a	17b	18a	18b	19a	19b	20a	20b
0.08	0.08	4.39E-08	2.62E-08	7.81E-08	4.89E-08	2.37E-08	2.20E-08	5.85E-08	5.69E-08	3.14E-08	3.07E-08
0.92	1	2.44E-08	2.37E-08	3.81E-08	3.96E-08	9.93E-09	1.04E-08	3.43E-08	3.42E-08	1.65E-08	1.87E-08
1	2	1.98E-08	1.97E-08	3.78E-08	3.42E-08	9.23E-09	9.36E-09	2.57E-08	2.48E-08	1.54E-08	1.96E-08
5	7	1.10E-08	1.01E-08	1.76E-08	1.67E-08	4.65E-09	4.36E-09	1.16E-08	1.09E-08	1.93E-08	1.57E-08
7	14	1.15E-08	1.21E-08	1.29E-08	1.69E-08	5.18E-09	4.88E-09	9.25E-09	1.02E-08	2.51E-08	1.25E-08
14	28	8.36E-09	8.94E-09	9.47E-09	9.14E-09	3.04E-09	3.22E-09	6.09E-09	5.18E-09	1.18E-08	6.55E-09
14	42	7.00E-09	7.63E-09	6.61E-09	6.98E-09	2.80E-09	2.89E-09	3.68E-09	3.64E-09	9.35E-09	6.76E-09
7	49	6.49E-09	6.69E-09	5.92E-09	6.30E-09	2.76E-09	2.85E-09	2.54E-09	2.78E-09	9.38E-09	6.38E-09
14	63	5.75E-09	5.91E-09	4.60E-09	4.33E-09	2.70E-09	2.46E-09	1.71E-09	2.01E-09	7.50E-09	6.51E-09
14	77	2.31E-09	3.40E-09	3.53E-09	3.44E-09	1.75E-09	2.12E-09	1.01E-09	1.11E-09	3.72E-09	5.16E-09
14	91	3.45E-09	3.45E-09	2.92E-09	2.65E-09	1.82E-09	2.26E-09	8.68E-10	9.77E-10	4.03E-09	5.03E-09

Table D-7. Sodium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Sodium Effective Diffusion Coefficient (cm ² /s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	1.50E-07	1.44E-07	5.69E-08	6.27E-08	9.95E-08	1.07E-07
0.92	1	9.58E-08	9.94E-08	3.55E-08	3.30E-08	1.01E-07	1.02E-07
1	2	7.57E-08	8.11E-08	2.71E-08	2.59E-08	1.02E-07	1.11E-07
5	7	3.51E-08	3.79E-08	1.14E-08	1.19E-08	5.96E-08	7.56E-08
7	14	3.10E-08	3.22E-08	9.27E-09	8.16E-09	5.07E-08	5.46E-08
14	28	1.53E-08	1.47E-08	5.49E-09	5.77E-09	3.38E-08	3.40E-08
14	42	1.13E-08	1.10E-08	5.17E-09	4.93E-09	2.42E-08	2.87E-08
7	49	9.14E-09	9.31E-09	4.61E-09	4.76E-09	2.30E-08	2.56E-08
14	63	6.46E-09	6.65E-09	3.92E-09	4.09E-09	1.72E-08	2.02E-08
14	77	2.81E-09	2.87E-09	2.91E-09	1.94E-09	8.71E-09	1.01E-08
14	91	2.70E-09	2.66E-09	3.82E-09	1.94E-09	6.16E-09	6.26E-09

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

Table D-8. Nitrate Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	4.73E-08	9.87E-08	4.41E-07	7.64E-07	3.41E-07	7.12E-09	1.83E-07	1.95E-07	4.06E-07	3.95E-07
0.92	1	2.33E-08	1.70E-08	1.03E-07	1.16E-07	9.85E-08	1.11E-07	1.66E-08	4.16E-08	4.13E-08	4.47E-08
1	2	3.83E-09	3.28E-09	4.21E-08	4.09E-08	2.76E-08	2.82E-08	1.83E-08	1.49E-08	3.39E-08	3.56E-08
5	7	2.62E-09	2.47E-09	3.17E-08	2.84E-08	1.89E-08	1.67E-08	3.95E-08	4.37E-08	2.95E-08	3.07E-08
7	14	1.53E-09	1.41E-09	1.90E-08	1.80E-08	9.65E-09	8.81E-09	1.47E-08	1.43E-08	1.74E-08	1.72E-08
14	28	1.19E-09	1.08E-09	1.08E-08	1.07E-08	5.58E-09	5.64E-09	7.78E-09	7.53E-09	1.17E-08	1.15E-08
14	42	6.07E-10	8.23E-10	5.21E-09	5.85E-09	4.24E-09	4.64E-09	5.59E-09	4.62E-09	5.17E-09	5.14E-09
7	49	7.97E-10	8.49E-10	5.80E-09	5.49E-09	4.34E-09	4.12E-09	4.50E-09	5.36E-09	4.81E-09	5.24E-09
14	63	7.93E-10	8.30E-10	4.81E-09	4.59E-09	4.12E-09	4.12E-09	4.51E-09	4.33E-09	3.78E-09	4.16E-09
14	77	7.79E-10	8.00E-10	2.60E-09	3.51E-09	3.60E-09	2.39E-09	2.71E-09	3.08E-09	2.59E-09	1.52E-09
14	91	7.54E-10	8.04E-10	3.07E-09	1.98E-09	3.37E-09	2.90E-09	3.19E-09	3.63E-09	1.46E-09	2.10E-09

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	9.11E-07	1.15E-06	4.18E-07	1.31E-07	5.02E-07	7.07E-07	2.06E-07	2.52E-07	4.46E-07	2.06E-07
0.92	1	8.02E-08	8.43E-08	3.46E-08	3.45E-08	1.28E-08	1.51E-08	3.55E-08	3.40E-08	4.01E-08	4.84E-08
1	2	5.80E-08	5.97E-08	2.97E-08	2.85E-08	6.23E-09	6.78E-09	2.21E-08	2.30E-08	3.25E-08	3.85E-08
5	7	4.65E-08	4.49E-08	1.94E-08	2.09E-08	5.98E-09	5.28E-09	1.31E-08	1.27E-08	2.54E-08	2.96E-08
7	14	2.60E-08	2.64E-08	1.18E-08	1.20E-08	4.31E-09	4.07E-09	6.95E-09	7.11E-09	1.47E-08	1.60E-08
14	28	1.61E-08	1.72E-08	8.08E-09	1.01E-08	3.35E-09	3.23E-09	5.42E-09	5.29E-09	9.80E-09	9.35E-09
14	42	7.80E-09	9.43E-09	3.22E-09	4.06E-09	2.27E-09	1.78E-09	2.73E-09	3.40E-09	5.20E-09	5.82E-09
7	49	9.30E-09	8.09E-09	3.23E-09	4.10E-09	2.85E-09	2.36E-09	4.41E-09	3.48E-09	5.60E-09	4.60E-09
14	63	4.96E-09	5.48E-09	2.37E-09	2.66E-09	2.53E-09	2.41E-09	3.94E-09	4.10E-09	4.28E-09	4.04E-09
14	77	1.65E-09	2.47E-09	9.46E-10	1.26E-09	1.87E-09	2.16E-09	2.42E-09	1.73E-09	2.66E-09	2.35E-09
14	91	1.45E-09	2.16E-09	1.14E-09	1.85E-09	1.19E-09	1.59E-09	3.41E-09	3.24E-09	2.62E-09	2.86E-09

Table D-8. Nitrate Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	1.98E-07	4.51E-07	1.45E-07	1.44E-07	3.15E-07	4.43E-07	4.71E-07	4.18E-07	5.33E-08	4.24E-08
0.92	1	5.34E-08	5.48E-08	8.70E-09	7.75E-09	5.97E-08	5.58E-08	4.43E-08	4.81E-08	4.11E-08	3.76E-08
1	2	3.56E-08	3.64E-08	6.22E-09	6.20E-09	4.25E-08	4.31E-08	3.82E-08	4.17E-08	3.10E-08	3.05E-08
5	7	2.15E-08	2.43E-08	7.51E-09	5.91E-09	3.53E-08	3.36E-08	3.02E-08	3.19E-08	2.27E-08	2.23E-08
7	14	1.20E-08	1.11E-08	4.79E-09	5.41E-09	2.18E-08	2.21E-08	1.64E-08	1.86E-08	1.61E-08	1.63E-08
14	28	9.76E-09	1.02E-08	4.16E-09	4.46E-09	1.42E-08	1.37E-08	9.71E-09	1.01E-08	1.08E-08	1.06E-08
14	42	7.98E-09	7.40E-09	3.48E-09	3.35E-09	8.74E-09	6.65E-09	5.05E-09	4.29E-09	4.20E-09	5.15E-09
7	49	8.06E-09	9.15E-09	4.13E-09	4.32E-09	8.11E-09	7.26E-09	6.60E-09	6.26E-09	8.27E-09	6.45E-09
14	63	7.48E-09	8.79E-09	4.05E-09	3.58E-09	5.73E-09	5.69E-09	3.93E-09	4.07E-09	5.83E-09	4.71E-09
14	77	3.57E-09	4.97E-09	2.92E-09	3.64E-09	4.18E-09	3.31E-09	2.77E-09	2.41E-09	3.03E-09	3.90E-09
14	91	2.69E-09	4.02E-09	3.91E-09	2.20E-09	3.87E-09	3.21E-09	1.94E-09	1.89E-09	2.53E-09	1.98E-09

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		16a	16b	17a	17b	18a	18b	19a	19b	20a	20b
0.08	0.08	5.60E-08	5.08E-08	7.20E-08	6.84E-08	3.33E-08	3.46E-08	8.30E-08	8.00E-08	3.86E-08	3.45E-08
0.92	1	3.16E-08	3.13E-08	5.93E-08	5.90E-08	1.50E-08	1.51E-08	5.07E-08	5.17E-08	1.54E-08	2.01E-08
1	2	2.69E-08	2.75E-08	5.22E-08	4.78E-08	1.23E-08	1.25E-08	3.71E-08	3.54E-08	1.25E-08	2.15E-08
5	7	1.67E-08	1.63E-08	2.95E-08	2.92E-08	7.84E-09	7.75E-09	1.94E-08	2.10E-08	2.73E-08	2.46E-08
7	14	1.38E-08	1.53E-08	1.84E-08	1.92E-08	5.50E-09	5.39E-09	1.20E-08	1.42E-08	2.68E-08	1.18E-08
14	28	1.03E-08	1.20E-08	1.15E-08	1.21E-08	3.89E-09	4.00E-09	8.90E-09	8.47E-09	1.00E-08	6.59E-09
14	42	6.49E-09	6.38E-09	5.57E-09	7.08E-09	2.47E-09	3.04E-09	4.37E-09	3.76E-09	5.69E-09	4.57E-09
7	49	8.14E-09	7.95E-09	6.86E-09	7.07E-09	3.44E-09	3.44E-09	3.60E-09	4.08E-09	6.47E-09	4.84E-09
14	63	6.62E-09	6.70E-09	5.72E-09	5.79E-09	3.16E-09	3.08E-09	2.34E-09	2.75E-09	5.58E-09	4.71E-09
14	77	2.78E-09	4.21E-09	4.82E-09	4.93E-09	2.42E-09	2.86E-09	1.38E-09	1.54E-09	2.92E-09	4.02E-09
14	91	4.02E-09	3.77E-09	3.83E-09	3.48E-09	2.23E-09	2.80E-09	1.00E-09	1.14E-09	2.73E-09	3.67E-09

Table D-8. Nitrate Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrate Effective Diffusion Coefficient (cm ² /s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	2.06E-07	2.01E-07	6.67E-08	7.24E-08	1.48E-07	1.48E-07
0.92	1	1.31E-07	1.28E-07	4.70E-08	4.19E-08	1.69E-07	1.74E-07
1	2	1.06E-07	1.06E-07	2.83E-08	2.77E-08	1.67E-07	1.77E-07
5	7	6.62E-08	6.10E-08	1.40E-08	1.32E-08	1.15E-07	1.36E-07
7	14	4.41E-08	3.79E-08	8.10E-09	7.99E-09	7.37E-08	8.89E-08
14	28	2.42E-08	2.31E-08	5.36E-09	5.33E-09	4.86E-08	5.75E-08
14	42	1.47E-08	1.20E-08	4.58E-09	3.18E-09	4.12E-08	3.66E-08
7	49	1.37E-08	1.37E-08	3.84E-09	4.38E-09	2.86E-08	3.37E-08
14	63	9.83E-09	9.97E-09	3.77E-09	4.06E-09	2.29E-08	2.84E-08
14	77	4.28E-09	4.36E-09	3.11E-09	1.76E-09	1.21E-08	1.35E-08
14	91	3.63E-09	3.44E-09	3.51E-09	1.74E-09	7.59E-09	7.62E-09

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

Table D-9. Nitrite Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	5.31E-08	8.63E-08	2.65E-07	8.01E-07	3.70E-07	5.93E-09	9.52E-08	1.01E-07	4.06E-07	3.91E-07
0.92	1	2.09E-08	1.55E-08	7.98E-08	9.17E-08	8.18E-08	9.12E-08	1.42E-08	3.45E-08	3.42E-08	3.68E-08
1	2	4.16E-09	3.59E-09	4.26E-08	4.14E-08	3.10E-08	3.15E-08	1.97E-08	1.63E-08	3.52E-08	3.70E-08
5	7	3.17E-09	2.97E-09	3.35E-08	2.95E-08	2.05E-08	1.84E-08	4.10E-08	4.40E-08	2.89E-08	2.99E-08
7	14	1.61E-09	1.52E-09	1.86E-08	1.74E-08	1.01E-08	8.83E-09	1.41E-08	1.38E-08	1.49E-08	1.52E-08
14	28	1.50E-09	1.33E-09	1.44E-08	1.34E-08	7.52E-09	7.15E-09	9.23E-09	8.64E-09	1.13E-08	1.09E-08
14	42	7.41E-10	9.77E-10	5.55E-09	6.37E-09	4.95E-09	5.30E-09	6.71E-09	5.37E-09	5.26E-09	5.11E-09
7	49	8.75E-10	9.37E-10	5.45E-09	5.11E-09	4.54E-09	4.37E-09	4.68E-09	5.59E-09	4.63E-09	5.08E-09
14	63	8.67E-10	9.16E-10	4.99E-09	4.65E-09	4.93E-09	4.73E-09	4.78E-09	4.48E-09	3.57E-09	3.92E-09
14	77	9.02E-10	8.45E-10	2.52E-09	3.31E-09	4.27E-09	2.80E-09	2.90E-09	3.00E-09	2.47E-09	1.39E-09
14	91	8.95E-10	9.34E-10	3.12E-09	2.10E-09	4.69E-09	3.95E-09	3.68E-09	4.16E-09	1.57E-09	2.23E-09

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	8.91E-07	1.06E-06	2.11E-07	8.54E-08	2.70E-07	3.90E-07	2.45E-07	2.92E-07	2.22E-07	1.18E-07
0.92	1	6.55E-08	6.83E-08	3.22E-08	3.19E-08	1.06E-08	1.22E-08	3.31E-08	3.18E-08	3.48E-08	4.17E-08
1	2	6.34E-08	6.50E-08	3.49E-08	3.42E-08	6.29E-09	6.65E-09	2.80E-08	2.86E-08	3.51E-08	4.18E-08
5	7	4.67E-08	4.47E-08	2.10E-08	2.37E-08	6.37E-09	5.64E-09	1.61E-08	1.55E-08	2.60E-08	3.12E-08
7	14	2.29E-08	2.40E-08	1.18E-08	1.18E-08	3.93E-09	3.79E-09	7.67E-09	7.74E-09	1.31E-08	1.39E-08
14	28	1.62E-08	1.67E-08	7.88E-09	9.45E-09	3.02E-09	2.92E-09	6.30E-09	5.93E-09	8.41E-09	8.10E-09
14	42	8.20E-09	9.92E-09	3.36E-09	4.30E-09	2.27E-09	1.73E-09	3.69E-09	4.43E-09	5.11E-09	5.80E-09
7	49	9.24E-09	8.30E-09	3.30E-09	4.15E-09	2.74E-09	2.35E-09	5.88E-09	4.53E-09	5.30E-09	4.44E-09
14	63	4.99E-09	5.37E-09	2.34E-09	2.66E-09	2.30E-09	2.16E-09	4.86E-09	4.88E-09	3.85E-09	3.45E-09
14	77	1.59E-09	2.44E-09	9.12E-10	1.22E-09	1.73E-09	2.04E-09	3.19E-09	2.30E-09	2.54E-09	2.15E-09
14	91	1.55E-09	2.35E-09	1.20E-09	1.96E-09	1.16E-09	1.54E-09	4.63E-09	4.99E-09	2.48E-09	2.71E-09

Table D-9. Nitrite Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	2.13E-07	4.46E-07	9.15E-08	8.75E-08	1.52E-07	2.11E-07	2.26E-07	2.21E-07	6.00E-08	4.76E-08
0.92	1	4.76E-08	4.83E-08	8.12E-09	7.20E-09	4.85E-08	4.54E-08	4.17E-08	4.44E-08	4.56E-08	4.22E-08
1	2	4.36E-08	4.47E-08	6.66E-09	6.67E-09	4.36E-08	4.42E-08	4.51E-08	4.88E-08	3.58E-08	3.61E-08
5	7	2.44E-08	2.66E-08	7.92E-09	6.23E-09	3.45E-08	3.27E-08	3.37E-08	3.55E-08	2.06E-08	2.02E-08
7	14	1.25E-08	1.15E-08	4.24E-09	4.76E-09	1.85E-08	1.83E-08	1.55E-08	1.66E-08	2.02E-08	1.88E-08
14	28	9.93E-09	1.02E-08	3.48E-09	3.71E-09	1.13E-08	1.08E-08	8.60E-09	9.01E-09	1.22E-08	1.03E-08
14	42	9.33E-09	8.81E-09	3.41E-09	3.26E-09	7.92E-09	6.19E-09	5.21E-09	4.43E-09	4.79E-09	5.73E-09
7	49	1.01E-08	1.16E-08	4.07E-09	4.23E-09	7.57E-09	6.74E-09	6.66E-09	6.36E-09	7.69E-09	6.04E-09
14	63	8.17E-09	9.58E-09	3.57E-09	3.15E-09	4.72E-09	4.48E-09	3.45E-09	3.69E-09	5.96E-09	4.66E-09
14	77	4.31E-09	6.05E-09	2.54E-09	3.36E-09	3.52E-09	2.78E-09	2.61E-09	2.23E-09	3.29E-09	4.18E-09
14	91	3.77E-09	5.41E-09	3.60E-09	1.95E-09	3.31E-09	2.84E-09	1.81E-09	1.80E-09	3.00E-09	2.31E-09

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

Table D-9. Nitrite Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Nitrite Effective Diffusion Coefficient (cm ² /s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	2.18E-07	2.16E-07	7.53E-08	8.24E-08	1.55E-07	1.56E-07
0.92	1	1.41E-07	1.40E-07	5.14E-08	4.65E-08	1.53E-07	1.66E-07
1	2	1.27E-07	1.19E-07	3.38E-08	3.30E-08	1.74E-07	1.87E-07
5	7	5.64E-08	5.10E-08	1.35E-08	1.29E-08	8.58E-08	9.79E-08
7	14	4.35E-08	3.73E-08	8.03E-09	8.01E-09	6.11E-08	7.51E-08
14	28	2.01E-08	1.90E-08	4.49E-09	4.57E-09	3.23E-08	3.65E-08
14	42	1.55E-08	1.25E-08	4.76E-09	3.40E-09	2.66E-08	2.88E-08
7	49	1.23E-08	1.28E-08	3.86E-09	4.35E-09	2.27E-08	2.62E-08
14	63	8.80E-09	9.35E-09	3.70E-09	3.99E-09	1.73E-08	2.06E-08
14	77	4.40E-09	4.42E-09	3.32E-09	1.88E-09	1.03E-08	1.05E-08
14	91	3.79E-09	3.58E-09	3.76E-09	1.87E-09	6.57E-09	6.15E-09

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

Table D-10. Iodine Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	1.19E-09	5.41E-08	1.83E-07	5.25E-07	3.53E-07	6.48E-07	8.16E-08	1.43E-07	3.28E-07	4.25E-07
0.92	1	1.71E-08	1.71E-08	1.05E-07	1.25E-07	1.33E-07	1.36E-07	1.27E-08	2.27E-08	3.77E-08	3.91E-08
1	2	3.15E-09	2.82E-09	3.99E-08	3.91E-08	3.39E-08	3.32E-08	1.31E-08	1.14E-08	3.45E-08	3.43E-08
5	7	1.67E-09	1.63E-09	2.50E-08	2.42E-08	1.76E-08	1.71E-08	2.41E-08	2.45E-08	2.33E-08	2.32E-08
7	14	1.05E-09	1.03E-09	1.64E-08	1.57E-08	9.73E-09	8.68E-09	9.18E-09	8.85E-09	1.44E-08	1.45E-08
14	28	8.53E-10	8.51E-10	1.09E-08	1.05E-08	6.32E-09	6.06E-09	5.38E-09	5.37E-09	9.32E-09	9.48E-09
14	42	7.01E-10	7.04E-10	7.17E-09	6.97E-09	4.72E-09	5.51E-09	4.01E-09	3.70E-09	5.23E-09	5.31E-09
7	49	6.18E-10	6.04E-10	5.79E-09	5.40E-09	4.54E-09	4.46E-09	3.02E-09	3.44E-09	3.55E-09	3.70E-09
14	63	6.00E-10	6.09E-10	4.76E-09	4.72E-09	4.18E-09	4.08E-09	3.19E-09	3.29E-09	2.77E-09	2.94E-09
14	77	5.88E-10	6.01E-10	2.61E-09	3.54E-09	3.59E-09	2.34E-09	2.06E-09	2.32E-09	1.74E-09	1.07E-09
14	91	5.70E-10	6.02E-10	2.97E-09	1.85E-09	3.17E-09	2.69E-09	2.34E-09	2.70E-09	9.51E-10	1.32E-09

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	8.06E-07	5.82E-07	6.90E-09	1.60E-07	1.07E-07	6.37E-08	5.66E-07	4.46E-07	3.04E-07	1.60E-07
0.92	1	9.52E-08	9.67E-08	3.77E-08	3.68E-08	1.17E-08	1.29E-08	4.78E-08	4.77E-08	2.83E-08	3.32E-08
1	2	7.19E-08	7.27E-08	3.21E-08	2.99E-08	6.18E-09	6.14E-09	3.03E-08	2.84E-08	2.29E-08	2.80E-08
5	7	4.60E-08	4.55E-08	1.90E-08	1.91E-08	4.74E-09	4.34E-09	1.45E-08	1.40E-08	1.65E-08	1.94E-08
7	14	2.68E-08	2.82E-08	1.12E-08	1.12E-08	3.79E-09	3.54E-09	8.01E-09	8.20E-09	1.06E-08	1.18E-08
14	28	1.75E-08	1.78E-08	7.09E-09	8.36E-09	3.19E-09	3.03E-09	6.41E-09	6.05E-09	6.66E-09	6.93E-09
14	42	1.05E-08	1.07E-08	3.93E-09	4.53E-09	2.68E-09	2.44E-09	5.08E-09	5.01E-09	4.47E-09	4.49E-09
7	49	6.99E-09	7.52E-09	2.68E-09	2.97E-09	2.42E-09	2.27E-09	4.70E-09	4.91E-09	3.58E-09	3.42E-09
14	63	4.74E-09	5.29E-09	2.02E-09	2.37E-09	2.22E-09	2.13E-09	4.62E-09	4.75E-09	3.08E-09	2.78E-09
14	77	1.46E-09	2.29E-09	8.01E-10	1.05E-09	1.68E-09	1.82E-09	2.77E-09	2.00E-09	1.93E-09	1.60E-09
14	91	1.14E-09	1.74E-09	9.78E-10	1.47E-09	1.07E-09	1.40E-09	3.22E-09	3.70E-09	1.91E-09	1.92E-09

Table D-10. Iodine Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	2.77E-07	4.74E-07	1.17E-08	3.67E-08	1.79E-07	3.77E-07	3.40E-07	2.80E-07	3.15E-08	2.89E-08
0.92	1	7.79E-08	7.97E-08	6.32E-09	6.32E-09	5.82E-08	5.44E-08	5.45E-08	5.85E-08	2.83E-08	2.58E-08
1	2	5.25E-08	5.17E-08	4.66E-09	4.79E-09	4.12E-08	4.01E-08	4.29E-08	4.50E-08	2.44E-08	2.33E-08
5	7	2.93E-08	2.94E-08	4.90E-09	4.04E-09	2.91E-08	2.74E-08	2.77E-08	2.98E-08	1.87E-08	1.87E-08
7	14	1.63E-08	1.60E-08	3.50E-09	3.84E-09	2.01E-08	1.95E-08	1.64E-08	1.84E-08	1.35E-08	1.30E-08
14	28	1.33E-08	1.32E-08	2.77E-09	3.12E-09	1.30E-08	1.24E-08	8.85E-09	9.05E-09	8.83E-09	8.26E-09
14	42	1.24E-08	1.29E-08	2.77E-09	2.74E-09	9.18E-09	8.54E-09	6.13E-09	5.94E-09	6.13E-09	5.52E-09
7	49	1.11E-08	1.24E-08	2.53E-09	2.56E-09	6.94E-09	6.54E-09	4.43E-09	4.56E-09	5.22E-09	4.16E-09
14	63	9.97E-09	1.19E-08	2.72E-09	2.54E-09	5.75E-09	5.50E-09	3.24E-09	3.40E-09	4.03E-09	3.20E-09
14	77	4.74E-09	6.64E-09	2.03E-09	2.64E-09	4.04E-09	3.30E-09	2.25E-09	2.01E-09	2.04E-09	2.52E-09
14	91	3.74E-09	5.43E-09	2.68E-09	1.62E-09	3.86E-09	3.12E-09	1.66E-09	1.63E-09	1.85E-09	1.37E-09

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

Table D-10. Iodine Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Iodine Effective Diffusion Coefficient (cm ² /s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	1.69E-07	1.80E-07	5.79E-08	6.40E-08	1.42E-07	1.45E-07
0.92	1	1.55E-07	1.51E-07	5.16E-08	4.42E-08	1.72E-07	1.79E-07
1	2	1.18E-07	1.19E-07	2.88E-08	2.69E-08	1.72E-07	1.91E-07
5	7	7.55E-08	7.27E-08	1.44E-08	1.39E-08	1.16E-07	1.41E-07
7	14	5.16E-08	4.92E-08	8.49E-09	8.38E-09	7.92E-08	9.82E-08
14	28	2.82E-08	2.79E-08	5.78E-09	5.53E-09	4.87E-08	5.94E-08
14	42	1.73E-08	1.75E-08	4.56E-09	4.40E-09	3.20E-08	3.84E-08
7	49	1.25E-08	1.28E-08	3.79E-09	3.99E-09	2.65E-08	2.97E-08
14	63	9.08E-09	9.11E-09	3.61E-09	3.71E-09	2.00E-08	2.41E-08
14	77	4.09E-09	3.97E-09	3.07E-09	1.68E-09	1.17E-08	1.18E-08
14	91	3.40E-09	3.23E-09	3.28E-09	1.69E-09	6.98E-09	5.81E-09

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

Table D-11. Technetium Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	3.67E-11	6.45E-09	2.60E-09	2.76E-09	2.08E-09	6.86E-10	3.75E-13	2.68E-12	5.24E-11	5.69E-11
0.92	1	5.25E-10	4.28E-10	4.06E-10	5.75E-10	4.04E-10	4.04E-10	2.91E-12	5.77E-12	1.08E-11	4.58E-11
1	2	1.39E-10	1.26E-10	9.22E-11	1.18E-10	8.18E-12	7.54E-12	9.30E-12	9.99E-12	2.37E-11	7.44E-11
5	7	1.12E-10	1.05E-10	4.79E-11	5.02E-11	1.26E-11	1.33E-11	1.47E-11	1.62E-11	2.76E-11	5.51E-11
7	14	7.50E-11	7.22E-11	5.37E-11	4.67E-11	2.51E-11	2.88E-11	2.05E-11	1.73E-11	5.34E-11	6.58E-11
14	28	5.19E-11	5.03E-11	7.06E-11	5.56E-11	3.13E-11	3.02E-11	1.92E-11	2.51E-11	5.00E-11	5.57E-11
14	42	4.22E-11	3.97E-11	7.23E-11	5.89E-11	3.41E-11	3.49E-11	2.28E-11	2.57E-11	8.41E-11	7.96E-11
7	49	4.43E-11	4.15E-11	6.59E-11	5.13E-11	3.61E-11	3.82E-11	2.90E-11	3.65E-11	8.58E-11	8.77E-11
14	63	5.14E-11	5.07E-11	5.86E-11	4.56E-11	3.71E-11	3.56E-11	3.05E-11	3.72E-11	6.44E-11	6.44E-11
14	77	4.89E-11	4.87E-11	3.32E-11	3.60E-11	3.29E-11	2.13E-11	2.28E-11	2.93E-11	3.73E-11	2.18E-11
14	91	4.39E-11	4.56E-11	3.89E-11	1.90E-11	3.01E-11	2.46E-11	2.66E-11	3.92E-11	2.16E-11	2.73E-11

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	2.44E-09	3.30E-09	1.09E-12	8.96E-12	2.19E-09	1.25E-09	8.99E-12	1.96E-10	7.07E-09	6.91E-09
0.92	1	5.49E-11	3.22E-11	1.03E-11	1.06E-11	2.28E-10	3.95E-10	2.40E-11	3.76E-11	1.11E-09	2.01E-09
1	2	7.61E-12	5.60E-12	9.74E-12	1.25E-11	2.51E-10	5.10E-10	5.74E-11	5.58E-11	5.99E-10	1.03E-09
5	7	7.09E-12	6.36E-12	7.97E-12	1.22E-11	2.89E-10	4.43E-10	3.74E-11	3.73E-11	3.03E-10	4.57E-10
7	14	1.80E-11	1.80E-11	7.91E-12	1.09E-11	2.75E-10	3.15E-10	4.73E-11	4.21E-11	1.99E-10	2.42E-10
14	28	4.22E-11	4.08E-11	8.74E-12	9.66E-12	2.22E-10	2.23E-10	2.98E-11	3.03E-11	1.40E-10	1.49E-10
14	42	1.26E-10	1.24E-10	9.22E-12	8.75E-12	2.21E-10	2.03E-10	3.92E-11	3.79E-11	1.13E-10	1.19E-10
7	49	2.52E-10	2.35E-10	9.51E-12	1.01E-11	2.20E-10	2.08E-10	4.16E-11	3.84E-11	1.16E-10	1.16E-10
14	63	1.96E-10	2.01E-10	8.40E-12	8.17E-12	2.26E-10	2.03E-10	3.20E-11	2.86E-11	1.15E-10	1.11E-10
14	77	8.05E-11	1.07E-10	3.58E-12	4.90E-12	1.62E-10	1.75E-10	1.88E-11	1.29E-11	8.20E-11	7.17E-11
14	91	8.11E-11	1.20E-10	5.85E-12	7.67E-12	8.25E-11	1.18E-10	2.14E-11	2.40E-11	8.35E-11	8.80E-11

Table D-11. Technetium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	5.09E-10	6.94E-11	1.16E-10	6.57E-11	7.59E-11	3.00E-10	4.64E-11	1.07E-10	2.30E-09	1.45E-09
0.92	1	3.75E-11	2.59E-11	4.13E-11	6.35E-11	7.16E-11	5.37E-11	1.87E-11	1.65E-11	9.35E-10	5.62E-10
1	2	4.43E-11	3.62E-11	4.47E-11	6.11E-11	2.20E-11	2.62E-11	1.94E-11	1.80E-11	3.02E-10	2.20E-10
5	7	1.90E-11	1.75E-11	4.56E-11	5.31E-11	2.26E-11	2.89E-11	1.32E-11	1.47E-11	1.78E-10	1.86E-10
7	14	2.96E-11	4.02E-11	3.79E-11	4.53E-11	5.13E-11	5.34E-11	9.07E-12	1.24E-11	1.25E-10	1.33E-10
14	28	5.01E-11	4.41E-11	3.91E-11	4.19E-11	7.94E-11	6.46E-11	7.03E-12	1.12E-11	5.98E-11	6.16E-11
14	42	8.84E-11	1.08E-10	4.10E-11	4.05E-11	9.71E-11	7.90E-11	8.44E-12	1.13E-11	4.27E-11	4.80E-11
7	49	1.84E-10	1.81E-10	4.13E-11	4.44E-11	9.73E-11	8.59E-11	9.40E-12	1.17E-11	4.37E-11	4.55E-11
14	63	1.97E-10	1.88E-10	4.46E-11	4.33E-11	8.46E-11	7.42E-11	7.54E-12	1.02E-11	3.43E-11	3.36E-11
14	77	8.95E-11	1.11E-10	3.36E-11	4.51E-11	6.10E-11	5.11E-11	7.28E-12	7.33E-12	1.73E-11	2.86E-11
14	91	7.67E-11	1.11E-10	3.81E-11	2.24E-11	5.73E-11	5.01E-11	5.35E-12	5.78E-12	1.81E-11	1.34E-11

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

Table D-11. Technetium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Technetium Effective Diffusion Coefficient (cm ² /s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	1.02E-08	3.05E-09	2.90E-09	2.37E-09	1.72E-09	4.67E-09
0.92	1	8.06E-09	9.79E-10	1.30E-09	4.32E-10	1.30E-09	3.88E-09
1	2	5.32E-10	1.19E-10	1.28E-10	1.49E-11	4.84E-10	1.17E-09
5	7	1.73E-11	1.44E-11	2.91E-11	9.59E-12	3.78E-10	4.58E-10
7	14	2.18E-11	1.41E-11	1.43E-11	1.29E-11	7.21E-10	4.05E-10
14	28	2.69E-11	1.97E-11	1.37E-11	1.68E-11	6.63E-10	4.29E-10
14	42	5.02E-11	4.61E-11	2.01E-11	2.62E-11	1.11E-09	6.90E-10
7	49	1.02E-10	1.61E-10	2.60E-11	3.49E-11	1.91E-09	1.19E-09
14	63	1.20E-10	2.27E-10	2.72E-11	3.36E-11	1.24E-09	8.68E-10
14	77	9.19E-11	2.26E-10	2.47E-11	1.64E-11	7.59E-10	6.01E-10
14	91	1.73E-10	3.52E-10	2.90E-11	1.43E-11	4.12E-10	3.98E-10

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

Table D-12. Chromium Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	6.59E-12	1.47E-09	1.14E-11	1.16E-11	2.30E-12	6.63E-13	1.10E-13	4.58E-13	1.79E-12	2.92E-12
0.92	1	3.76E-11	2.74E-11	2.25E-12	2.57E-12	3.10E-12	2.91E-12	8.12E-14	1.66E-13	1.90E-13	2.02E-12
1	2	9.19E-12	7.82E-12	1.05E-12	1.09E-12	8.56E-13	9.59E-13	1.42E-13	1.42E-13	2.17E-13	2.62E-12
5	7	3.01E-12	2.60E-12	4.43E-13	4.42E-13	2.74E-13	3.12E-13	9.58E-14	9.96E-14	5.77E-14	5.59E-13
7	14	6.76E-13	5.99E-13	1.96E-13	1.92E-13	2.20E-13	2.44E-13	7.27E-14	6.20E-14	4.93E-14	1.14E-13
14	28	2.80E-13	2.42E-13	1.15E-13	1.14E-13	1.68E-13	1.66E-13	6.12E-14	6.52E-14	4.37E-14	6.32E-14
14	42	1.53E-13	1.37E-13	7.04E-14	7.12E-14	1.57E-13	1.50E-13	4.33E-14	4.86E-14	5.77E-14	6.11E-14
7	49	1.91E-13	1.65E-13	8.38E-14	7.31E-14	2.31E-13	2.15E-13	6.64E-14	7.51E-14	1.16E-13	1.19E-13
14	63	1.64E-13	1.64E-13	6.32E-14	6.73E-14	1.09E-13	1.16E-13	6.87E-14	7.44E-14	9.14E-14	8.97E-14
14	77	1.66E-13	1.74E-13	4.59E-14	5.62E-14	1.16E-13	8.57E-14	5.26E-14	5.68E-14	8.16E-14	4.35E-14
14	91	1.79E-13	1.68E-13	5.02E-14	3.05E-14	1.06E-13	9.14E-14	5.85E-14	6.27E-14	6.16E-14	7.96E-14

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	5.72E-13	2.44E-13	<3.82E-14	<4.00E-14	7.57E-10	2.36E-10	2.14E-13	1.42E-11	4.31E-11	4.56E-11
0.92	1	9.85E-14	6.92E-14	2.81E-14	3.19E-14	2.83E-11	4.44E-11	7.08E-13	1.78E-12	6.77E-12	1.23E-11
1	2	8.63E-14	7.43E-14	2.29E-14	3.13E-14	5.22E-12	1.23E-11	1.06E-12	1.87E-12	5.69E-12	1.26E-11
5	7	4.87E-14	4.78E-14	1.11E-14	1.61E-14	1.17E-12	1.33E-12	4.57E-13	7.04E-13	1.96E-12	4.71E-12
7	14	6.39E-14	6.03E-14	7.17E-15	6.74E-15	7.84E-13	9.84E-13	3.27E-13	4.27E-13	8.34E-13	2.19E-12
14	28	8.39E-14	7.83E-14	6.36E-15	6.49E-15	6.52E-13	8.23E-13	1.67E-13	2.16E-13	5.08E-13	1.42E-12
14	42	1.15E-13	1.01E-13	5.32E-15	5.54E-15	5.55E-13	6.25E-13	1.30E-13	1.72E-13	2.97E-13	9.18E-13
7	49	2.73E-13	1.83E-13	1.00E-14	1.14E-14	7.39E-13	8.51E-13	1.72E-13	2.00E-13	3.38E-13	9.56E-13
14	63	1.92E-13	1.96E-13	1.01E-14	1.11E-14	6.90E-13	7.82E-13	9.97E-14	1.20E-13	3.21E-13	8.31E-13
14	77	8.48E-14	1.28E-13	6.13E-15	8.29E-15	5.73E-13	6.43E-13	5.58E-14	4.62E-14	2.30E-13	5.08E-13
14	91	1.11E-13	1.56E-13	7.93E-15	1.13E-14	2.96E-13	5.14E-13	6.05E-14	8.33E-14	2.18E-13	5.69E-13

Table D-12. Chromium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	1.42E-11	1.84E-13	1.70E-12	8.95E-13	8.13E-13	1.38E-12	1.12E-13	2.37E-13	4.49E-12	3.60E-12
0.92	1	6.89E-13	9.16E-14	1.05E-13	1.13E-13	5.99E-13	3.11E-13	6.68E-14	4.76E-14	3.70E-12	3.04E-12
1	2	4.87E-13	1.34E-13	1.90E-13	1.67E-13	5.14E-13	2.48E-13	4.62E-14	3.30E-14	5.23E-12	4.17E-12
5	7	1.56E-13	9.75E-14	1.44E-13	1.75E-13	2.04E-13	1.20E-13	1.87E-14	9.79E-15	5.01E-12	4.15E-12
7	14	8.85E-14	1.01E-13	1.15E-13	2.63E-13	1.00E-13	7.64E-14	9.73E-15	9.00E-15	3.03E-12	2.48E-12
14	28	1.08E-13	1.02E-13	1.09E-13	1.18E-13	6.77E-14	5.51E-14	8.49E-15	8.72E-15	9.09E-13	6.84E-13
14	42	1.07E-13	1.08E-13	1.10E-13	1.13E-13	4.82E-14	3.91E-14	6.88E-15	7.55E-15	2.24E-13	1.59E-13
7	49	1.89E-13	1.80E-13	2.14E-13	5.54E-13	5.38E-14	6.54E-14	1.20E-14	1.11E-14	1.93E-13	1.53E-13
14	63	1.39E-13	1.41E-13	2.21E-13	1.77E-13	5.61E-14	5.54E-14	1.27E-14	1.18E-14	1.10E-13	8.98E-14
14	77	6.03E-14	7.88E-14	1.28E-13	1.70E-13	5.24E-14	4.53E-14	1.57E-14	1.23E-14	1.17E-13	1.04E-13
14	91	5.29E-14	7.67E-14	1.58E-13	8.14E-14	5.16E-14	3.99E-14	1.02E-14	1.05E-14	7.13E-14	4.62E-14

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

Table D-12. Chromium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Chromium Effective Diffusion Coefficient (cm ² /s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	3.60E-13	6.47E-13	8.69E-12	2.47E-12	5.78E-13	1.48E-13
0.92	1	2.87E-13	3.38E-13	5.03E-12	1.68E-12	2.51E-13	1.99E-13
1	2	1.17E-13	1.35E-13	3.38E-12	1.30E-12	6.04E-13	2.71E-13
5	7	4.89E-14	4.60E-14	1.15E-12	5.89E-13	6.17E-13	3.84E-13
7	14	5.52E-14	5.67E-14	4.37E-13	3.42E-13	9.03E-13	5.85E-13
14	28	6.98E-14	8.15E-14	2.21E-13	1.94E-13	9.15E-13	6.96E-13
14	42	1.03E-13	1.09E-13	1.93E-13	1.62E-13	1.01E-12	8.70E-13
7	49	1.99E-13	2.14E-13	2.51E-13	2.16E-13	1.79E-12	1.55E-12
14	63	2.09E-13	2.23E-13	1.08E-13	8.87E-14	1.24E-12	1.19E-12
14	77	1.72E-13	1.91E-13	1.41E-13	6.91E-14	9.21E-13	9.47E-13
14	91	2.39E-13	2.35E-13	1.26E-13	4.48E-14	5.10E-13	5.82E-13

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

Table D-13. Uranium Effective Diffusion Coefficients

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		1a	1b	2a	2b	3a	3b	4a	4b	5a	5b
0.08	0.08	3.02E-15	6.93E-13	5.96E-15	6.62E-15	2.76E-15	9.99E-16	4.66E-16	5.20E-15	1.05E-14	2.46E-15
0.92	1	4.98E-16	4.99E-16	2.81E-16	2.92E-16	6.22E-17	6.58E-17	1.92E-16	1.15E-16	4.68E-17	9.08E-17
1	2	2.33E-14	2.68E-16	3.47E-16	1.96E-16	<5.79E-17	<5.96E-17	1.93E-16	<1.95E-16	1.06E-16	7.25E-17
5	7	8.67E-17	8.16E-17	1.03E-16	9.04E-17	5.62E-16	<1.51E-17	2.21E-16	2.23E-15	7.72E-17	4.94E-17
7	14	9.84E-17	1.48E-16	1.11E-16	9.98E-17	<1.88E-17	<1.94E-17	1.05E-16	<6.32E-17	<2.35E-17	<2.35E-17
14	28	9.72E-17	1.12E-16	7.64E-17	6.51E-17	<9.30E-18	<9.57E-18	<3.10E-17	<3.13E-17	<1.16E-17	<1.16E-17
14	42	1.86E-16	1.97E-16	6.04E-17	5.58E-17	<1.58E-17	<1.62E-17	<5.24E-17	<5.30E-17	<1.97E-17	<1.97E-17
7	49	5.59E-16	6.66E-16	<2.16E-16	<2.11E-16	<8.26E-17	<8.50E-17	<2.74E-16	<2.78E-16	<1.03E-16	<1.03E-16
14	63	3.15E-16	3.37E-16	<6.65E-17	<6.52E-17	<2.55E-17	<2.62E-17	<8.47E-17	<8.56E-17	<3.19E-17	<3.19E-17
14	77	3.84E-16	4.39E-16	<8.31E-17	<8.14E-17	<3.18E-17	<3.25E-17	<1.06E-16	<1.07E-16	<3.98E-17	<3.97E-17
14	91	5.22E-16	5.06E-16	<9.89E-17	<9.69E-17	<3.79E-17	<3.90E-17	<1.26E-16	<1.27E-16	<4.74E-17	<4.67E-17

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		6a	6b	7a	7b	8a	8b	9a	9b	10a	10b
0.08	0.08	6.72E-15	2.93E-15	<1.52E-16	5.67E-16	6.00E-13	6.20E-14	<1.15E-16	5.33E-16	5.82E-14	3.49E-13
0.92	1	1.40E-16	1.04E-16	4.43E-17	3.88E-17	1.63E-15	2.05E-15	<1.89E-17	<1.87E-17	4.16E-16	4.16E-16
1	2	<7.81E-17	<7.94E-17	<7.38E-17	4.71E-14	1.25E-15	1.32E-15	<5.58E-17	<5.50E-17	2.25E-16	1.38E-13
5	7	3.04E-17	6.43E-14	7.02E-16	2.73E-17	2.04E-16	2.26E-16	<1.41E-17	1.72E-17	8.01E-17	8.95E-17
7	14	3.93E-17	2.94E-17	<2.40E-17	<2.52E-17	2.80E-16	2.73E-16	<1.82E-17	<1.79E-17	6.03E-17	8.30E-17
14	28	1.52E-17	1.55E-17	1.42E-17	1.28E-17	1.83E-16	1.89E-16	<8.96E-18	<8.83E-18	4.68E-17	4.79E-17
14	42	<2.13E-17	<2.17E-17	<2.01E-17	<2.11E-17	2.32E-16	2.14E-16	<1.52E-17	<1.50E-17	<3.70E-17	5.53E-17
7	49	2.17E-16	<1.13E-16	<1.05E-16	<1.10E-16	1.03E-15	9.41E-16	<7.96E-17	<7.85E-17	<1.94E-16	<1.96E-16
14	63	<3.44E-17	<3.49E-17	<3.25E-17	<3.40E-17	6.00E-16	4.68E-16	<2.46E-17	<2.42E-17	<5.97E-17	<6.06E-17
14	77	<4.29E-17	<4.36E-17	<4.05E-17	<4.25E-17	5.84E-16	7.19E-16	<3.06E-17	<3.02E-17	<7.45E-17	<7.56E-17
14	91	<5.12E-17	<5.20E-17	<4.84E-17	<5.07E-17	3.37E-16	8.98E-16	<3.66E-17	<3.61E-17	<8.89E-17	<9.02E-17

Table D-13. Uranium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm ² /s)									
		Mix Number and Duplicate (Repeat)									
		11a	11b	12a	12b	13a	13b	14a	14b	15a	15b
0.08	0.08	6.40E-15	1.56E-15	5.76E-15	3.63E-15	1.07E-15	4.61E-15	8.72E-16	3.47E-15	5.89E-16	5.87E-16
0.92	1	2.46E-17	2.72E-17	<4.57E-17	<4.63E-17	2.34E-16	2.91E-16	2.82E-17	3.52E-17	5.56E-17	5.29E-17
1	2	<6.05E-17	<5.96E-17	<1.35E-16	<1.37E-16	2.08E-16	2.37E-16	<7.37E-17	<7.58E-17	<7.47E-17	<7.42E-17
5	7	<1.53E-17	<1.51E-17	<3.41E-17	<3.45E-17	1.36E-16	1.28E-16	<1.87E-17	<1.92E-17	<1.90E-17	<1.90E-17
7	14	<1.98E-17	<1.95E-17	<4.41E-17	8.94E-17	1.09E-16	1.10E-16	2.57E-17	<2.47E-17	<2.40E-17	<2.39E-17
14	28	<9.70E-18	<9.55E-18	<2.16E-17	<2.19E-17	7.14E-17	6.65E-17	1.43E-17	<1.22E-17	<1.20E-17	<1.20E-17
14	42	<1.65E-17	<1.62E-17	<3.67E-17	4.46E-17	4.93E-17	5.02E-17	2.33E-17	<2.07E-17	<2.04E-17	<2.03E-17
7	49	<8.63E-17	<8.50E-17	<1.92E-16	1.09E-15	<2.22E-16	<2.23E-16	<1.05E-16	<1.08E-16	1.08E-16	<1.07E-16
14	63	<2.66E-17	<2.62E-17	<5.92E-17	6.27E-17	<6.83E-17	<6.87E-17	<3.24E-17	<3.33E-17	8.09E-17	<3.27E-17
14	77	<3.32E-17	<3.27E-17	<7.39E-17	9.60E-17	<8.52E-17	<8.56E-17	<4.04E-17	<4.16E-17	7.06E-17	<4.09E-17
14	91	<3.97E-17	<3.91E-17	<8.84E-17	<8.96E-17	<1.02E-16	<1.02E-16	<4.84E-17	<4.97E-17	6.73E-17	<4.90E-17

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

Table D-13. Uranium Effective Diffusion Coefficients (contd)

Interval (days)	Total Duration (days)	Uranium Effective Diffusion Coefficient (cm ² /s)					
		Mix Number and Duplicate (Repeat)					
		21a	21b	22a	22b	23a	23b
0.08	0.08	2.41E-15	1.32E-15	1.67E-16	1.40E-16	2.73E-15	3.57E-15
0.92	1	1.94E-16	1.72E-16	2.72E-17	<1.96E-17	2.66E-16	7.47E-16
1	2	1.01E-16	7.51E-17	<5.67E-17	<5.77E-17	3.63E-16	6.59E-16
5	7	3.70E-17	3.36E-17	<1.44E-17	1.19E-16	1.42E-16	2.31E-16
7	14	5.43E-17	4.85E-17	<1.81E-17	3.90E-17	2.55E-16	3.69E-16
14	28	3.79E-17	3.36E-17	<9.12E-18	<9.29E-18	2.18E-16	2.62E-16
14	42	2.39E-17	3.19E-17	<1.55E-17	<1.57E-17	2.20E-16	3.39E-16
7	49	<8.40E-17	<8.47E-17	<8.12E-17	<8.28E-17	6.15E-16	1.90E-15
14	63	<2.58E-17	2.99E-17	<2.49E-17	<2.54E-17	2.12E-16	4.83E-16
14	77	<3.22E-17	<3.25E-17	<3.11E-17	<3.17E-17	1.82E-16	2.64E-16
14	91	<3.86E-17	<3.89E-17	<3.73E-17	<3.80E-17	8.98E-17	2.91E-16

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

Table D-14. Duplicate Values, Means, and Standard Deviations of Effective Diffusion Coefficients from EPA Draft Method 1315 Leach Tests Averaged Over the 28-, 42-, 49-, and 63-Day Results^(a)

Test/ Mix # ^(b)	Random Run Order	Na Effective Diffusion Coefficient (cm ² /s)				NO ₃ Effective Diffusion Coefficient (cm ² /s)				NO ₂ Effective Diffusion Coefficient (cm ² /s)			
		Duplicate #1	Duplicate #2	Mean	SD ^(c)	Duplicate #1	Duplicate #2	Mean	SD ^(c)	Duplicate #1	Duplicate #2	Mean	SD ^(c)
1	7	1.20E-09	1.22E-09	1.21E-09	1.41E-11	8.46E-10	8.94E-10	8.70E-10	3.39E-11	9.96E-10	1.04E-09	1.02E-09	3.11E-11
2 @	11	6.00E-09	7.84E-09	6.92E-09	1.30E-09	6.66E-09	6.65E-09	6.65E-09	7.07E-12	7.61E-09	7.38E-09	7.50E-09	1.63E-10
3 \$	3	4.67E-09	5.32E-09	4.99E-09	4.60E-10	4.57E-09	4.63E-09	4.60E-09	4.24E-11	5.48E-09	5.39E-09	5.44E-09	6.36E-11
4	5	1.07E-08	7.67E-09	9.18E-09	2.14E-09	5.59E-09	5.46E-09	5.52E-09	9.19E-11	6.35E-09	6.02E-09	6.19E-09	2.33E-10
5	9	4.77E-09	6.35E-09	5.56E-09	1.12E-09	6.37E-09	6.52E-09	6.45E-09	1.06E-10	6.20E-09	6.24E-09	6.22E-09	2.83E-11
6	12	6.32E-09	6.67E-09	6.49E-09	2.47E-10	9.53E-09	1.00E-08	9.77E-09	3.32E-10	9.66E-09	1.01E-08	9.88E-09	3.11E-10
7 #	14	3.26E-09	3.77E-09	3.52E-09	3.61E-10	4.23E-09	5.22E-09	4.72E-09	7.00E-10	4.22E-09	5.14E-09	4.68E-09	6.51E-10
8	1	2.45E-09	2.36E-09	2.40E-09	6.36E-11	2.75E-09	2.45E-09	2.60E-09	2.12E-10	2.58E-09	2.29E-09	2.44E-09	2.05E-10
9	10	4.65E-09	4.61E-09	4.63E-09	2.83E-11	4.12E-09	4.07E-09	4.09E-09	3.54E-11	5.18E-09	4.94E-09	5.06E-09	1.70E-10
10	13	7.39E-09	7.66E-09	7.52E-09	1.91E-10	6.22E-09	5.95E-09	6.08E-09	1.91E-10	5.67E-09	5.45E-09	5.56E-09	1.56E-10
11	6	6.10E-09	6.23E-09	6.17E-09	9.19E-11	8.32E-09	8.87E-09	8.59E-09	3.89E-10	9.37E-09	1.00E-08	9.69E-09	4.45E-10
12	8	4.10E-09	3.92E-09	4.01E-09	1.27E-10	3.95E-09	3.93E-09	3.94E-09	1.41E-11	3.63E-09	3.59E-09	3.61E-09	2.83E-11
13 @	4	6.56E-09	6.32E-09	6.44E-09	1.70E-10	9.19E-09	8.31E-09	8.75E-09	6.22E-10	7.87E-09	7.07E-09	7.47E-09	5.66E-11
14 #	2	4.39E-09	4.50E-09	4.44E-09	7.78E-11	6.32E-09	6.19E-09	6.26E-09	9.19E-11	5.98E-09	5.87E-09	5.93E-09	7.78E-11
15 %	17	7.76E-09	6.90E-09	7.33E-09	6.08E-10	7.28E-09	6.74E-09	7.01E-09	3.82E-10	7.66E-09	6.68E-09	7.17E-09	6.93E-10
16	22	6.90E-09	7.29E-09	7.10E-09	2.76E-10	7.89E-09	8.26E-09	8.07E-09	2.62E-10	7.26E-09	7.53E-09	7.40E-09	1.91E-10
17	25	6.65E-09	6.69E-09	6.67E-09	2.83E-11	7.41E-09	8.01E-09	7.71E-09	4.24E-10	7.26E-09	7.47E-09	7.37E-09	1.48E-10
18	16	2.82E-09	2.86E-09	2.84E-09	2.83E-11	3.24E-09	3.39E-09	3.32E-09	1.06E-10	2.84E-09	3.03E-09	2.94E-09	1.34E-10
19	15	3.51E-09	3.40E-09	3.46E-09	7.78E-11	4.80E-09	4.77E-09	4.78E-09	2.12E-11	5.34E-09	5.19E-09	5.26E-09	1.06E-10
20	24	9.51E-09	6.55E-09	8.03E-09	2.09E-09	6.95E-09	5.18E-09	6.07E-09	1.25E-09	6.49E-09	4.99E-09	5.74E-09	1.06E-09
21 &	23	1.05E-08	1.04E-08	1.05E-08	7.07E-11	1.56E-08	1.47E-08	1.52E-08	6.36E-10	1.42E-08	1.34E-08	1.38E-08	5.66E-10
22 \$	19	4.80E-09	4.89E-09	4.84E-09	6.36E-11	4.39E-09	4.24E-09	4.32E-09	1.06E-10	4.20E-09	4.08E-09	4.14E-09	8.49E-11
23	20	2.45E-08	2.71E-08	2.58E-08	1.84E-09	3.53E-08	3.91E-08	3.72E-08	2.69E-09	2.47E-08	2.80E-08	2.64E-08	2.33E-09
24	26	9.96E-09	9.68E-09	9.82E-09	1.98E-10	9.59E-09	9.70E-09	9.64E-09	7.78E-11	7.94E-09	8.03E-09	7.99E-09	6.36E-11
25 %	21	7.00E-09	6.73E-09	6.86E-09	1.91E-10	7.22E-09	6.76E-09	6.99E-09	3.25E-10	5.94E-09	5.66E-09	5.80E-09	1.98E-10
26 &	18	7.97E-09	8.18E-09	8.07E-09	1.48E-10	8.02E-09	7.69E-09	7.85E-09	2.33E-10	7.21E-09	6.97E-09	7.09E-09	1.70E-10

- (a) The duplicate diffusion coefficients for each test were rounded to two decimal places in scientific notation before calculating the mean and SD. After the mean and SD were calculated, they were rounded to two decimal places in scientific notation.
- (b) Test numbers with the same symbols are replicates.
- (c) The SD of duplicate results represents the variation in results of making duplicate samples from each test/mix, then testing them and averaging the results over the 28-, 42-, 49-, and 63-day periods. This "Duplicate or Repeat SD" is distinct from "Replicate SD," which takes into account variations in making the mixes and testing with different beginning dates over the period of the screening matrix testing. See Section 8.5 for discussion of replicate variation.

Table D-14. Duplicate Values, Means, and Standard Deviations of Effective Diffusion Coefficients from EPA Draft Method 1315 Leach Tests Averaged Over the 28-, 42-, 49-, and 63-Day Results (contd)^(a)

Test/ Mix # ^(b)	Random Run Order	I Effective Diffusion Coefficient (cm ² /s)				Tc Effective Diffusion Coefficient (cm ² /s)				Cr Effective Diffusion Coefficient (cm ² /s)			
		Duplicate #1	Duplicate #2	Mean	SD ^(c)	Duplicate #1	Duplicate #2	Mean	SD ^(c)	Duplicate #1	Duplicate #2	Mean	SD ^(c)
1	7	6.93E-10	6.92E-10	6.93E-10	7.07E-13	4.75E-11	4.55E-11	4.65E-11	1.41E-12	1.97E-13	1.77E-13	1.87E-13	1.41E-14
2 @	11	7.14E-09	6.89E-09	7.02E-09	1.77E-10	6.69E-11	5.29E-11	5.99E-11	9.90E-12	8.30E-14	8.14E-14	8.22E-14	1.13E-15
3 \$	3	4.94E-09	5.03E-09	4.98E-09	6.36E-11	3.46E-11	3.47E-11	3.47E-11	7.07E-14	1.66E-13	1.62E-13	1.64E-13	2.83E-15
4	5	3.90E-09	3.95E-09	3.93E-09	3.54E-11	2.54E-11	3.11E-11	2.82E-11	4.03E-12	5.99E-14	6.58E-14	6.29E-14	4.17E-15
5	9	5.22E-09	5.36E-09	5.29E-09	9.90E-11	7.11E-11	7.18E-11	7.14E-11	4.95E-13	7.73E-14	8.33E-14	8.03E-14	4.24E-15
6	12	9.94E-09	1.03E-08	1.01E-08	2.55E-10	1.54E-10	1.50E-10	1.52E-10	2.83E-12	1.66E-13	1.40E-13	1.53E-13	1.84E-14
7 #	14	3.93E-09	4.56E-09	4.25E-09	4.45E-10	8.97E-12	9.16E-12	9.06E-12	1.34E-13	7.95E-15	8.62E-15	8.28E-15	4.74E-16
8	1	2.63E-09	2.47E-09	2.55E-09	1.13E-10	2.22E-10	2.09E-10	2.16E-10	9.19E-12	6.59E-13	7.70E-13	7.15E-13	7.85E-14
9	10	5.20E-09	5.18E-09	5.19E-09	1.41E-11	3.56E-11	3.38E-11	3.47E-11	1.27E-12	1.42E-13	1.77E-13	1.60E-13	2.47E-14
10	13	4.45E-09	4.41E-09	4.43E-09	2.83E-11	1.21E-10	1.24E-10	1.22E-10	2.12E-12	3.66E-13	1.03E-12	6.98E-13	4.70E-13
11	6	1.17E-08	1.26E-08	1.22E-08	6.36E-10	1.30E-10	1.30E-10	1.30E-10	0.00E+00	1.36E-13	1.33E-13	1.34E-13	2.12E-15
12	8	2.70E-09	2.74E-09	2.72E-09	2.83E-11	4.15E-11	4.25E-11	4.20E-11	7.07E-13	1.64E-13	2.41E-13	2.03E-13	5.44E-14
13 @	4	8.72E-09	8.24E-09	8.48E-09	3.39E-10	8.96E-11	7.59E-11	8.27E-11	9.69E-12	5.64E-14	5.37E-14	5.51E-14	1.91E-15
14 #	2	5.66E-09	5.74E-09	5.70E-09	5.66E-11	8.11E-12	1.11E-11	9.61E-12	2.11E-12	1.00E-14	9.79E-15	9.90E-15	1.48E-16
15 %	17	6.05E-09	5.29E-09	5.67E-09	5.37E-10	4.51E-11	4.72E-11	4.62E-11	1.48E-12	3.59E-13	2.72E-13	3.16E-13	6.15E-14
16	22	7.63E-09	8.15E-09	7.89E-09	3.68E-10	1.54E-10	1.61E-10	1.58E-10	4.95E-12	2.34E-13	2.57E-13	2.46E-13	1.63E-14
17	25	7.46E-09	7.71E-09	7.58E-09	1.77E-10	1.81E-10	1.71E-10	1.76E-10	7.07E-12	1.76E-13	1.55E-13	1.66E-13	1.48E-14
18	16	3.07E-09	3.21E-09	3.14E-09	9.90E-11	5.57E-12	5.77E-12	5.67E-12	1.41E-13	1.81E-14	1.86E-14	1.83E-14	3.54E-16
19	15	5.45E-09	5.49E-09	5.47E-09	2.83E-11	4.70E-11	4.43E-11	4.56E-11	1.91E-12	5.58E-14	6.07E-14	5.83E-14	3.46E-15
20	24	4.50E-09	3.12E-09	3.81E-09	9.76E-10	4.12E-11	3.43E-11	3.77E-11	4.88E-12	8.00E-14	7.14E-14	7.57E-14	6.08E-15
21 &	23	1.68E-08	1.68E-08	1.68E-08	0.00E+00	7.48E-11	1.14E-10	9.44E-11	2.77E-11	1.45E-13	1.57E-13	1.51E-13	8.49E-15
22 \$	19	4.43E-09	4.41E-09	4.42E-09	1.41E-11	2.18E-11	2.79E-11	2.48E-11	4.31E-12	1.93E-13	1.65E-13	1.79E-13	1.98E-14
23	20	3.18E-08	3.79E-08	3.48E-08	4.31E-09	1.23E-09	7.94E-10	1.01E-09	3.08E-10	1.24E-12	1.08E-12	1.16E-12	1.13E-13
24	26	8.24E-09	8.07E-09	8.15E-09	1.20E-10	6.68E-11	6.41E-11	6.55E-11	1.91E-12	1.16E-13	7.26E-14	9.43E-14	3.07E-14
25 %	21	4.96E-09	4.78E-09	4.87E-09	1.27E-10	3.37E-11	2.40E-11	2.88E-11	6.86E-12	3.32E-13	4.71E-13	4.02E-13	9.83E-14
26 &	18	7.27E-09	7.32E-09	7.30E-09	3.54E-11	1.76E-10	1.56E-10	1.66E-10	1.41E-11	1.04E-13	9.92E-14	1.02E-13	3.39E-15

- (a) The duplicate diffusion coefficients for each test were rounded to two decimal places in scientific notation before calculating the mean and SD. After the mean and SD were calculated, they were rounded to two decimal places in scientific notation.
- (b) Test numbers with the same symbols are replicates.
- (c) The SD of duplicate results represents the variation in results of making duplicate samples from each test/mix, then testing them and averaging results over the 28-, 42-, 49-, and 63-day periods. This "Duplicate or Repeat SD" is distinct from "Replicate SD," which takes into account variations in making the mixes and testing with different beginning dates over the period of the screening matrix testing. See Section 8.5 for discussion of replicate variation.

Table D-14. Duplicate Values, Means, and Standard Deviations of Effective Diffusion Coefficients from EPA Draft Method 1315 Leach Tests Averaged Over the 28-, 42-, 49-, and 63-Day Results (contd)^(a)

Test/Mix # ^(b)	Random Run Order	U Effective Diffusion Coefficient (cm ² /s)			
		Duplicate #1	Duplicate #2	Mean	SD ^(c)
1	7	2.90E-16	3.28E-16	3.09E-16	2.69E-17
2 @	11	1.05E-16	9.93E-17	1.02E-16	4.03E-18
3 \$	3	3.33E-17	3.43E-17	3.38E-17	7.07E-19
4	5	1.11E-16	1.12E-16	1.11E-16	7.07E-19
5	9	4.16E-17	4.17E-17	4.16E-17	7.07E-20
6	12	7.20E-17	4.64E-17	5.92E-17	1.81E-17
7 #	14	4.30E-17	4.46E-17	4.38E-17	1.13E-18
8	1	5.10E-16	4.53E-16	4.82E-16	4.03E-17
9	10	3.21E-17	3.16E-17	3.19E-17	3.54E-19
10	13	8.43E-17	9.00E-17	8.71E-17	4.03E-18
11	6	3.48E-17	3.42E-17	3.45E-17	4.24E-19
12	8	7.75E-17	3.05E-16	1.91E-16	1.61E-16
13 @	4	1.03E-16	1.02E-16	1.03E-16	7.07E-19
14 #	2	4.38E-17	4.36E-17	4.37E-17	1.41E-19
15 %	17	5.52E-17	4.29E-17	4.91E-17	8.70E-18
16	22	9.52E-17	1.53E-16	1.24E-16	4.09E-17
17	25	8.88E-17	8.69E-17	8.79E-17	1.34E-18
18	16	1.53E-16	3.43E-16	2.48E-16	1.34E-16
19	15	4.55E-17	4.51E-17	4.53E-17	2.83E-19
20	24	9.65E-17	9.43E-17	9.54E-17	1.56E-18
21 &	23	4.29E-17	4.50E-17	4.39E-17	1.48E-18
22 \$	19	3.27E-17	3.33E-17	3.30E-17	4.24E-19
23	20	3.16E-16	7.46E-16	5.31E-16	3.04E-16
24	26	8.09E-17	8.36E-17	8.23E-17	1.91E-18
25 %	21	4.31E-17	4.46E-17	4.39E-17	1.06E-18
26 &	18	3.35E-17	3.33E-17	3.34E-17	1.41E-19

(a) The duplicate diffusion coefficients for each test were rounded to two decimal places in scientific notation before calculating the mean and SD. After the mean and SD were calculated, they were rounded to two decimal places in scientific notation.

(b) Test numbers with the same symbols are replicates.

(c) The SD of duplicate results represents the variation in results of making duplicate samples from each test/mix, then testing them and averaging results over the 28-, 42-, 49-, and 63-day periods. This “Duplicate or Repeat SD” is distinct from “Replicate SD,” which takes into account variations in making the mixes and testing with different beginning dates over the period of the screening matrix testing. See Section 8.5 for discussion of replicate variation.

Table D-15. Duplicate Values, Means, and Standard Deviations of Leachability Index Values from EPA Draft Method 1315 Leach Tests Averaged Over the 28-, 42-, 49-, and 63-Day Results^(a)

Test/ Mix # ^(b)	Random Run Order	Na Leachability Index ($-\log_{10}(\text{cm}^2/\text{s})$)				NO ₃ Leachability Index ($-\log_{10}(\text{cm}^2/\text{s})$)				NO ₂ Leachability Index ($-\log_{10}(\text{cm}^2/\text{s})$)			
		Duplicate #1	Duplicate #2	Mean	SD ^(c)	Duplicate #1	Duplicate #2	Mean	SD ^(c)	Duplicate #1	Duplicate #2	Mean	SD ^(c)
1	7	8.92	8.92	8.92	0.000	9.09	9.05	9.07	0.028	9.02	8.99	9.00	0.021
2 @	11	8.24	8.16	8.20	0.057	8.20	8.20	8.20	0.000	8.17	8.17	8.17	0.000
3 \$	3	8.36	8.28	8.32	0.057	8.34	8.34	8.34	0.000	8.27	8.28	8.27	0.007
4	5	8.05	8.12	8.09	0.049	8.26	8.27	8.27	0.007	8.21	8.23	8.22	0.014
5	9	8.33	8.22	8.28	0.078	8.24	8.22	8.23	0.014	8.25	8.24	8.25	0.007
6	12	8.23	8.21	8.22	0.014	8.06	8.04	8.05	0.014	8.05	8.03	8.04	0.014
7 #	14	8.51	8.45	8.48	0.042	8.43	8.34	8.38	0.064	8.42	8.34	8.38	0.057
8	1	8.61	8.63	8.62	0.014	8.56	8.62	8.59	0.042	8.59	8.65	8.62	0.042
9	10	8.34	8.34	8.34	0.000	8.40	8.40	8.40	0.000	8.29	8.31	8.30	0.014
10	13	8.14	8.13	8.14	0.007	8.23	8.25	8.24	0.014	8.26	8.29	8.27	0.021
11	6	8.23	8.21	8.22	0.014	8.08	8.05	8.07	0.021	8.03	8.00	8.02	0.021
12	8	8.39	8.41	8.40	0.014	8.40	8.41	8.41	0.007	8.44	8.45	8.45	0.007
13 @	4	8.20	8.21	8.21	0.007	8.06	8.11	8.09	0.035	8.12	8.17	8.14	0.035
14 #	2	8.37	8.36	8.36	0.007	8.22	8.24	8.23	0.014	8.25	8.26	8.25	0.007
15 %	17	8.12	8.17	8.14	0.035	8.17	8.19	8.18	0.014	8.14	8.20	8.17	0.042
16	22	8.17	8.14	8.16	0.021	8.11	8.10	8.11	0.007	8.15	8.13	8.14	0.014
17	25	8.19	8.19	8.19	0.000	8.15	8.11	8.13	0.028	8.16	8.14	8.15	0.014
18	16	8.55	8.55	8.55	0.000	8.50	8.47	8.48	0.021	8.55	8.52	8.54	0.021
19	15	8.50	8.49	8.50	0.007	8.37	8.36	8.36	0.007	8.31	8.31	8.31	0.000
20	24	8.03	8.18	8.11	0.106	8.17	8.29	8.23	0.085	8.19	8.30	8.25	0.078
21 &	23	8.00	8.00	8.00	0.000	7.83	7.86	7.85	0.021	7.87	7.89	7.88	0.014
22 \$	19	8.32	8.31	8.32	0.007	8.36	8.38	8.37	0.014	8.38	8.39	8.39	0.007
23	20	7.62	7.57	7.60	0.035	7.47	7.42	7.45	0.035	7.62	7.56	7.59	0.042
24	26	8.01	8.03	8.02	0.014	8.04	8.03	8.04	0.007	8.11	8.10	8.11	0.007
25 %	21	8.17	8.18	8.18	0.007	8.17	8.19	8.18	0.014	8.24	8.26	8.25	0.014
26 &	18	8.12	8.11	8.11	0.007	8.12	8.14	8.13	0.014	8.16	8.17	8.16	0.007

- (a) The duplicate LI values were calculated by converting the effective diffusion coefficients for 28-, 42-, 49-, and 63-day periods to LI values. Then the LI values were averaged over those four periods and the averages were rounded to two decimal places. Finally, the mean and SD were calculated from the rounded values, and the means and SDs themselves were rounded to two and three decimal places, respectively.
- (b) Test numbers with the same symbols are replicates.
- (c) The SD of duplicate results represents the variation in results of making duplicate samples from each test/mix, then testing them and averaging results over the 28-, 42-, 49-, and 63-day periods. This “Duplicate or Repeat SD” is distinct from “Replicate SD,” which takes into account variations in making the mixes and testing with different beginning dates over the period of the screening matrix testing. See Section 8.5 for discussion of replicate variation.

Table D-15. Duplicate Values, Means, and Standard Deviations of Leachability Index Values from EPA Draft Method 1315 Leach Tests Averaged Over the 28-, 42-, 49-, and 63-Day Results (contd)^(a)

Test/ Mix # ^(b)	Random Run Order	I Leachability Index ($-\log_{10}(\text{cm}^2/\text{s})$)				Tc Leachability Index ($-\log_{10}(\text{cm}^2/\text{s})$)				Cr Leachability Index ($-\log_{10}(\text{cm}^2/\text{s})$)			
		Duplicate #1	Duplicate #2	Mean	SD ^(c)	Duplicate #1	Duplicate #2	Mean	SD ^(c)	Duplicate #1	Duplicate #2	Mean	SD ^(c)
1	7	9.16	9.16	9.16	0.000	10.33	10.34	10.34	0.007	12.72	12.76	12.74	0.028
2 @	11	8.17	8.18	8.18	0.007	10.18	10.28	10.23	0.071	13.09	13.10	13.09	0.007
3 \$	3	8.31	8.30	8.30	0.007	10.46	10.46	10.46	0.000	12.79	12.80	12.80	0.007
4	5	8.42	8.41	8.41	0.007	10.60	10.51	10.56	0.064	13.23	13.19	13.21	0.028
5	9	8.33	8.32	8.32	0.007	10.16	10.15	10.16	0.007	13.14	13.10	13.12	0.028
6	12	8.05	8.03	8.04	0.014	9.90	9.91	9.91	0.007	12.82	12.89	12.86	0.049
7 #	14	8.46	8.39	8.43	0.049	11.05	11.04	11.05	0.007	14.12	14.09	14.11	0.021
8	1	8.58	8.61	8.59	0.021	9.65	9.68	9.66	0.021	12.18	12.12	12.15	0.042
9	10	8.29	8.29	8.29	0.000	10.45	10.47	10.46	0.014	12.86	12.76	12.81	0.071
10	13	8.37	8.38	8.38	0.007	9.92	9.91	9.91	0.007	12.45	12.00	12.23	0.318
11	6	7.93	7.90	7.92	0.021	9.95	9.95	9.95	0.000	12.88	12.89	12.89	0.007
12	8	8.57	8.56	8.57	0.007	10.38	10.37	10.38	0.007	12.81	12.72	12.77	0.064
13 @	4	8.08	8.10	8.09	0.014	10.05	10.12	10.09	0.049	13.25	13.28	13.27	0.021
14 #	2	8.28	8.27	8.27	0.007	11.09	10.96	11.03	0.092	14.01	14.02	14.02	0.007
15 %	17	8.24	8.30	8.27	0.042	10.35	10.34	10.34	0.007	12.59	12.71	12.65	0.085
16	22	8.13	8.10	8.12	0.021	9.82	9.80	9.81	0.014	12.64	12.60	12.62	0.028
17	25	8.15	8.13	8.14	0.014	9.75	9.77	9.76	0.014	12.76	12.81	12.79	0.035
18	16	8.52	8.50	8.51	0.014	11.26	11.24	11.25	0.014	13.82	13.80	13.81	0.014
19	15	8.34	8.31	8.32	0.021	10.43	10.44	10.43	0.007	13.26	13.23	13.25	0.021
20	24	8.36	8.51	8.43	0.106	10.39	10.47	10.43	0.057	13.11	13.15	13.13	0.028
21 &	23	7.81	7.81	7.81	0.000	10.20	10.12	10.16	0.057	12.88	12.84	12.86	0.028
22 \$	19	8.36	8.36	8.36	0.000	10.68	10.57	10.63	0.078	12.73	12.80	12.77	0.049
23	20	7.52	7.45	7.48	0.049	8.94	9.13	9.04	0.134	11.92	11.99	11.96	0.049
24	26	8.11	8.12	8.11	0.007	10.18	10.20	10.19	0.014	12.95	13.15	13.05	0.141
25 %	21	8.34	8.35	8.34	0.007	10.47	10.62	10.55	0.106	12.60	12.52	12.56	0.057
26 &	18	8.17	8.17	8.17	0.000	9.85	9.90	9.88	0.035	13.02	13.04	13.03	0.014

(a) The duplicate LI values were calculated by converting the effective diffusion coefficients for 28-, 42-, 49-, and 63-day periods to LI values. Then the LI values were averaged over those four periods and the averages were rounded to two decimal places. Finally, the mean and SD were calculated from the rounded values, and the means and SDs themselves were rounded to two and three decimal places, respectively.

(b) Test numbers with the same symbols are replicates.

(c) The SD of duplicate results represents the variation in results of making duplicate samples from each test/mix, then testing them and averaging results over the 28-, 42-, 49-, and 63-day periods. This “Duplicate or Repeat SD” is distinct from “Replicate SD,” which takes into account variations in making the mixes and testing with different beginning dates over the period of the screening matrix testing. See Section 8.5 for discussion of replicate variation.

Table D-15. Duplicate Values, Means, and Standard Deviations of Leachability Index Values from EPA Draft Method 1315 Leach Tests Averaged Over the 28-, 42-, 49-, and 63-Day Results (contd)^(a)

Test/Mix # ^(b)	Random Run Order	U Leachability Index ($-\log_{10}(\text{cm}^2/\text{s})$)			
		Duplicate #1	Duplicate #2	Mean	SD ^(c)
1	7	15.62	15.58	15.60	0.028
2 @	11	16.04	16.08	16.06	0.028
3 \$	3	16.63	16.62	16.63	0.007
4	5	16.11	16.10	16.11	0.007
5	9	16.53	16.53	16.53	0.000
6	12	16.40	16.47	16.43	0.049
7 #	14	16.50	16.50	16.50	0.000
8	1	15.40	15.44	15.42	0.028
9	10	16.64	16.65	16.65	0.007
10	13	16.17	16.13	16.15	0.028
11	6	16.61	16.62	16.62	0.007
12	8	16.26	16.04	16.15	0.156
13 @	4	16.07	16.07	16.07	0.000
14 #	2	16.49	16.51	16.50	0.014
15 %	17	16.42	16.52	16.47	0.071
16	22	16.09	15.87	15.98	0.156
17	25	16.15	16.16	16.16	0.007
18	16	15.85	15.56	15.71	0.205
19	15	16.49	16.50	16.49	0.007
20	24	16.17	16.18	16.18	0.007
21 &	23	16.43	16.39	16.41	0.028
22 \$	19	16.64	16.63	16.63	0.007
23	20	15.55	15.27	15.41	0.198
24	26	16.23	16.22	16.23	0.007
25 %	21	16.52	16.48	16.50	0.028
26 &	18	16.62	16.63	16.63	0.007

(a) The duplicate LI values were calculated by converting the effective diffusion coefficients for 28-, 42-, 49-, and 63-day periods to LI values. Then the LI values were averaged over those four periods and the averages rounded to two decimal places. Finally, the mean and SD were calculated from the rounded values, and the means and SDs themselves were rounded to two and three decimal places, respectively.

(b) Test numbers with the same symbols are replicates.

(c) The SD of duplicate results represents the variation in results of making duplicate samples from each test/mix, then testing them and averaging results over the 28-, 42-, 49-, and 63-day periods. This “Duplicate or Repeat SD” is distinct from “Replicate SD,” which takes into account variations in making the mixes and testing with different beginning dates over the period of the screening matrix testing. See Section 8.5 for discussion of replicate variation.

Appendix E. Data-Interaction Plots for Selected Cast Stone Properties

This appendix contains graphical displays referred to as *data-interaction plots* for Cast Stone properties that are statistically analyzed in Section 8.0. The properties tested by SRNL (Plastic Viscosity, Heat Generation at 300 Hours, and Time to Peak Heat Generation,) have data for 38 mixes, because 12 of the original 26 mixes (Mixes 1–26) were mis-batched and tested, (denoted Mixes 27–38) and then subsequently those 12 mixes were correctly batched and tested. The remaining properties that are statistically analyzed in Section 8.0 (compressive strength; EPA 1315 leachability indices for Na, NO₃, I, Tc, and Cr) were batched and tested by PNNL, so there are only data for the original 26 mixes (Mixes 1–26). Hence, the data-interaction plots are based on different numbers of test results depending on the property.

The data-interaction plots have a specific Cast Stone property on the y-axis, one of the test parameters on the x-axis (Parameter A), and another of the test parameters (Parameter B) whose values are represented by different plotting symbols and line segments. The property values for the mixes are plotted using different plotting symbols and colors. Also plotted is a line segment for each value of Parameter B, where the values at the endpoints of the line segment are the means of all property values having the possible values of Parameter A.

For each property two groups of plots are presented. The first group of plots is for pairs of test parameters for which interactions were accounted for in designing the screening test matrix. This group of plots includes the following pairs of components:

- Na Molarity and Simulant
- Fly Ash and Simulant
- Mix Ratio and Simulant
- Na Molarity and Fly Ash
- Na Molarity and Mix Ratio
- Mix Ratio and Fly Ash.

In this group of plots, the line segments are shown as solid lines.

The second group of plots is for pairs of test parameters for which interactions were assumed to be non-significant in designing the screening test matrix. This group of plots includes the following pairs of components:

- blast furnace slag and fly ash
- Na molarity and blast furnace slag
- fly ash and blast furnace slag
- mix ratio and blast furnace slag.

In this group of plots, the line segments are shown as dashed lines as a reminder that two-parameter interactions with BFS were originally assumed to be non-significant when developing the test matrix for Mixes 1–26. However, because the 12 mixes mis-batched by SRNL involved switching the BFS sources (NW and SE), and those 12 mixes were subsequently correctly batched and tested, the data for the SRNL properties noted above contain additional information regarding the individual and two-parameter interaction effects of BFS.

The primary purpose of a data-interaction plot is to graphically assess whether Parameters A and B have a significant interactive effect on a property. A significant interaction occurs when the effect of one parameter on the property depends significantly on the value of the second

parameter. Graphically, a significant interaction exists when the line segments in a plot are significantly non-parallel. When subjectively judging whether line segments are significantly non-parallel the following four things are considered:

- The spread of the endpoints of the line segments must be considered relative to the spread of the data points. If the spread of the endpoints is small relative to the spread of the data points, that means that test parameters other than Parameters A and B in a plot account for substantial variation in the data. In such cases, the potential interactive effect of Parameters A and B may be small compared to the effects of other parameters, and hence less likely to be significant. On the other hand, if the spread of the endpoints of line segments accounts for a substantial fraction of the spread of data points, that means Parameters A and/or B have substantive effects, and hence are more likely to interact.
- Outlying data points may substantively affect the mean property values at the endpoints of the line segments, and hence may be the cause of what appears to be a significant interactive effect of Parameters A and B on the property.
- The screening test matrix was designed based on the assumptions that certain individual parameter effects and two-parameter interactions were not significant (see Appendix A). This results in aliasing of parameter effects and interactions (i.e., some interactions are strongly correlated with other interactions and/or individual parameter effects).
- Mixes 1–26 in the planned test matrix are not “balanced” in that each value of each test parameter does not appear an equal number of times in the test matrix. It was not possible to generate a balanced design given the restrictions on the number of tests that could be performed, and the assumptions about non-significant effects of some individual and two-parameter interaction effects. Mixes 1–38 for the SRNL properties are also not balanced. The consequence of unbalanced data is that when averaging over subsets of data to form the endpoints of the lines in the plots, there can be unequal effects of other parameters than the two parameters whose interaction is being assessed with a given plot. Thus, the parallelism or non-parallelism of lines in a given data-interaction plot can be affected by the lack of balance. The statistical analyses of the data in Section 8.0 account for the structure of the data, and hence may result in conclusions that may appear to differ, in some cases, from what is shown in the data-interaction plots in this appendix.

The following sections present the data-interaction plots for the properties of mixes in the screening test matrix that are statistically analyzed in Section 8.0. Note that the figures in this appendix are provided “For Information Only” (for the reason in the last bullet above), and hence the PNNL quality assurance procedures were not applied for these figures. Similar figures that are based on the models fit as part of statistical data analyses are presented in Section 8.0, and the required QA procedures were completed for those figures.

E.1 Plastic Viscosity

Figure E-1 to Figure E-10 display the data-interaction plots for the plastic viscosity of Cast Stone Mixes 1 through 38. The data in these plots are from Table C-5 in Appendix C.

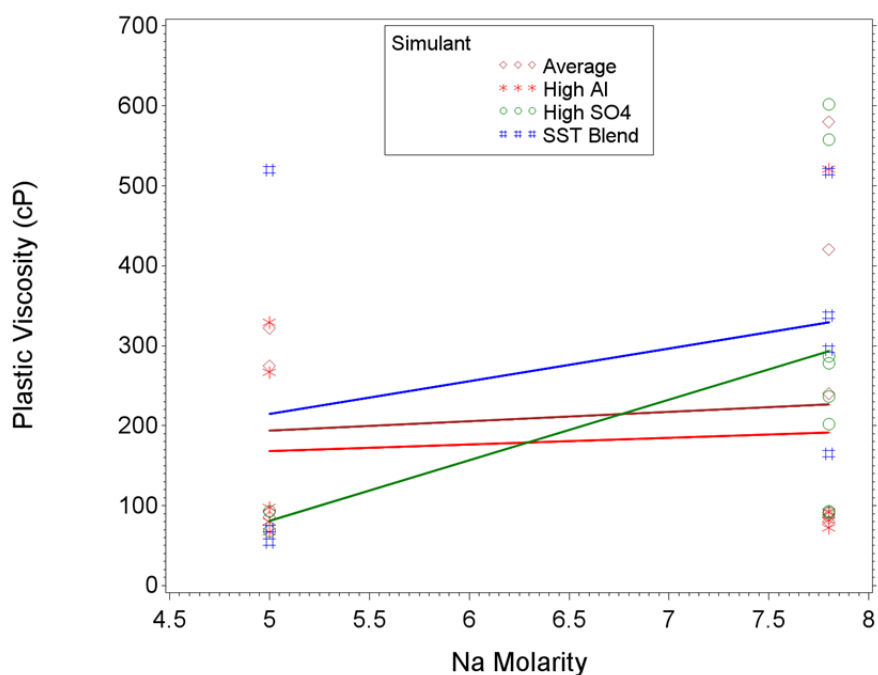


Figure E-1. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Plastic Viscosity

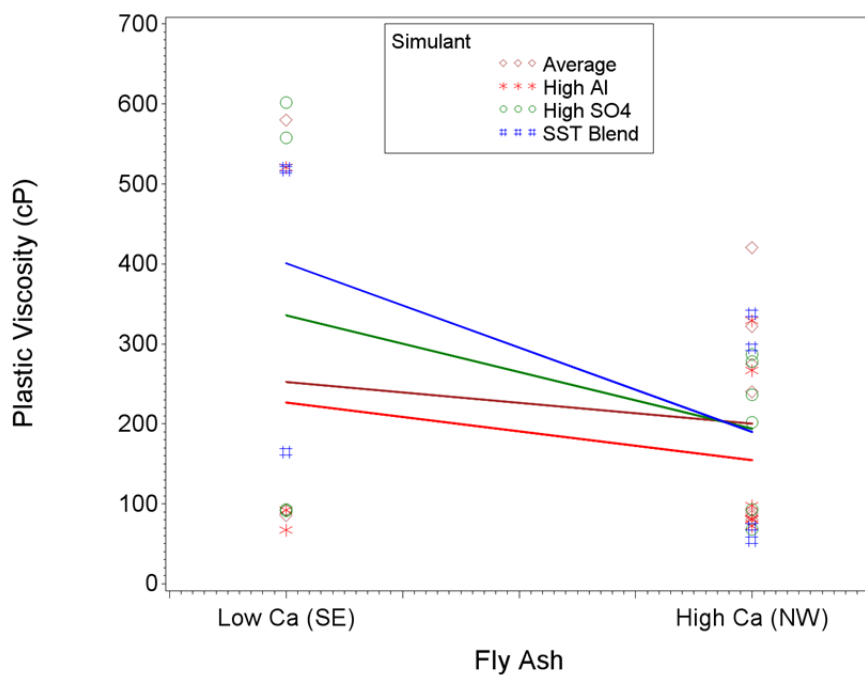


Figure E-2. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Plastic Viscosity

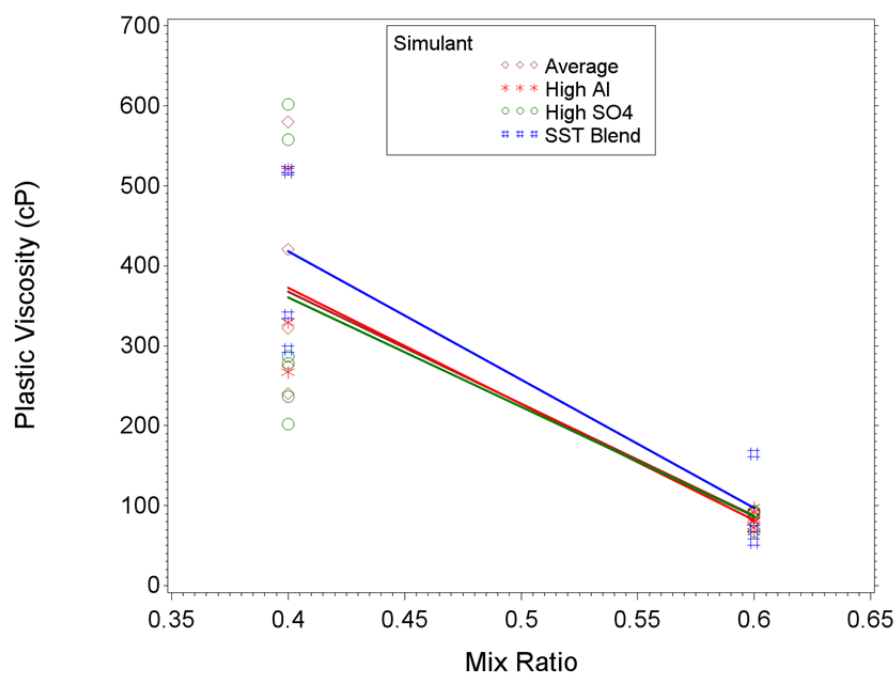


Figure E-3. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Plastic Viscosity

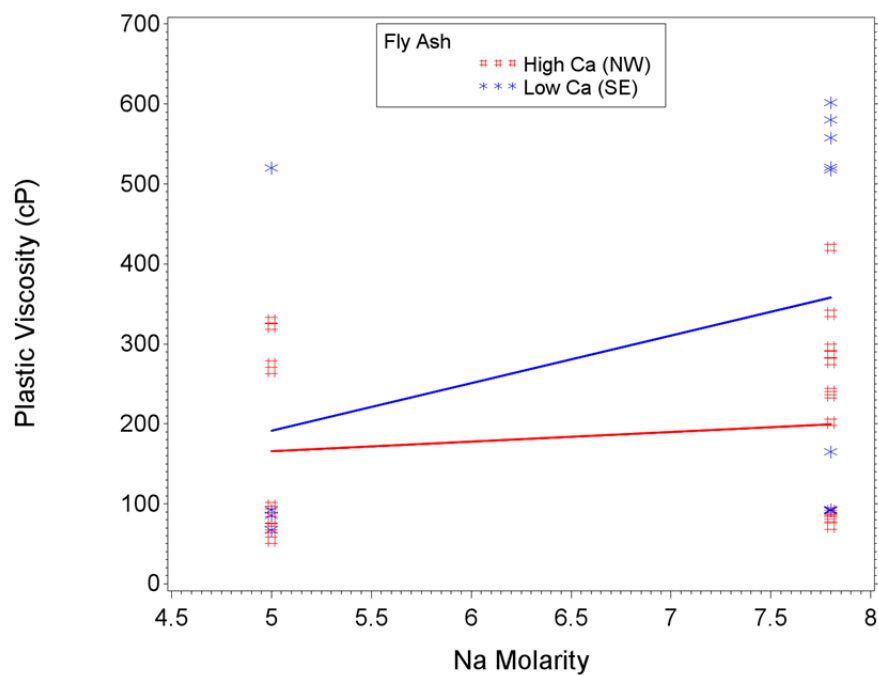


Figure E-4. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Plastic Viscosity

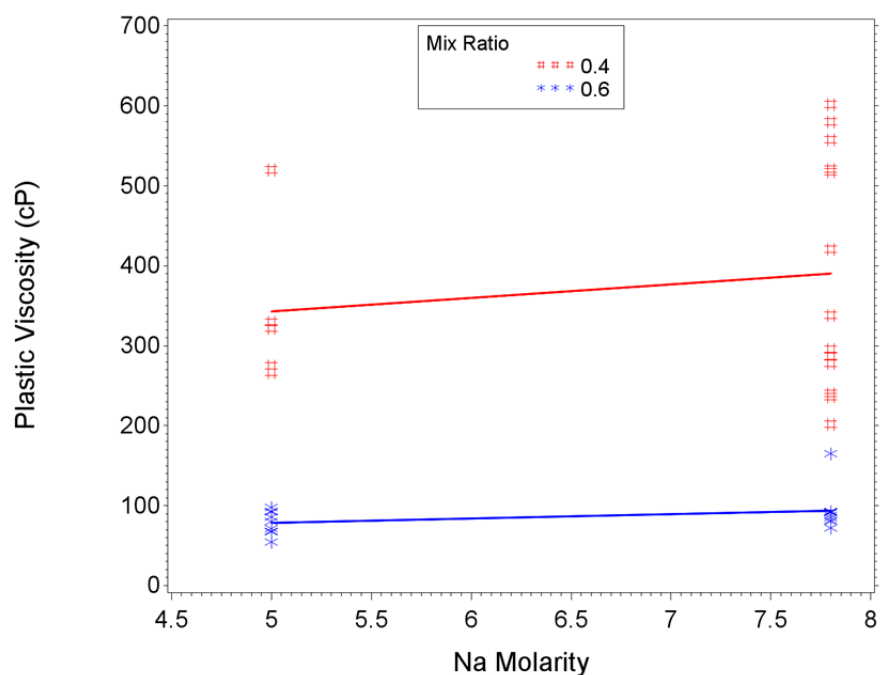


Figure E-5. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Plastic Viscosity

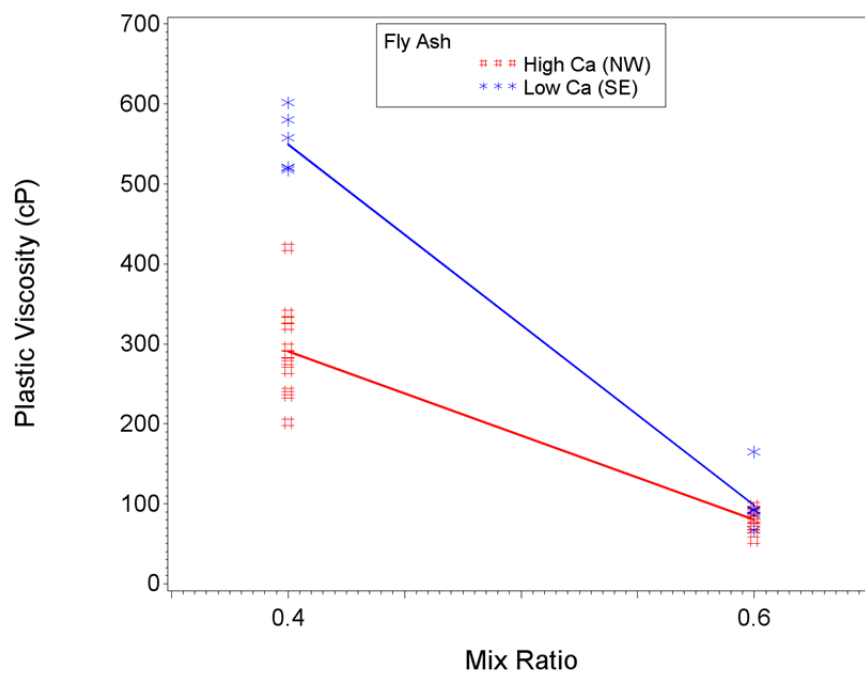


Figure E-6. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Plastic Viscosity

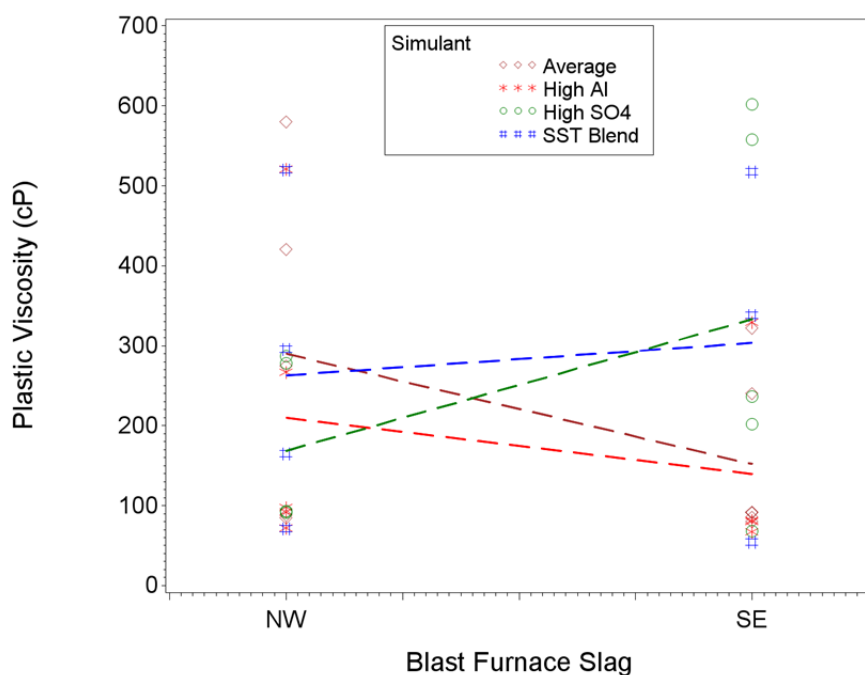


Figure E-7. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Plastic Viscosity

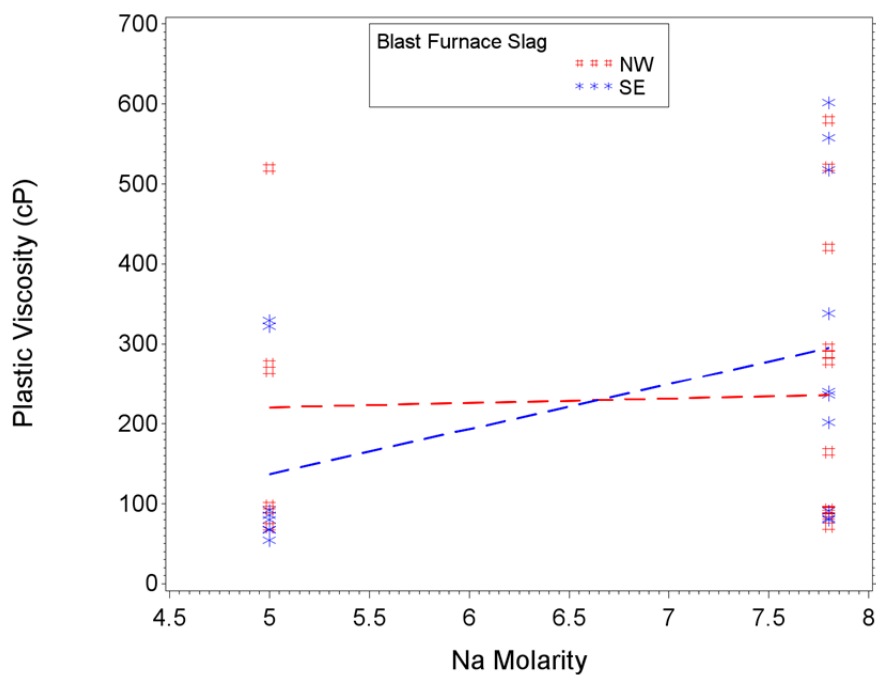


Figure E-8. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Plastic Viscosity

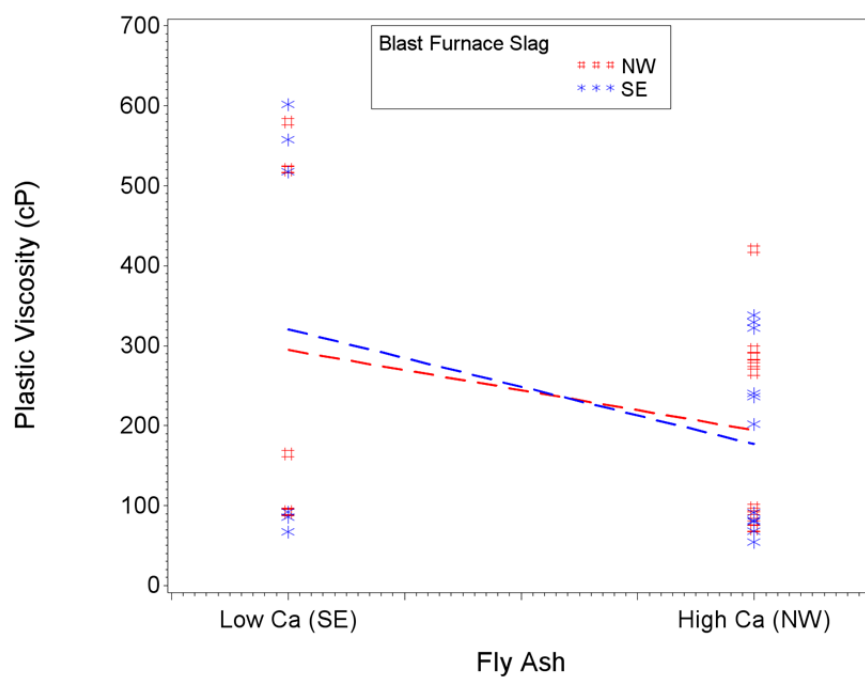


Figure E-9. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Plastic Viscosity

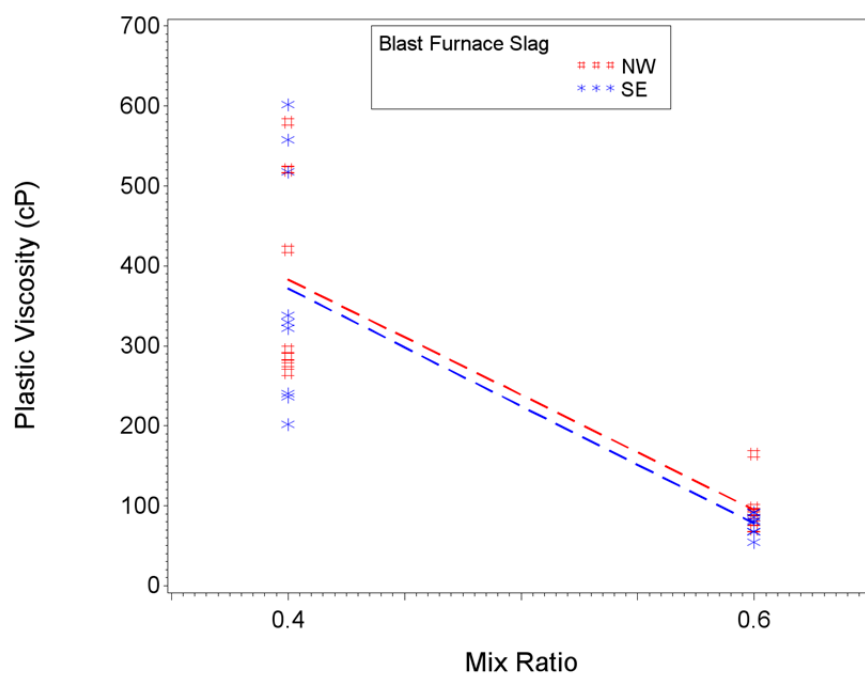


Figure E-10. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Plastic Viscosity

E.2 Heat Generation

This section presents data-interaction plots for two heat-generation properties (Heat Generation at 300 Hours and Time to Peak Heat Generation) of Cast Stone Mixes 1–38. The numerical values (data) in these plots are from Table C-8 in Appendix C.

E.2.1 *Heat Generation at 300 Hours*

Figure E-11 through Figure E-20 display the data-interaction plots for Heat Generation at 300 Hours using the data from Cast Stone Mixes 1–38.

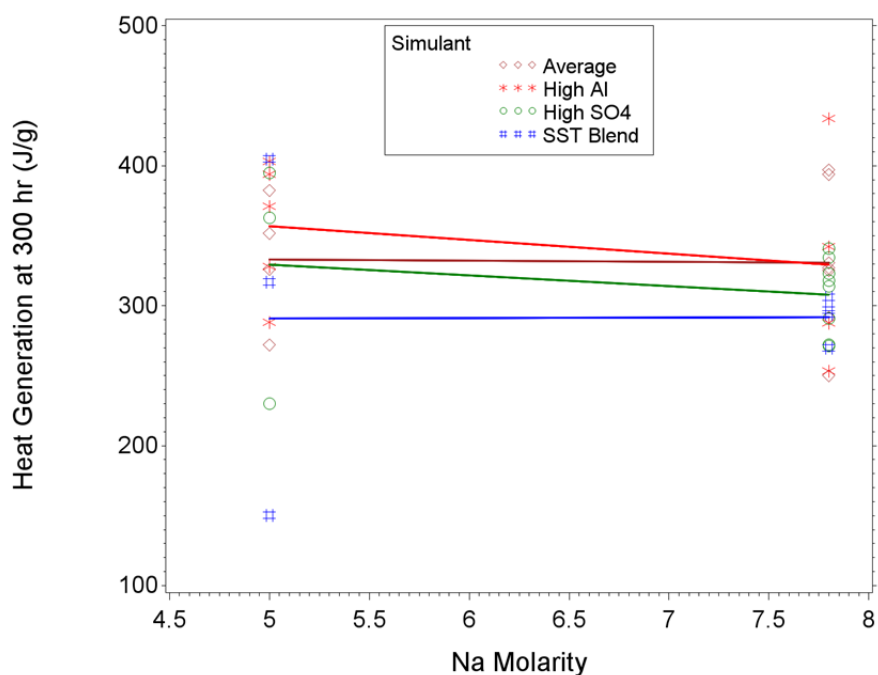


Figure E-11. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Heat Generation at 300 Hours

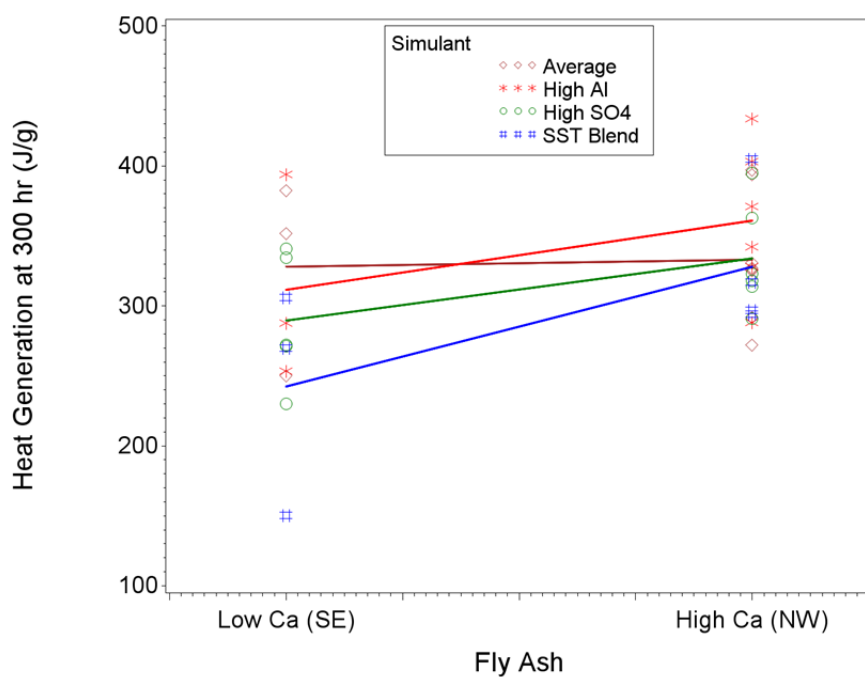


Figure E-12. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Heat Generation at 300 Hours

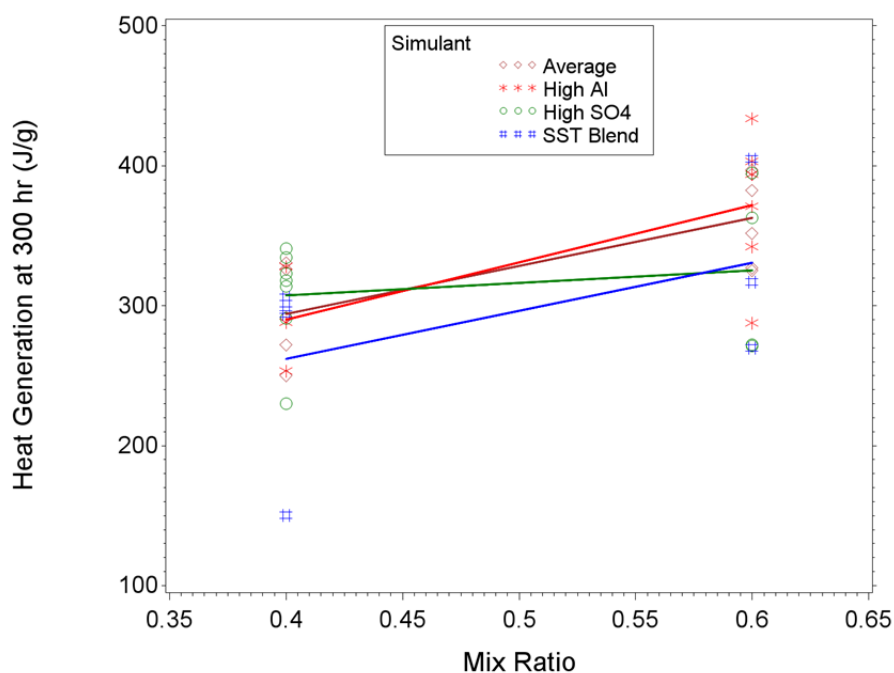


Figure E-13. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Heat Generation at 300 Hours

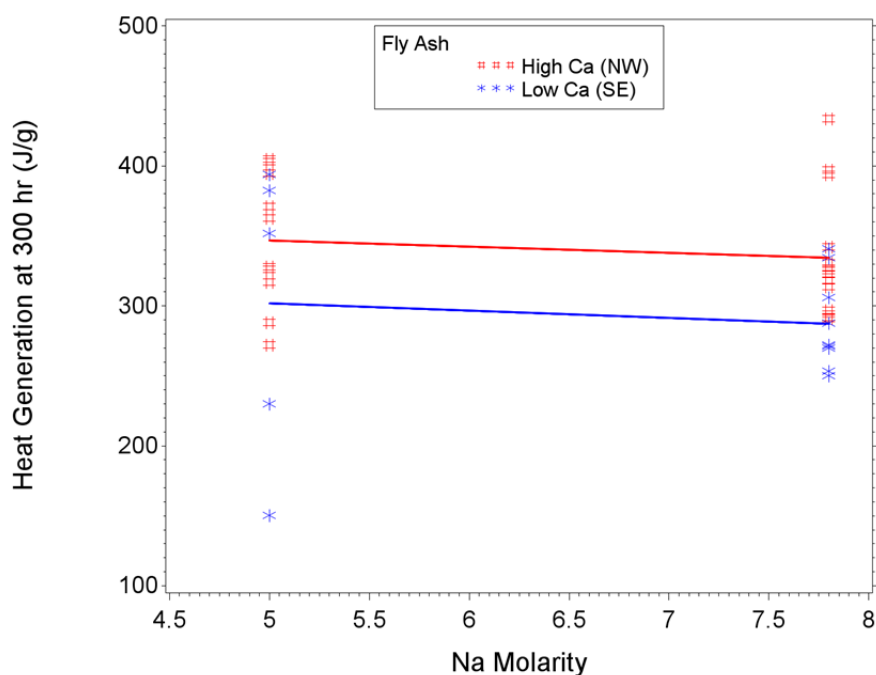


Figure E-14. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Heat Generation at 300 Hours

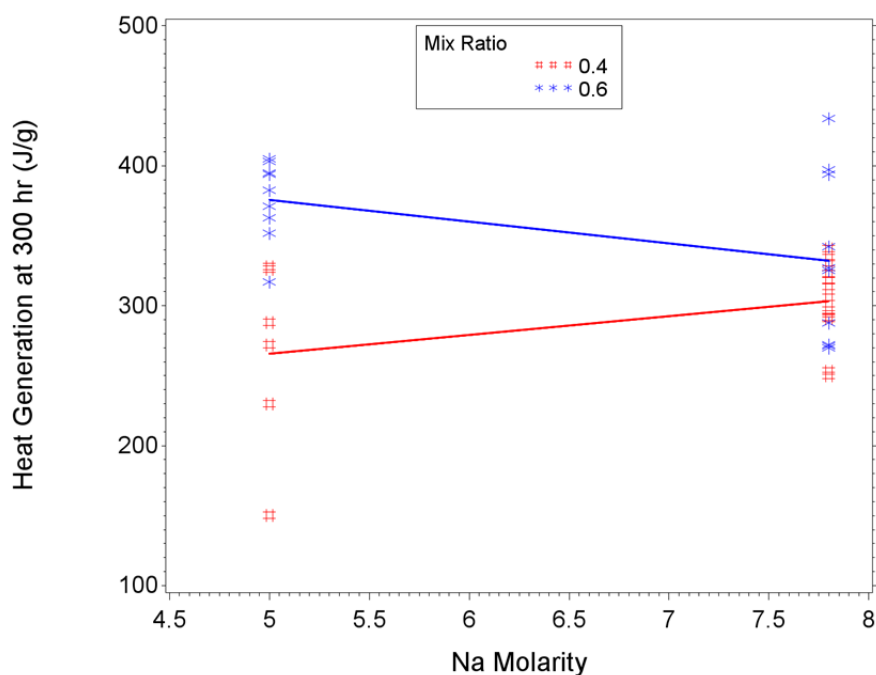


Figure E-15. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Heat Generation at 300 Hours

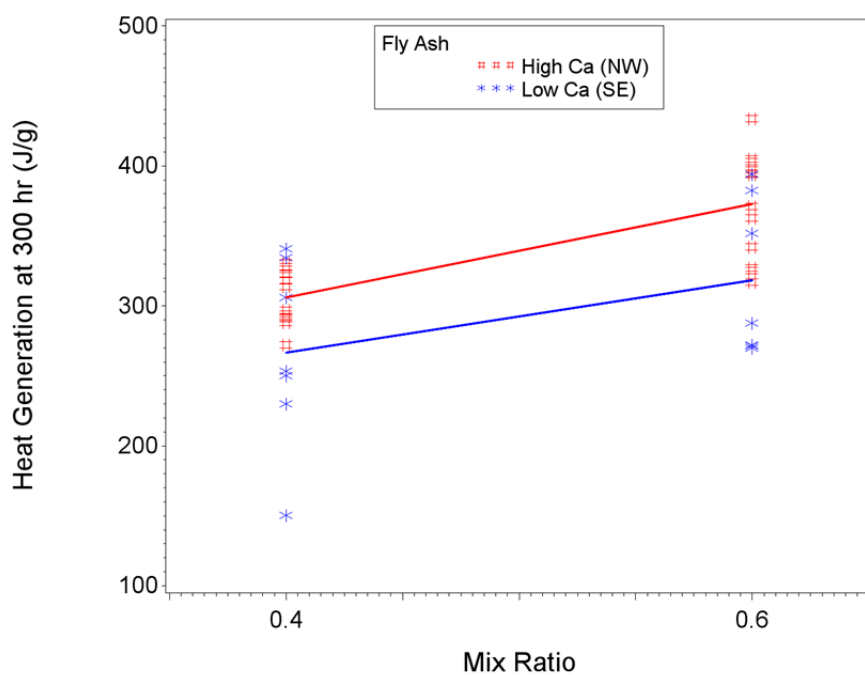


Figure E-16. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Heat Generation at 300 Hours

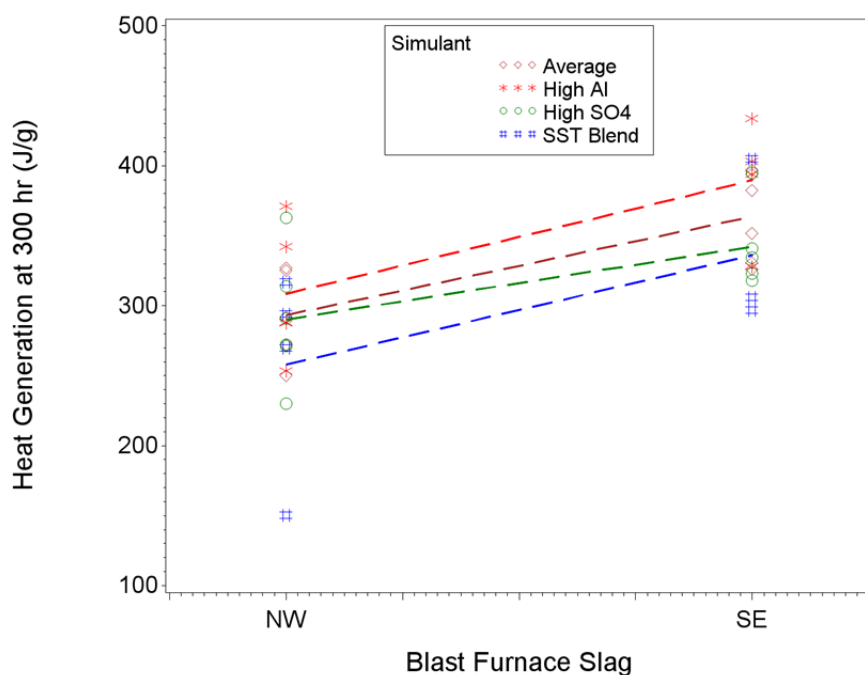


Figure E-17. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Heat Generation at 300 Hours

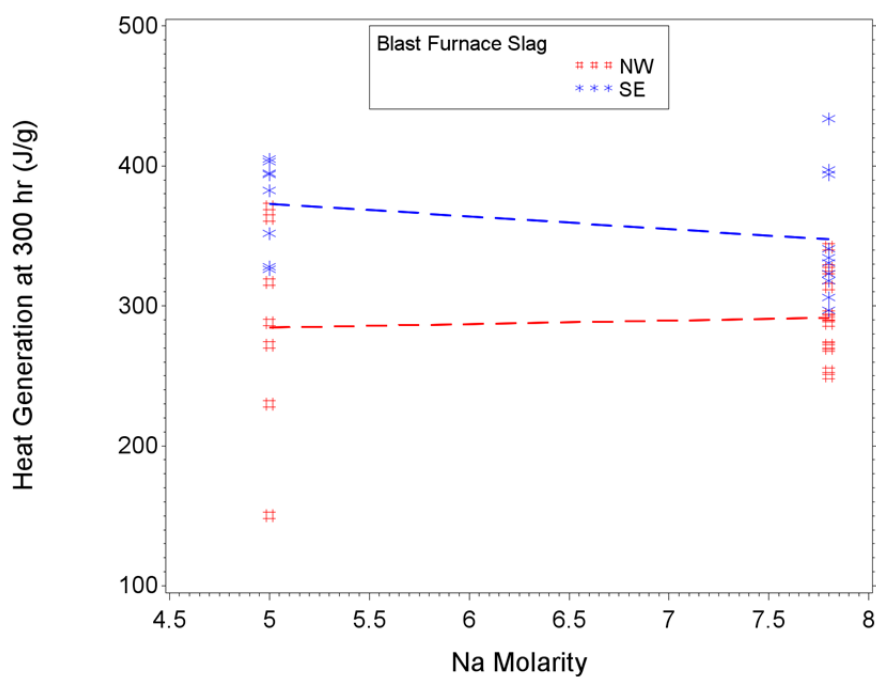


Figure E-18. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Heat Generation at 300 Hours

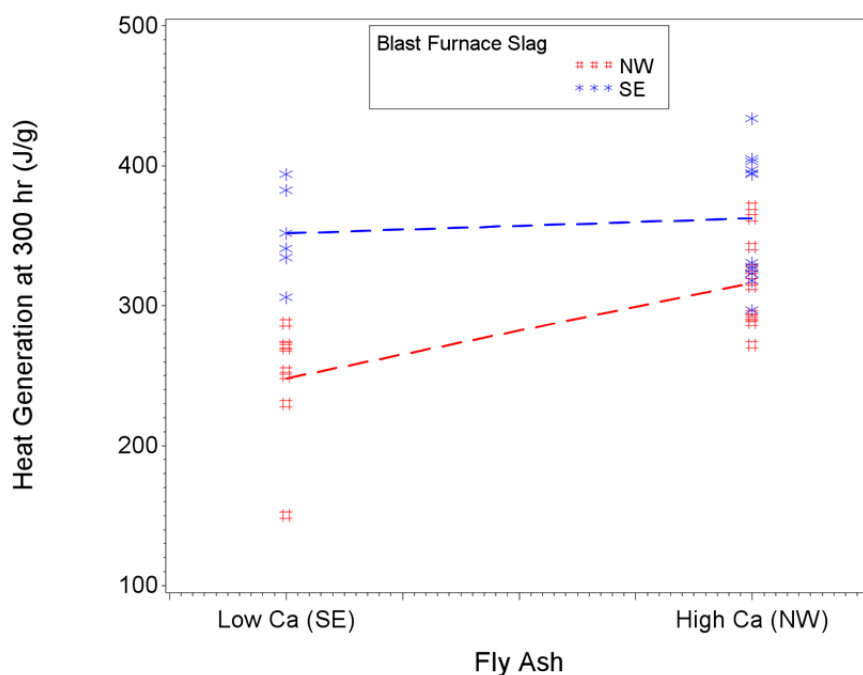


Figure E-19. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Heat Generation at 300 Hours

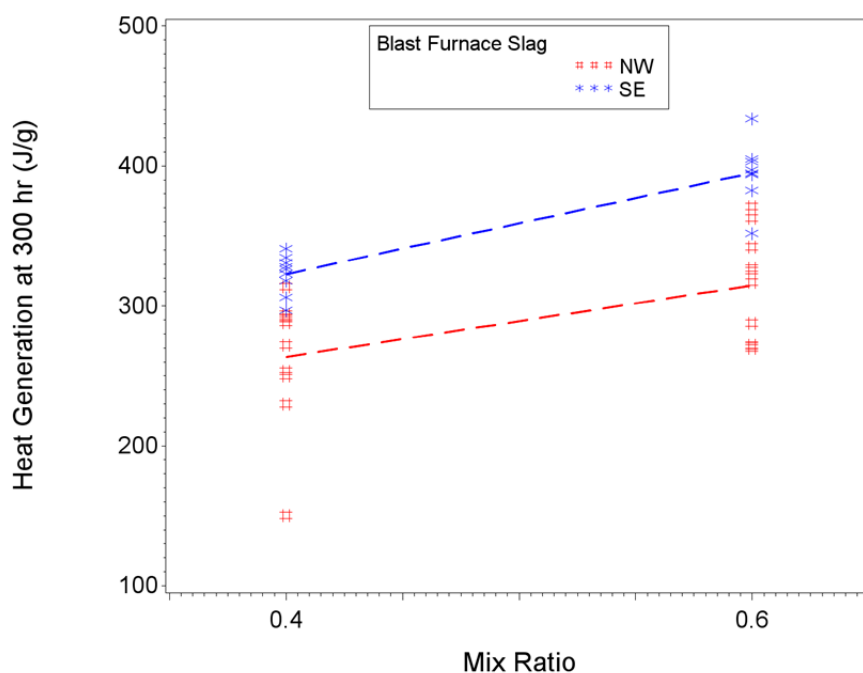


Figure E-20. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Heat Generation at 300 Hours

E.2.2 Heat Generation, Time to Peak Heat Generation

Figure E-21 through Figure E-30 display the data-interaction plots for Time to Peak Heat Generation using the data from Cast Stone Mixes 1–38.

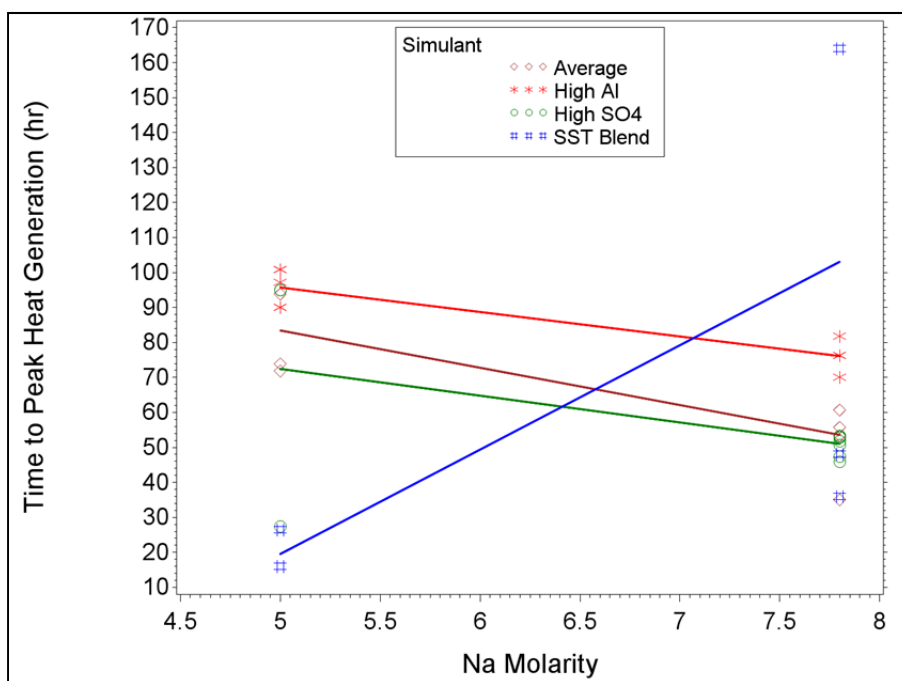


Figure E-21. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Time to Peak Heat Generation

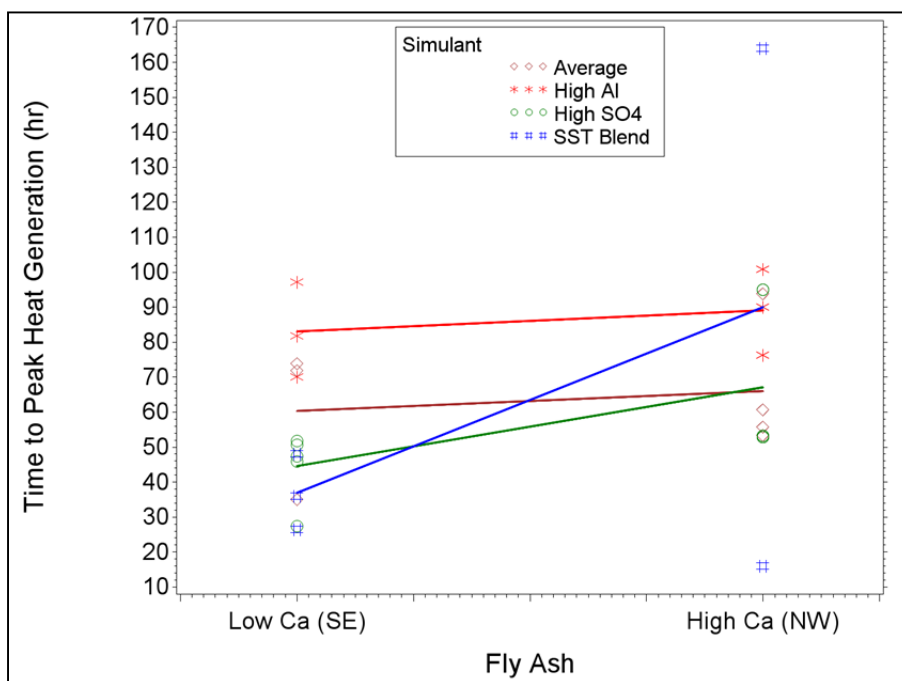


Figure E-22. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Time to Peak Heat Generation

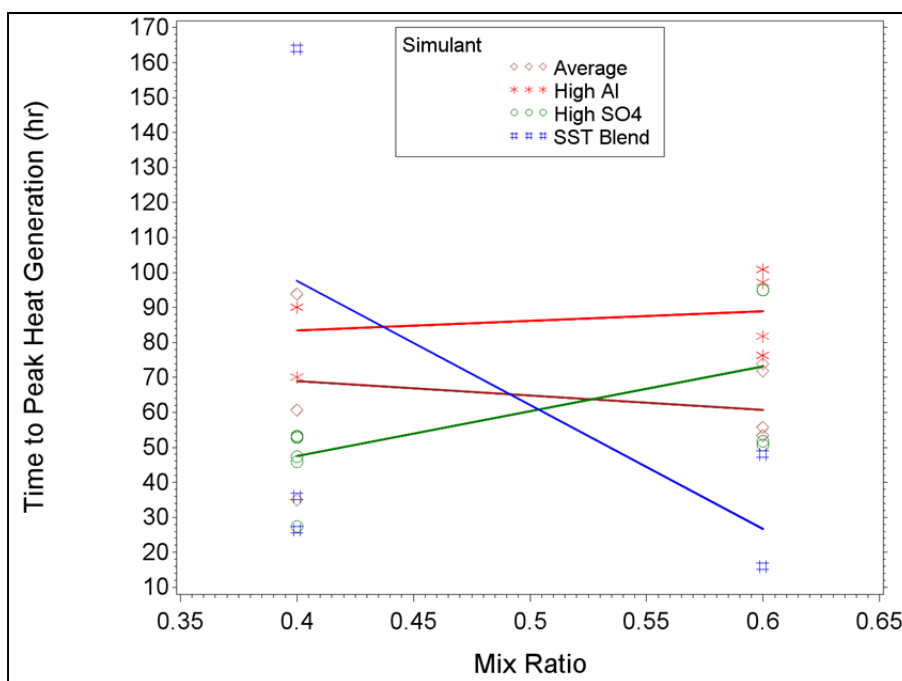


Figure E-23. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Time to Peak Heat Generation

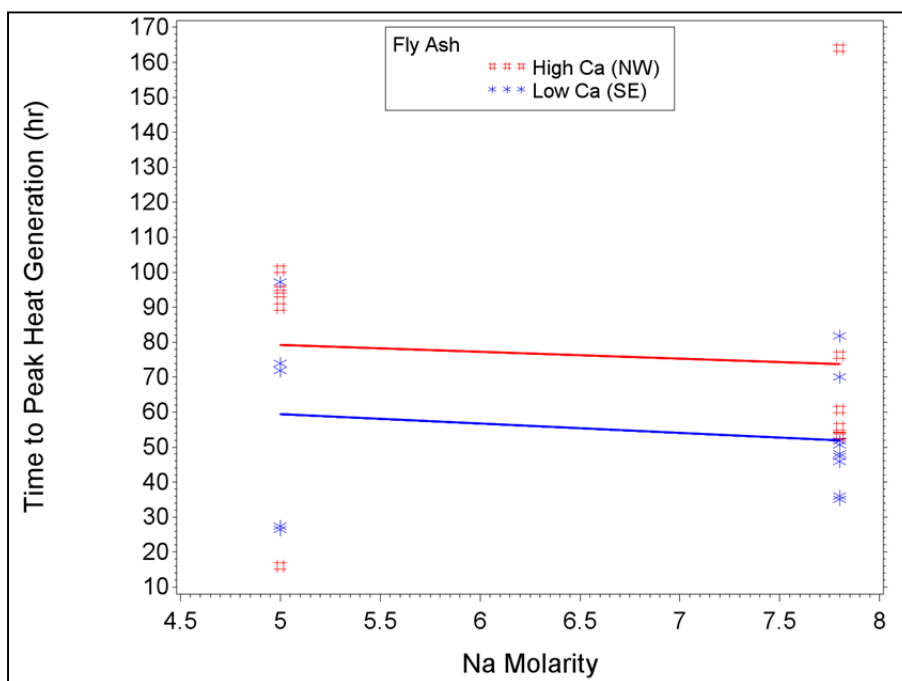


Figure E-24. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Time to Peak Heat Generation

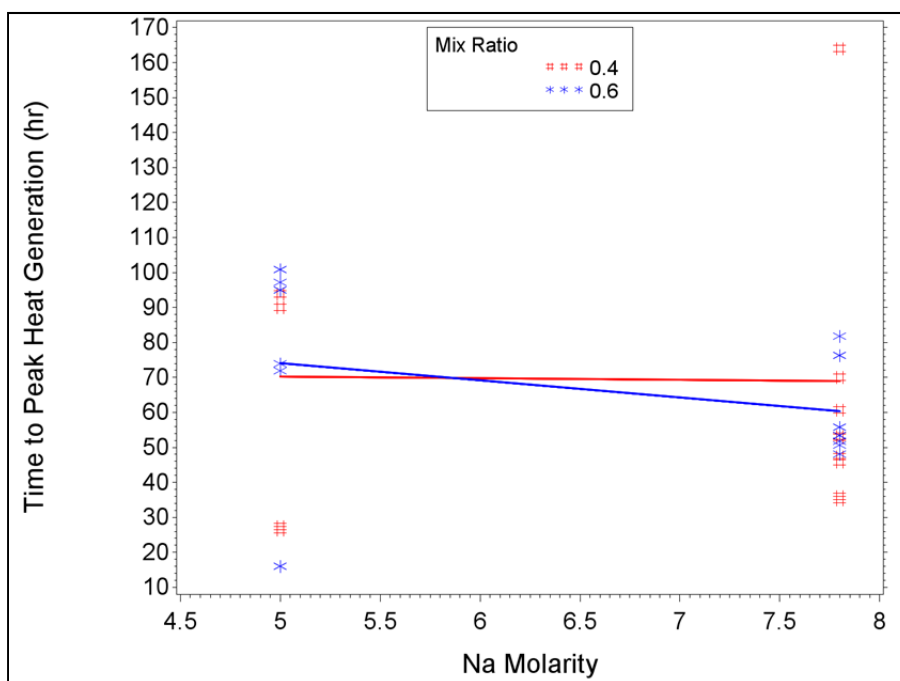


Figure E-25. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Time to Peak Heat Generation

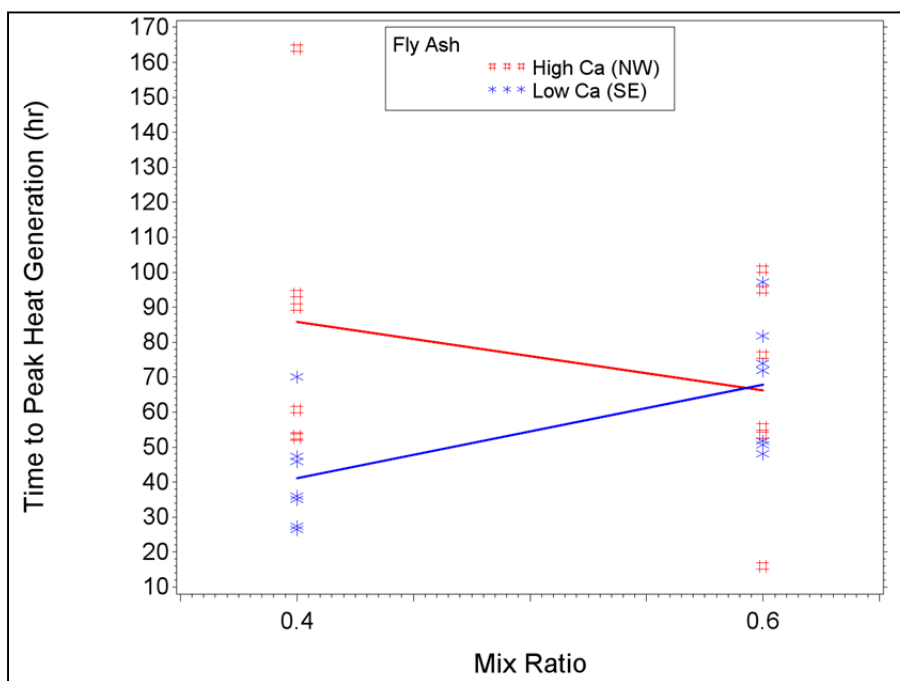


Figure E-26. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Time to Peak Heat Generation

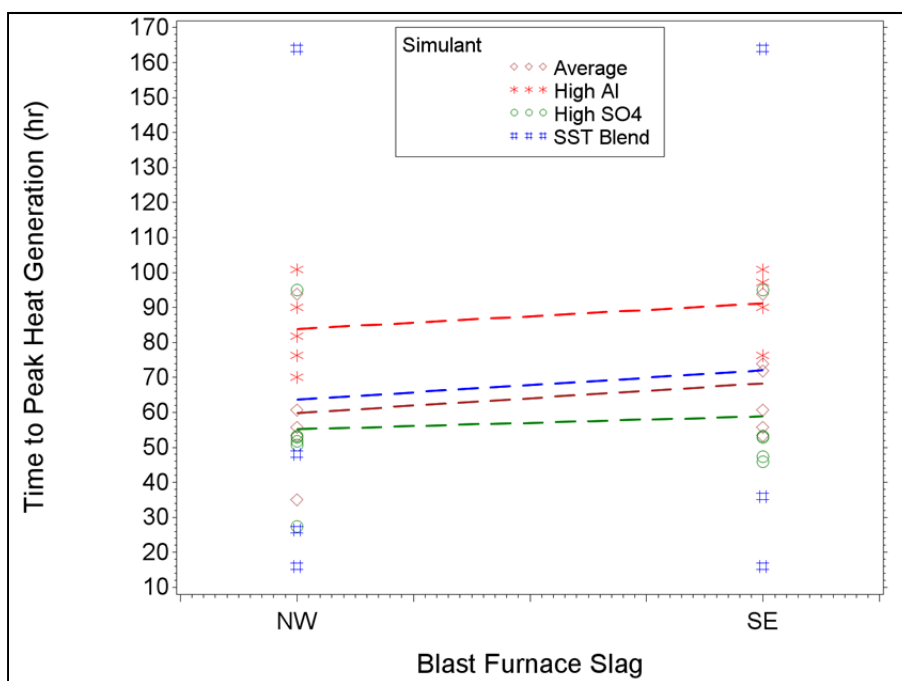


Figure E-27. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Time to Peak Heat Generation

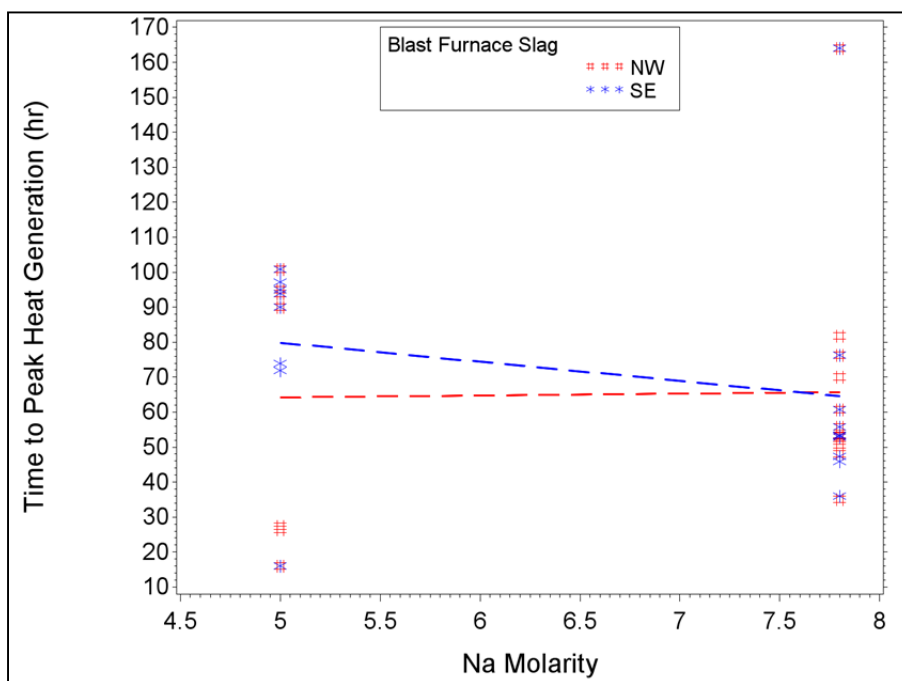


Figure E-28. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Time to Peak Heat Generation

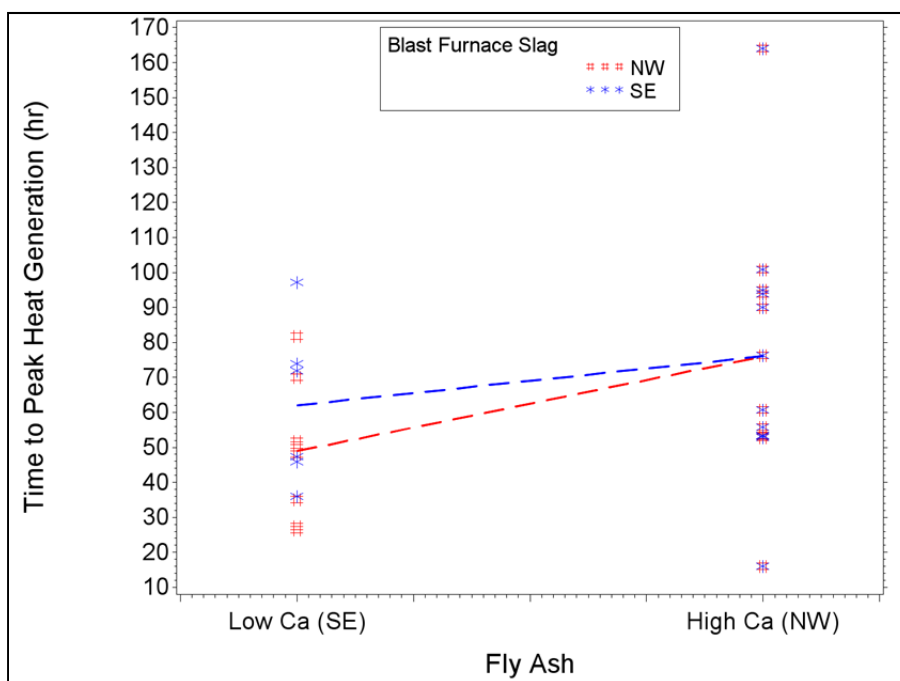


Figure E-29. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Time to Peak Heat Generation

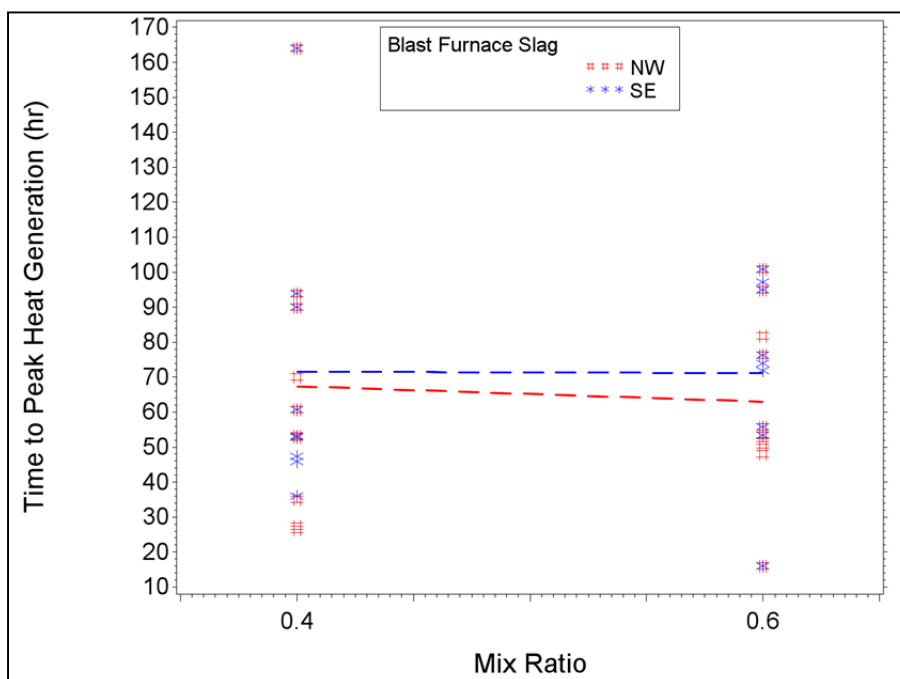


Figure E-30. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Time to Peak Heat Generation

E.3 Compressive Strength

This section presents data-interaction plots of the compressive strength data from for the 26 Cast Stone mixes (1–26) in the screening text matrix. Cylindrical samples of the 26 mixes were made and tested for compressive strength at PNNL. The PNNL data in these plots are from the “Mean” column in Table D-2 in Appendix D.

Figure E-31 through Figure E-40 display the data-interaction plots using the Compressive Strength data measured at PNNL.

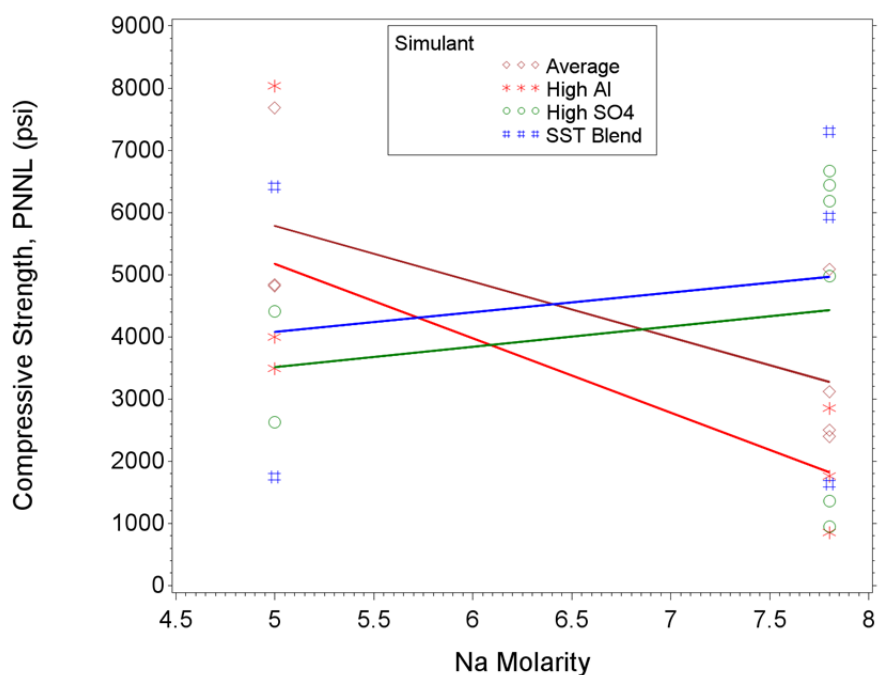


Figure E-31. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Compressive Strength Tested at PNNL

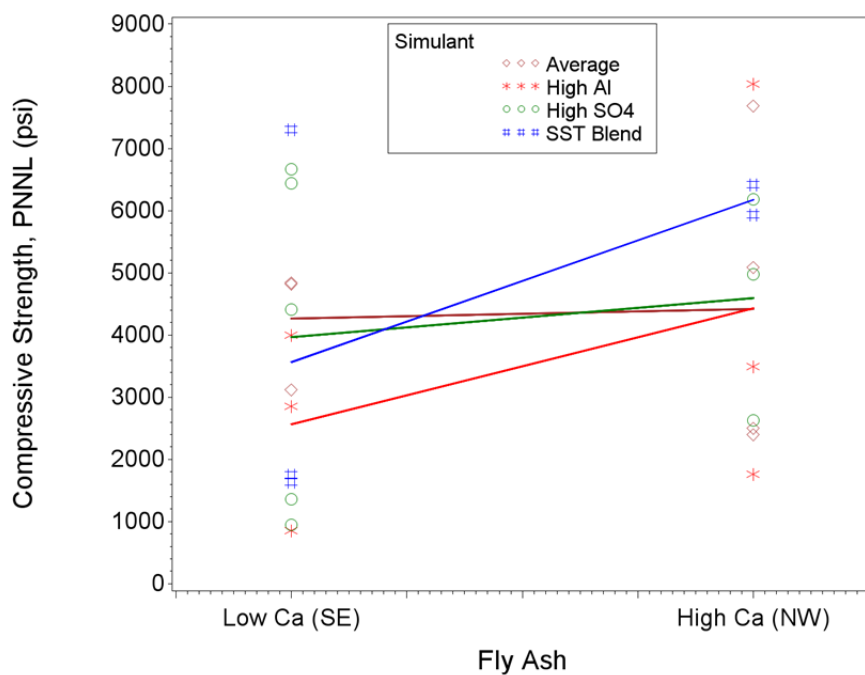


Figure E-32. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Compressive Strength Tested at PNNL

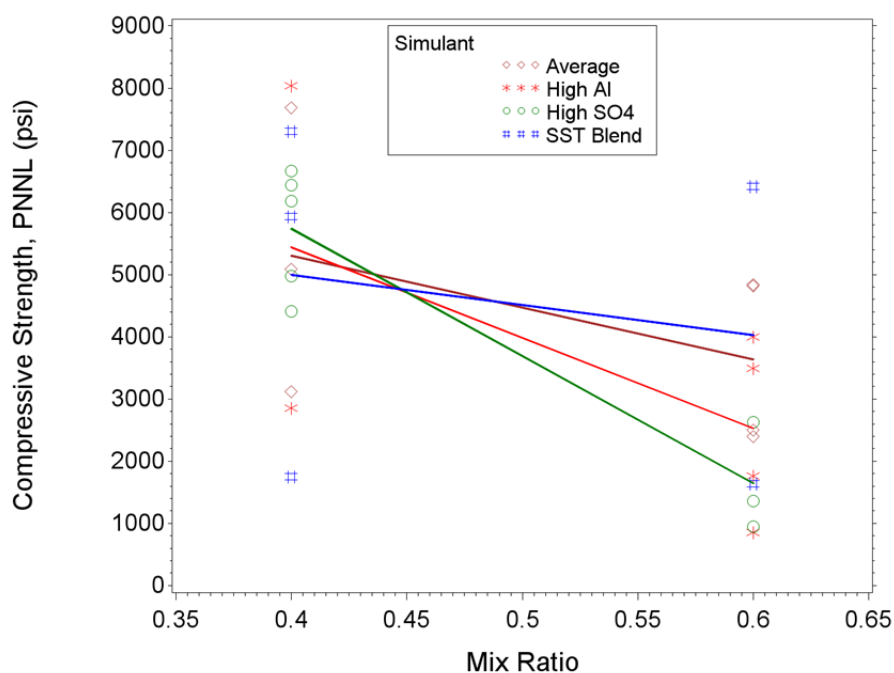


Figure E-33. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Compressive Strength Tested at PNNL

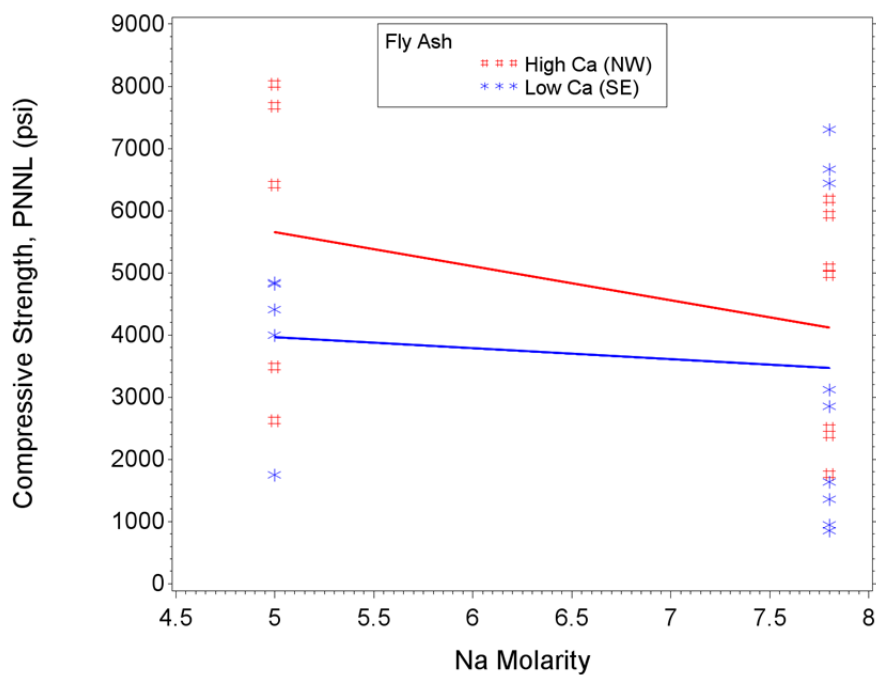


Figure E-34. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Compressive Strength Tested at PNNL

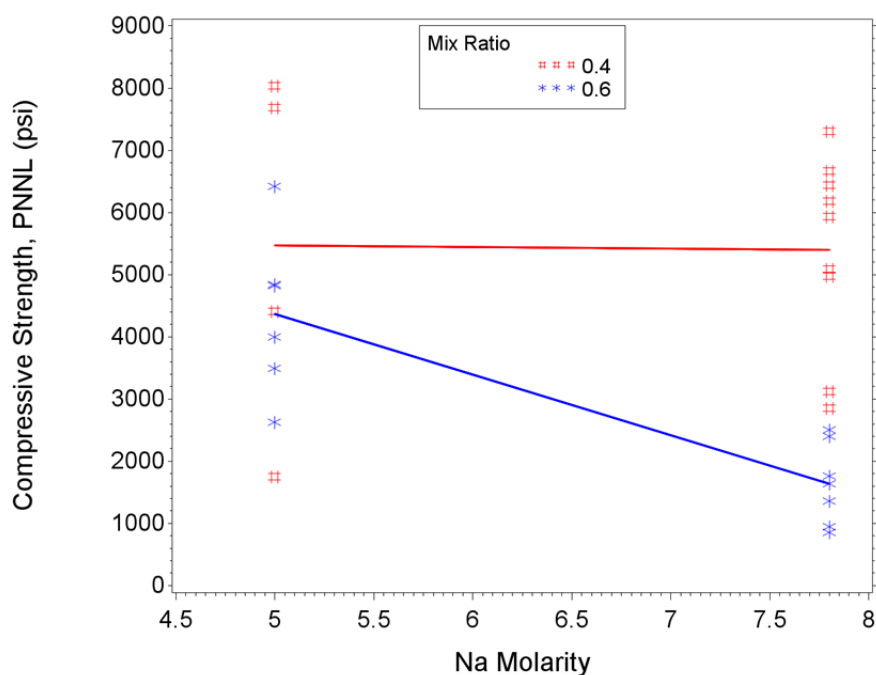


Figure E-35. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Compressive Strength Tested at PNNL

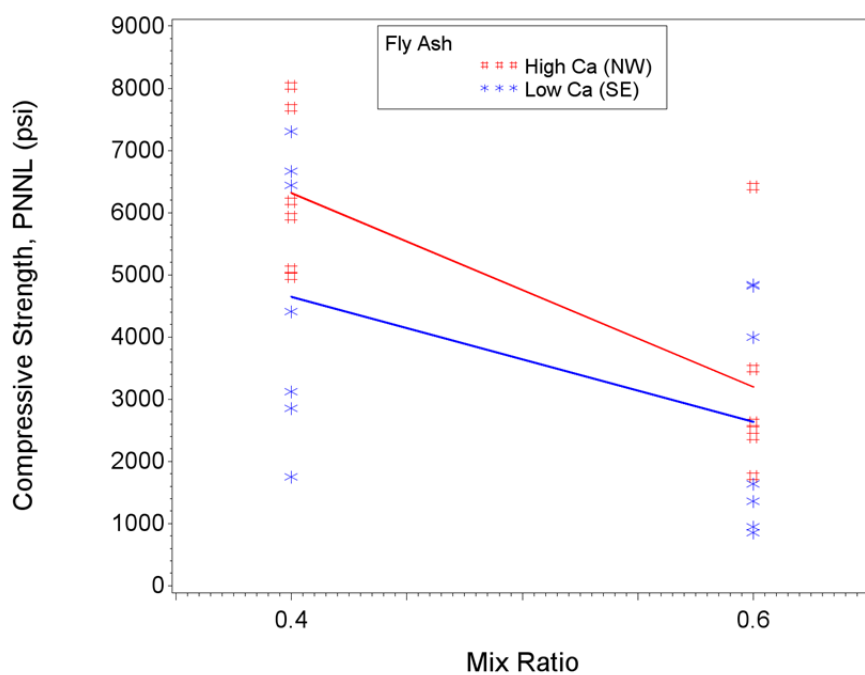


Figure E-36. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Compressive Strength Tested at PNNL

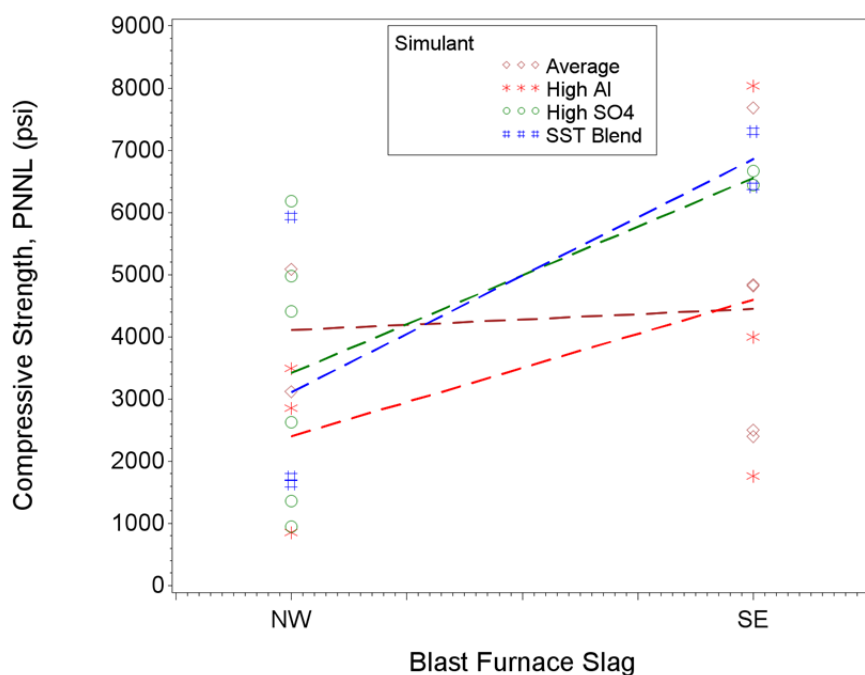


Figure E-37. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Compressive Strength Tested at PNNL

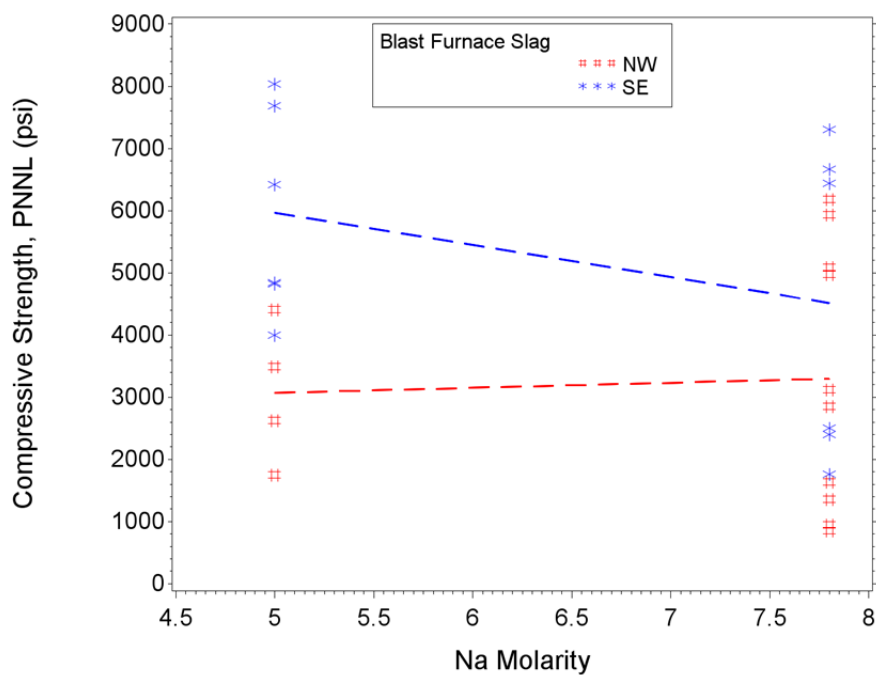


Figure E-38. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Compressive Strength Tested at PNNL

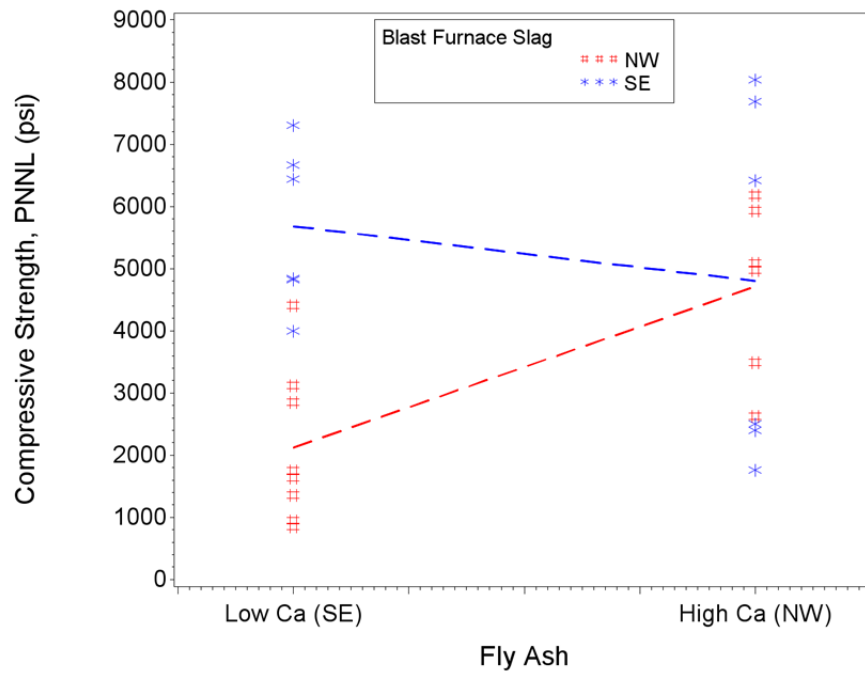


Figure E-39. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Compressive Strength Tested at PNNL

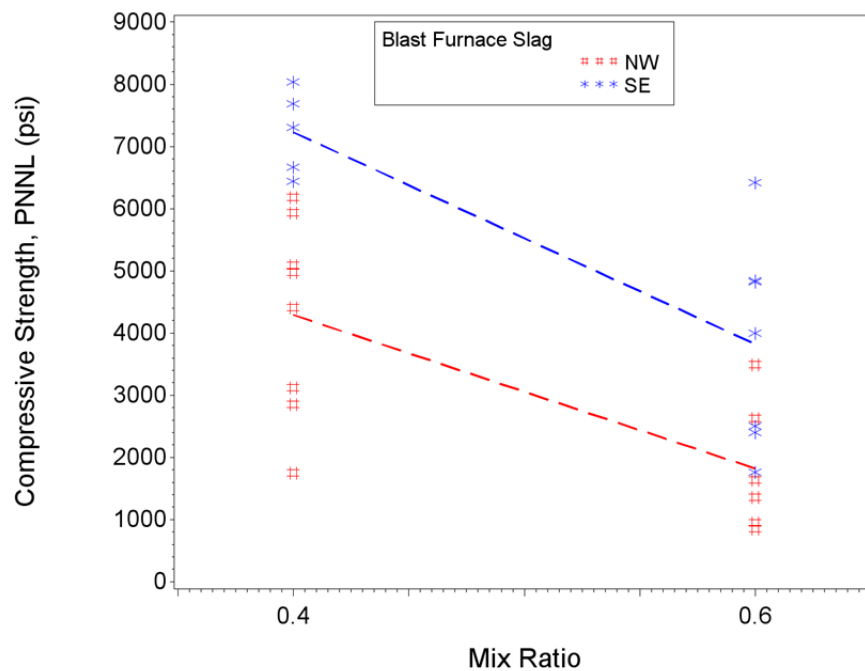


Figure E-40. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Compressive Strength Tested at PNNL

E.4 Leachability Index, EPA Draft Method 1315 Leach Test

This section presents data-interaction plots for averaged leachability indices (LIs) over the cumulative leach times of 28 to 63 cumulative days of Na, NO₃, I, Tc, and Cr. Plots are not included for NO₂ because the nitrate and nitrite data are very strongly correlated (see Figure 8-3). Leachability indices were calculated from effective diffusivities using Equation (7-2). The averaged LIs were determined using Equation (D-1). The data plotted in the figures are listed in the “Mean” column for each of the leachates in Table D-15 in Appendix D.

E.4.1 *Sodium Leachability Index*

Figure E-41 through Figure E-50 display the data-interaction plots for Na LI of the 26 Cast Stone mixes in the screening text matrix.

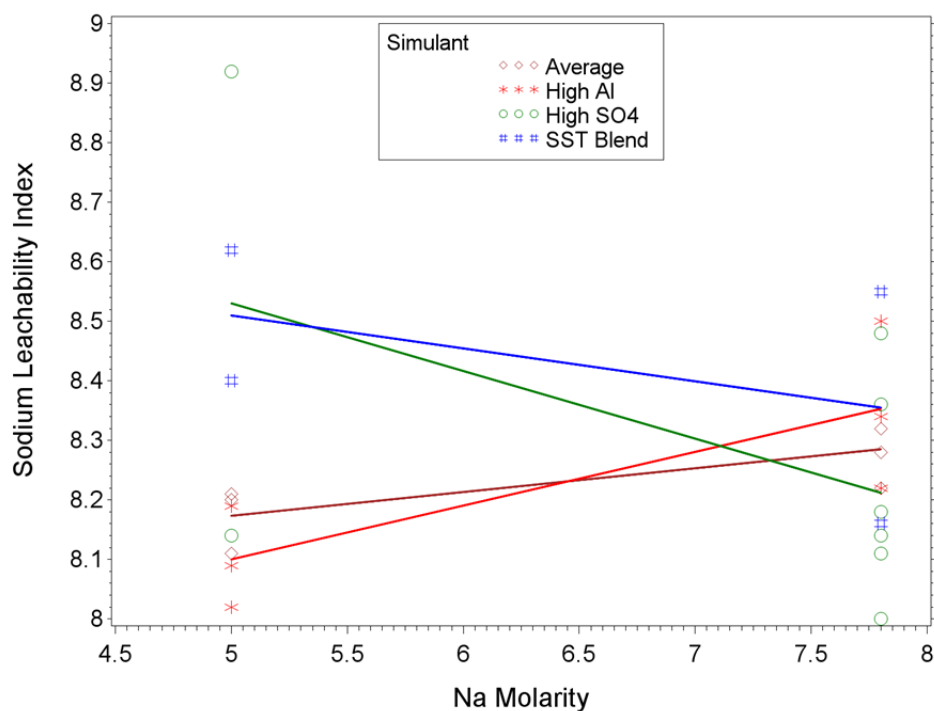


Figure E-41. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Sodium Leachability Index

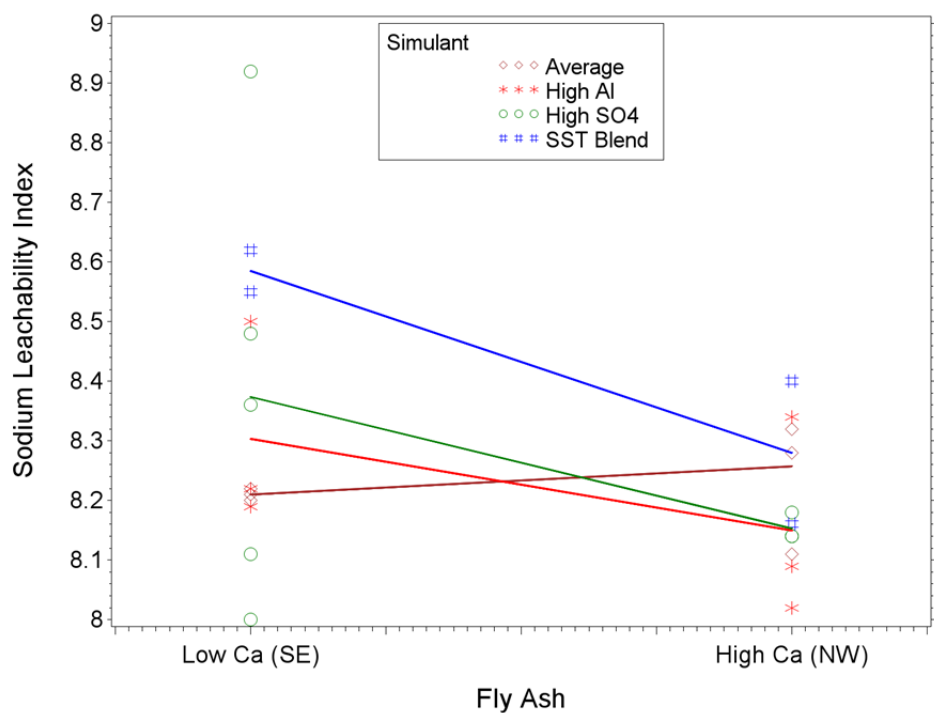


Figure E-42. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Sodium Leachability Index

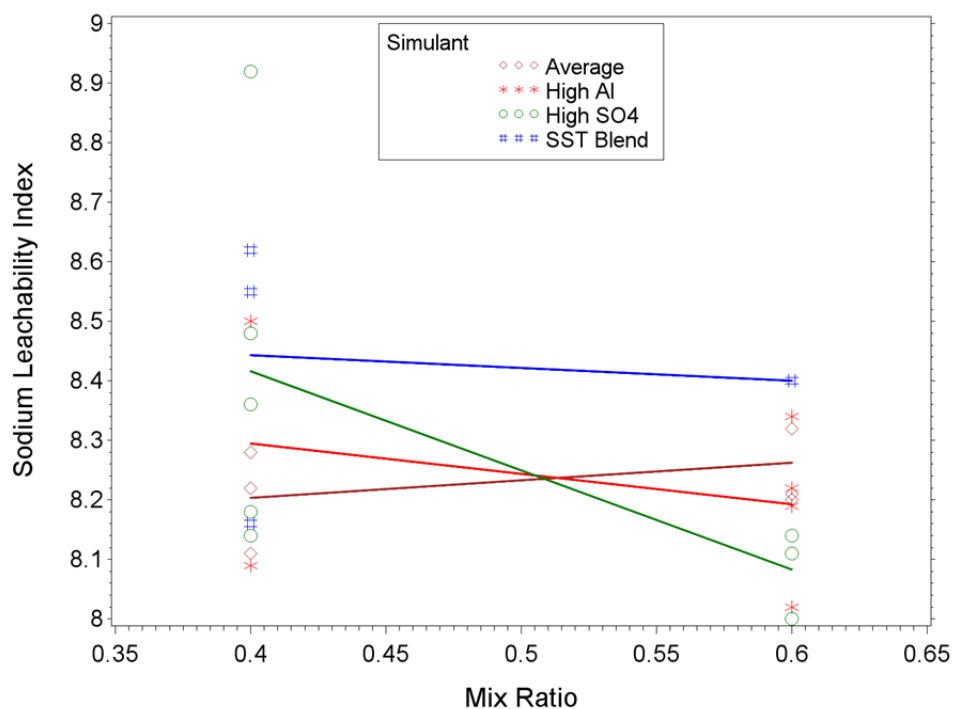


Figure E-43. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Sodium Leachability Index

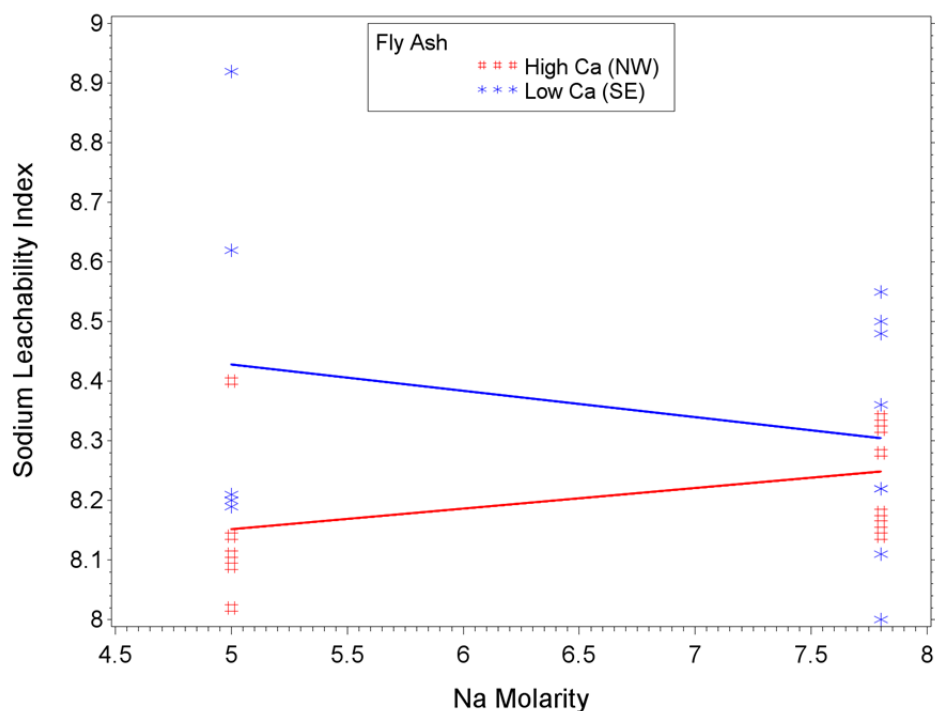


Figure E-44. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Sodium Leachability Index

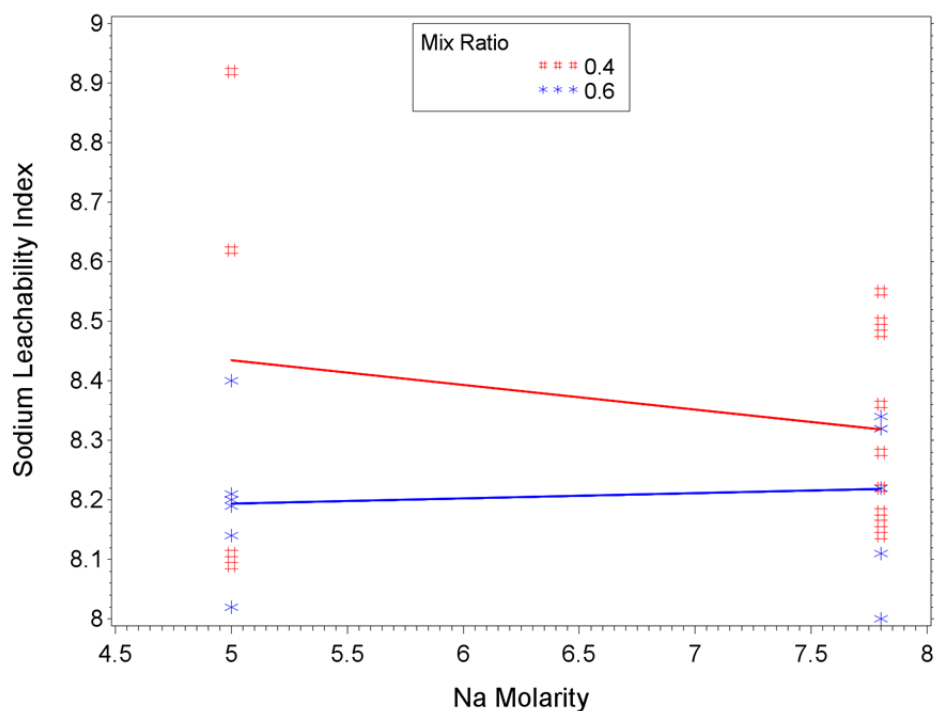


Figure E-45. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Sodium Leachability Index

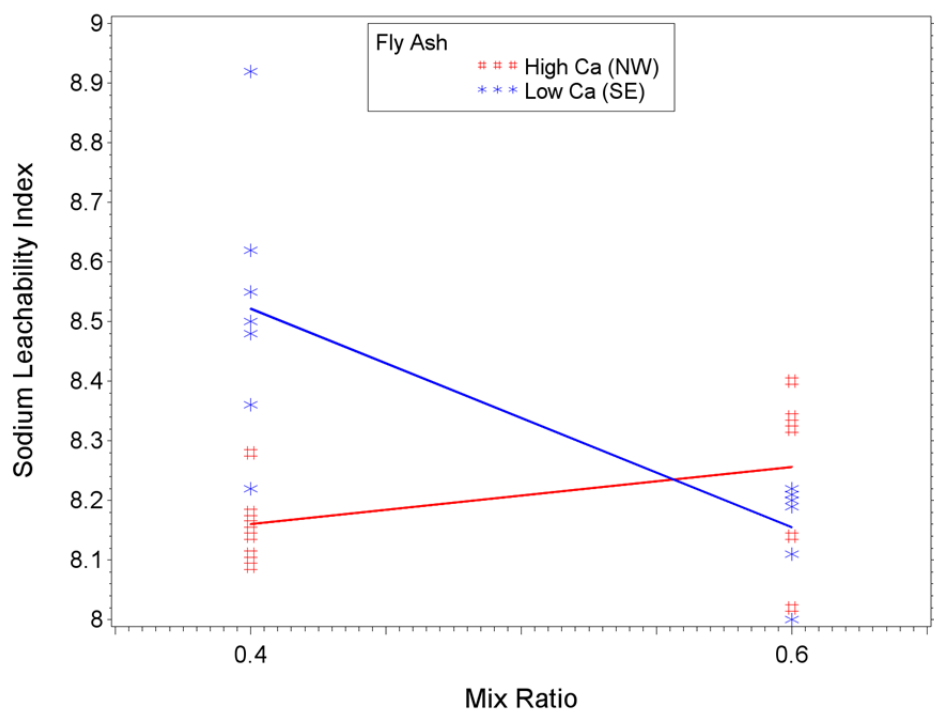


Figure E-46. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Sodium Leachability Index

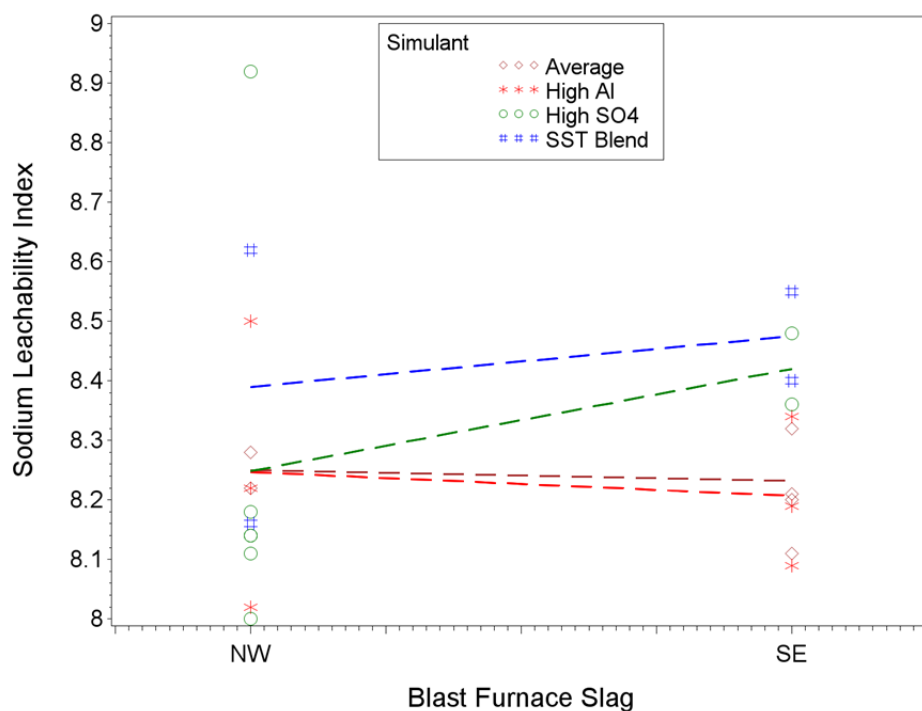


Figure E-47. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Sodium Leachability Index

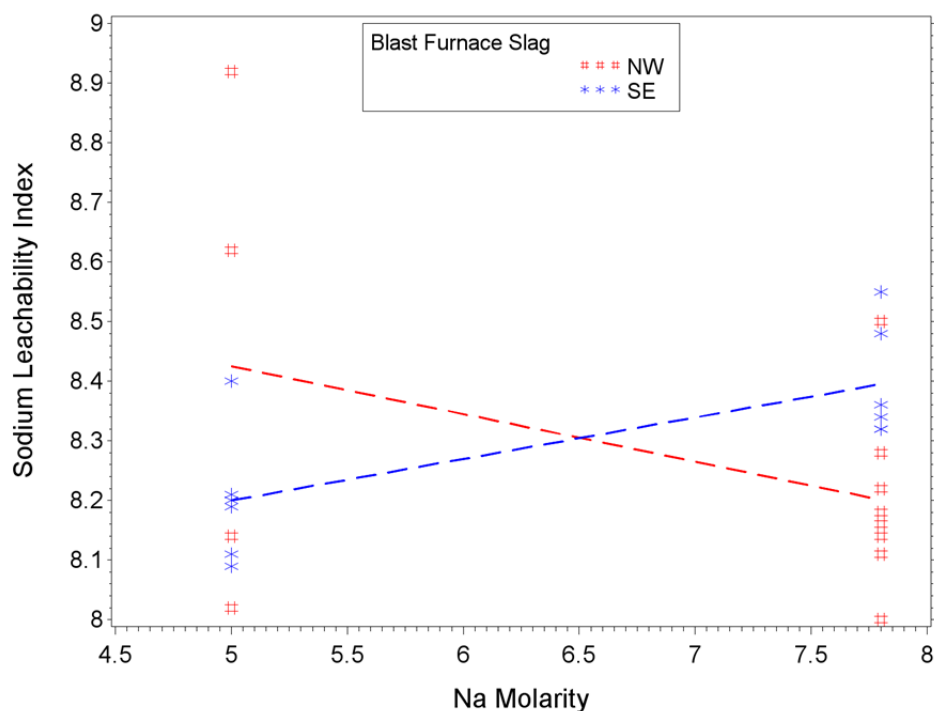


Figure E-48. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Sodium Leachability Index

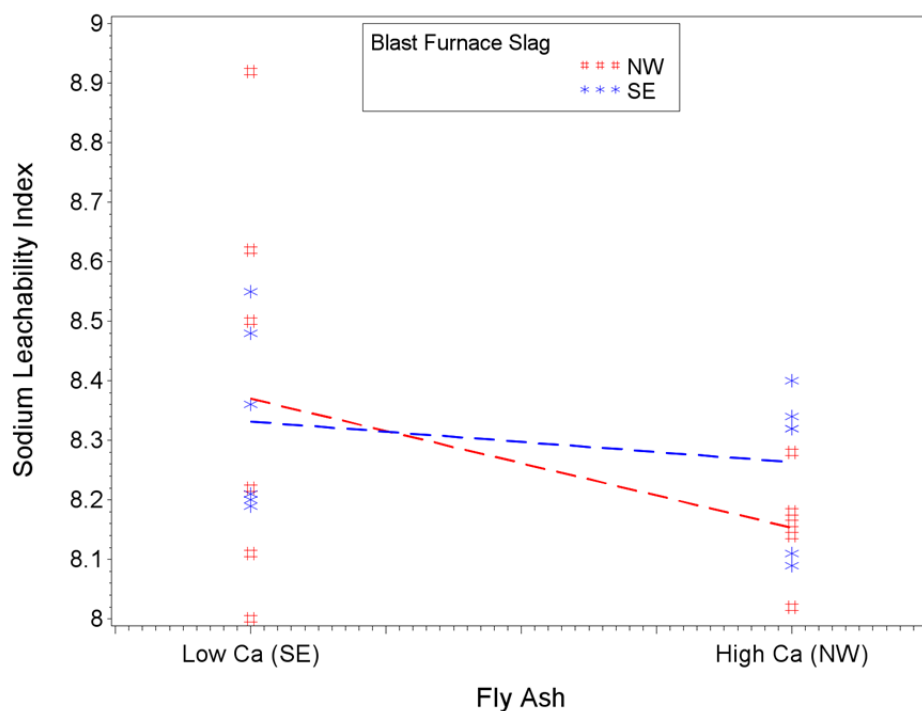


Figure E-49. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Sodium Leachability Index

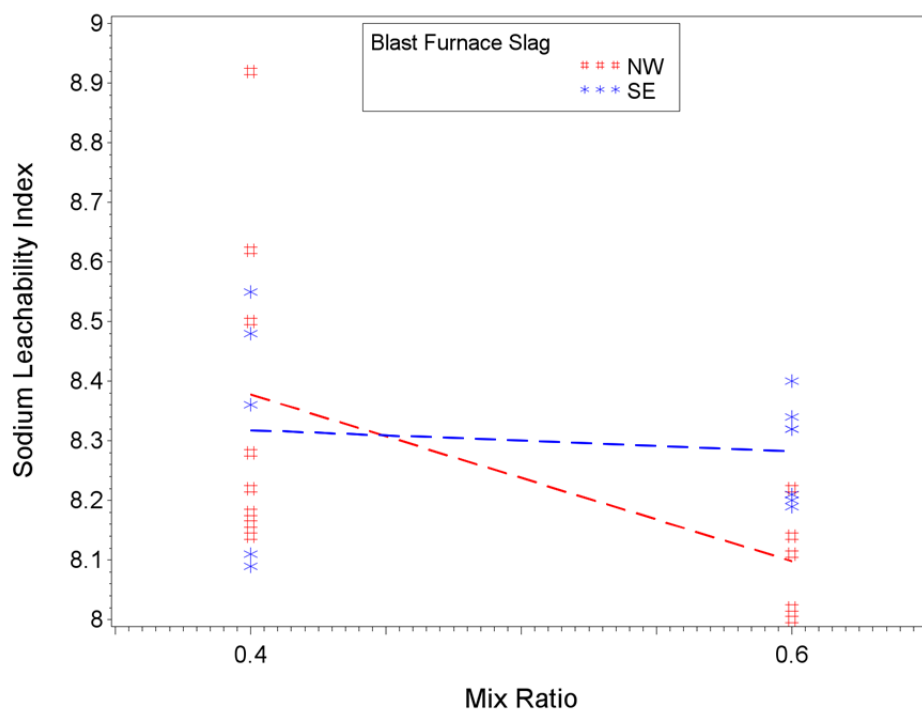


Figure E-50. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Sodium Leachability Index

E.4.2 *Nitrate Leachability Index*

Figure E-51 through Figure E-60 display the data-interaction plots for the averaged NO_3 LI of the 26 Cast Stone mixes in the screening text matrix.

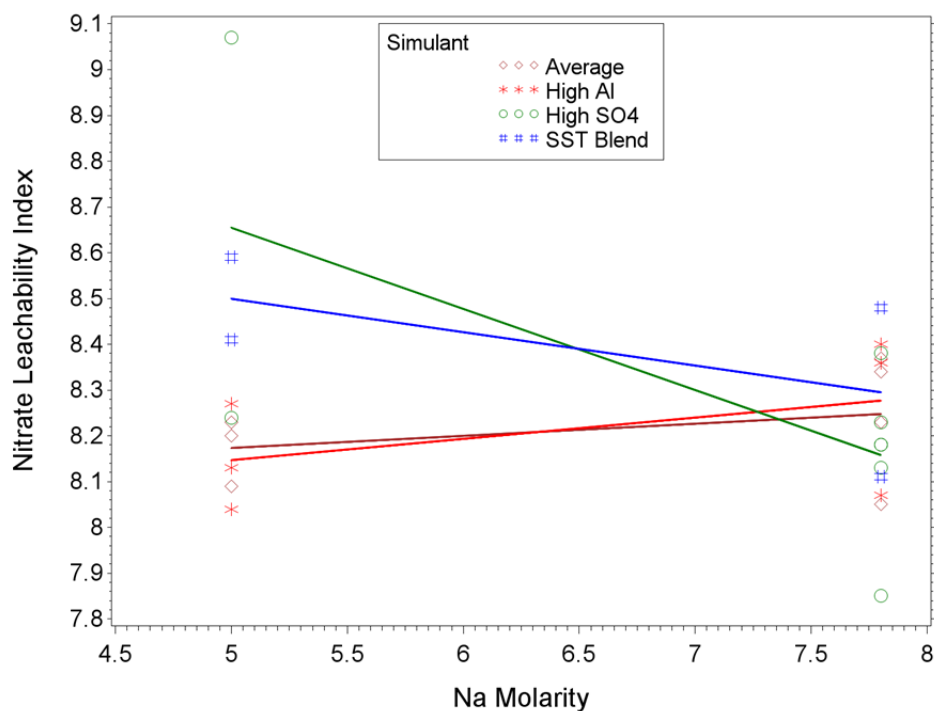


Figure E-51. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Nitrate Leachability Index

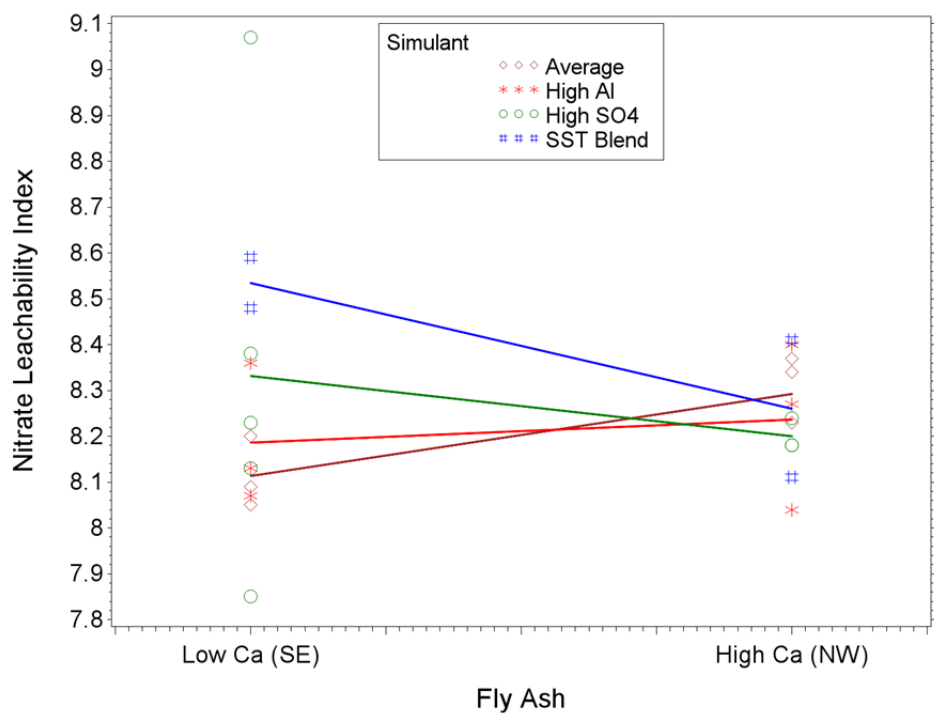


Figure E-52. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Nitrate Leachability Index

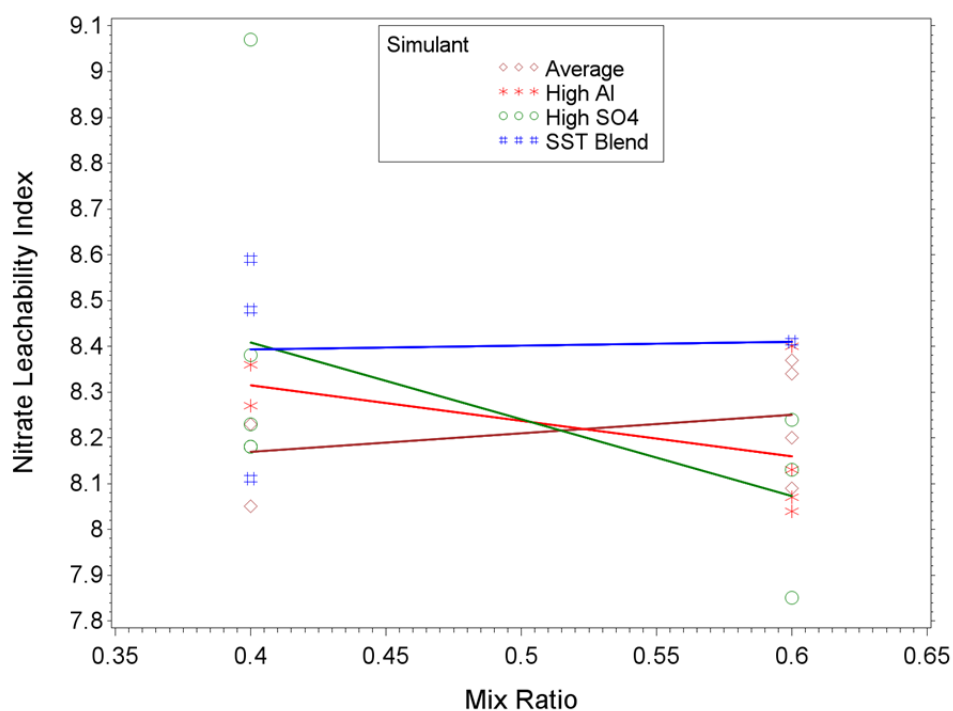


Figure E-53. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Nitrate Leachability Index

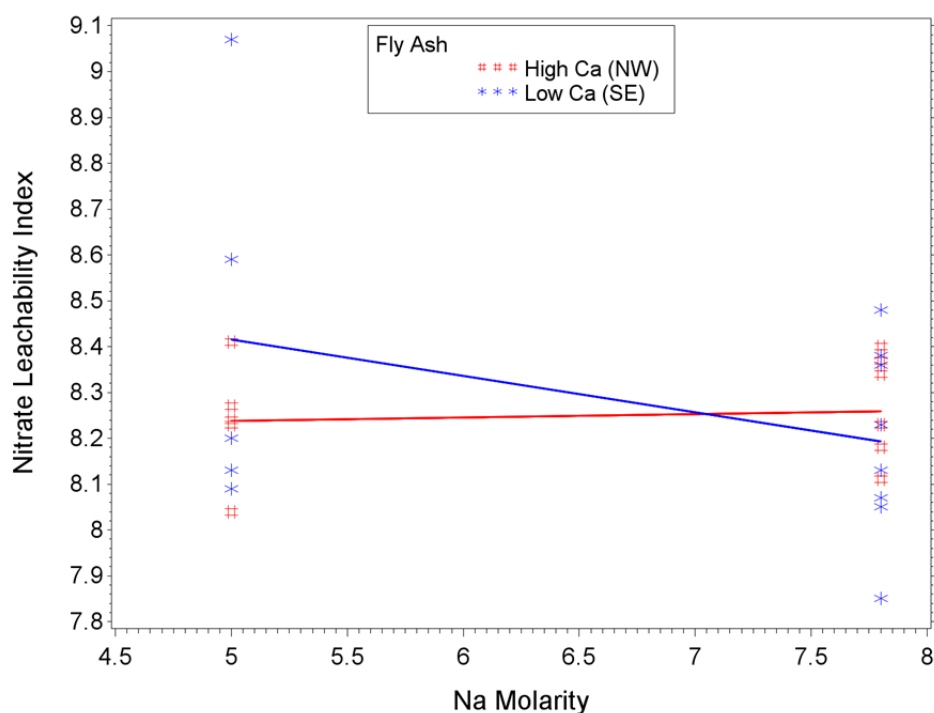


Figure E-54. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Nitrate Leachability Index

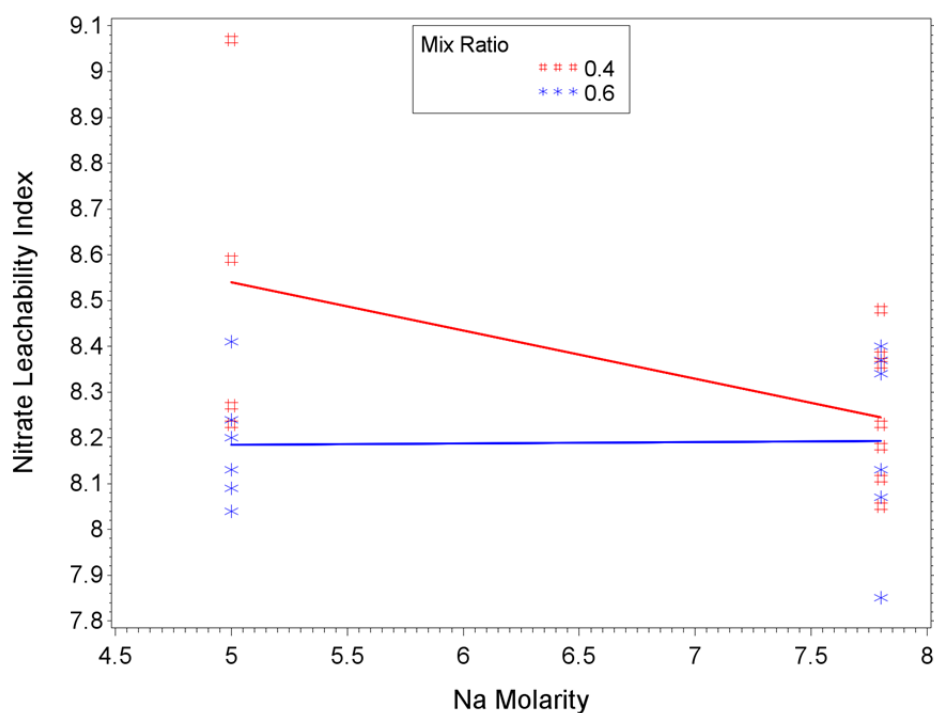


Figure E-55. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Nitrate Leachability Index

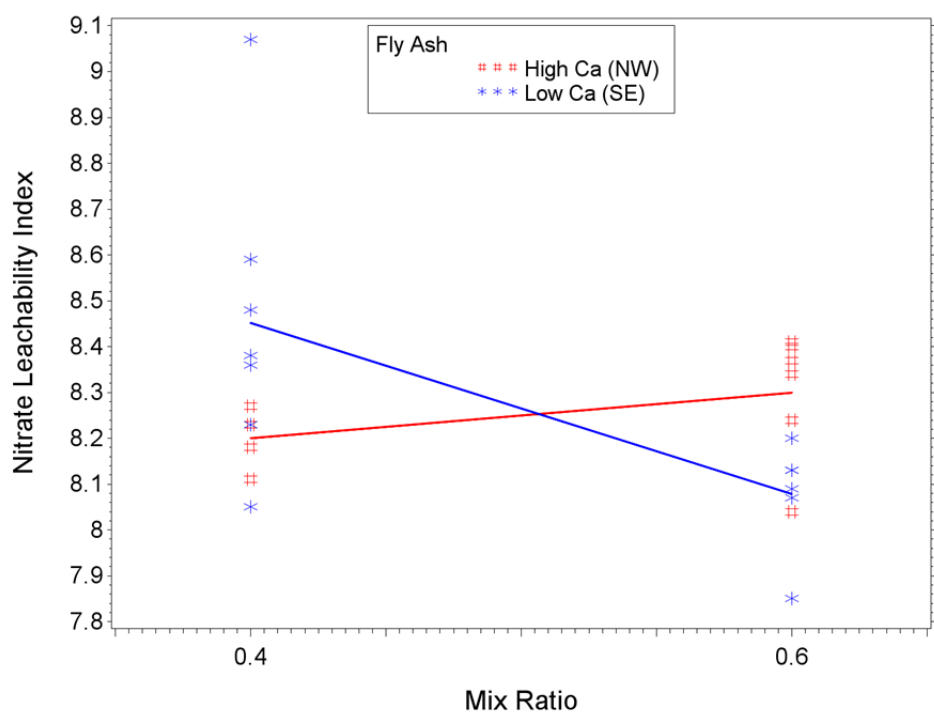


Figure E-56. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Nitrate Leachability Index

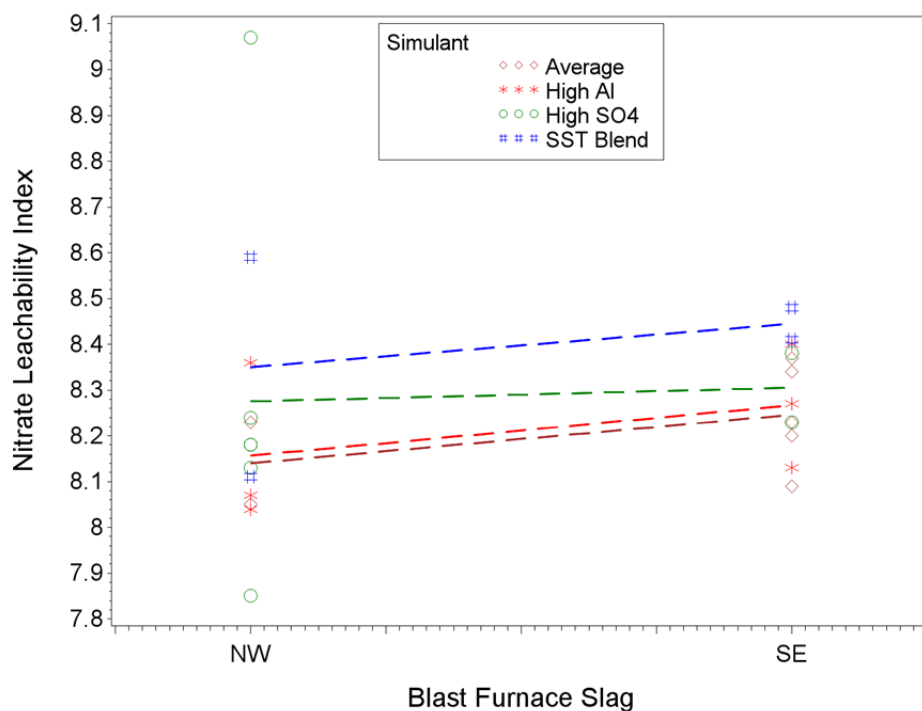


Figure E-57. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Nitrate Leachability Index

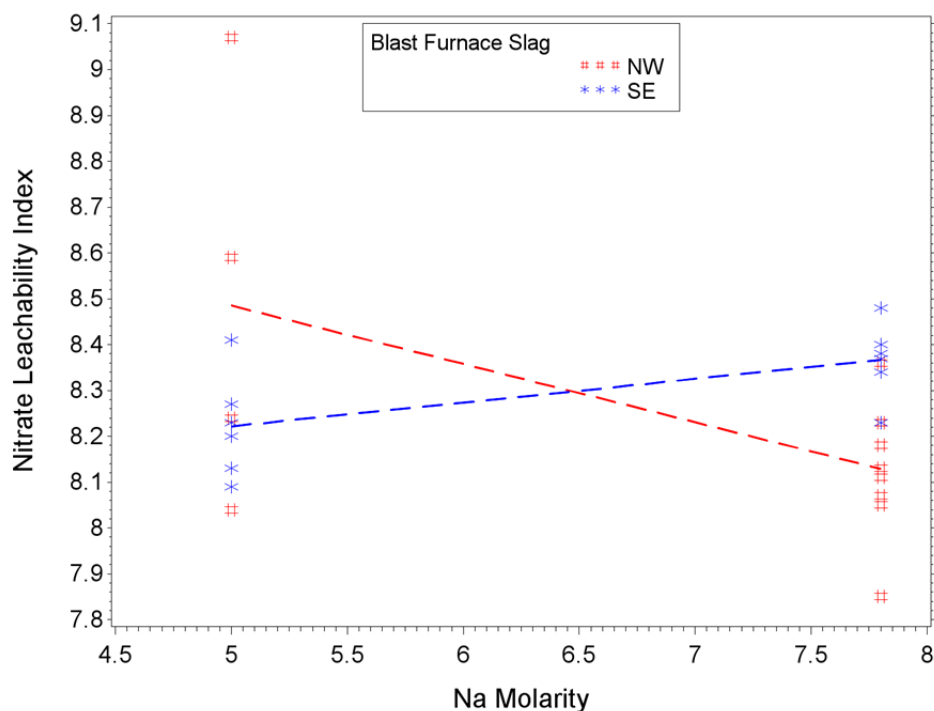


Figure E-58. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Nitrate Leachability Index

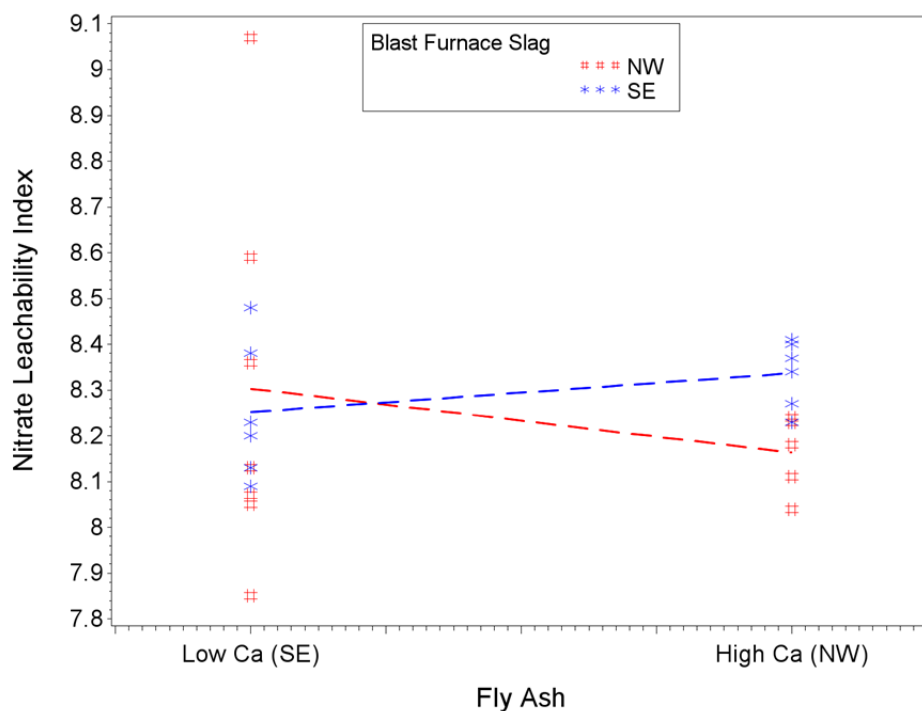


Figure E-59. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Nitrate Leachability Index

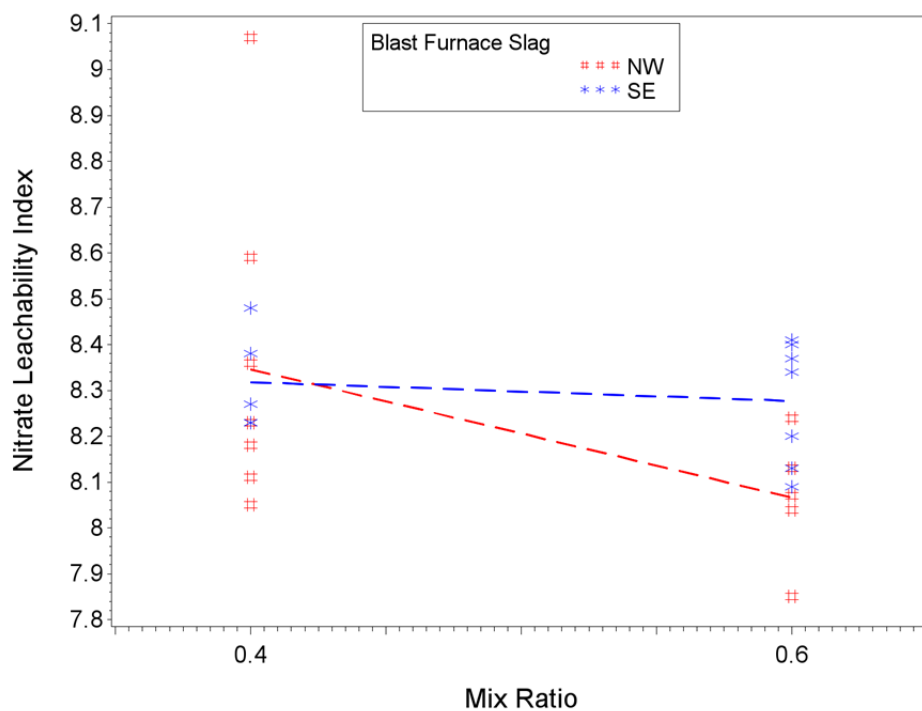


Figure E-60. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Nitrate Leachability Index

E.4.3 *Iodine Leachability Index*

Figure E-61 through Figure E-70 display the data-interaction plots for the averaged I LI of the 26 Cast Stone mixes in the screening text matrix.

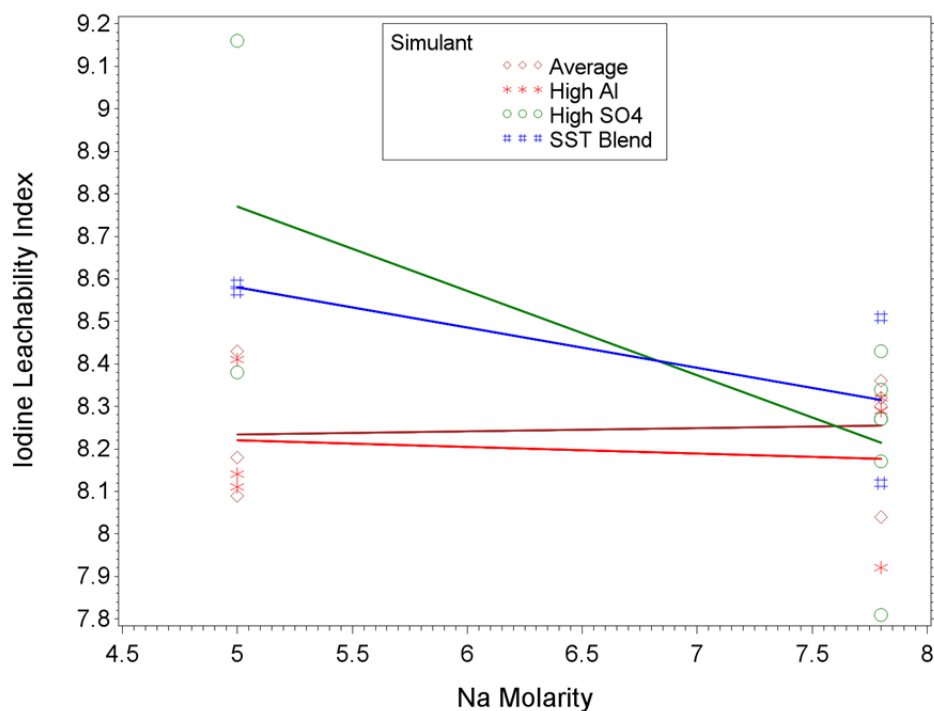


Figure E-61. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Iodine Leachability Index

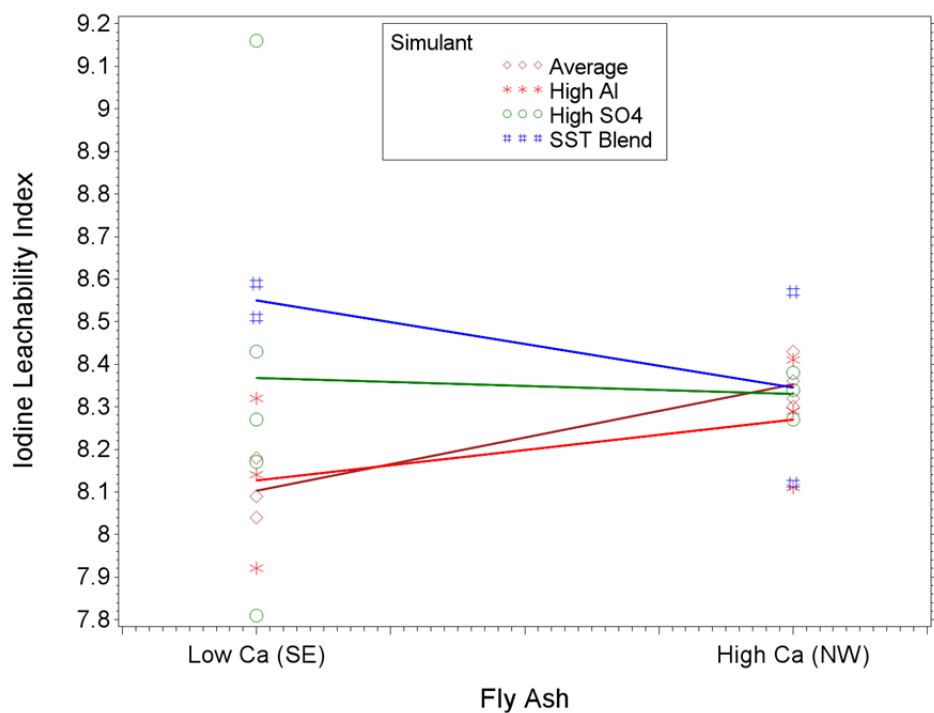


Figure E-62. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Iodine Leachability Index

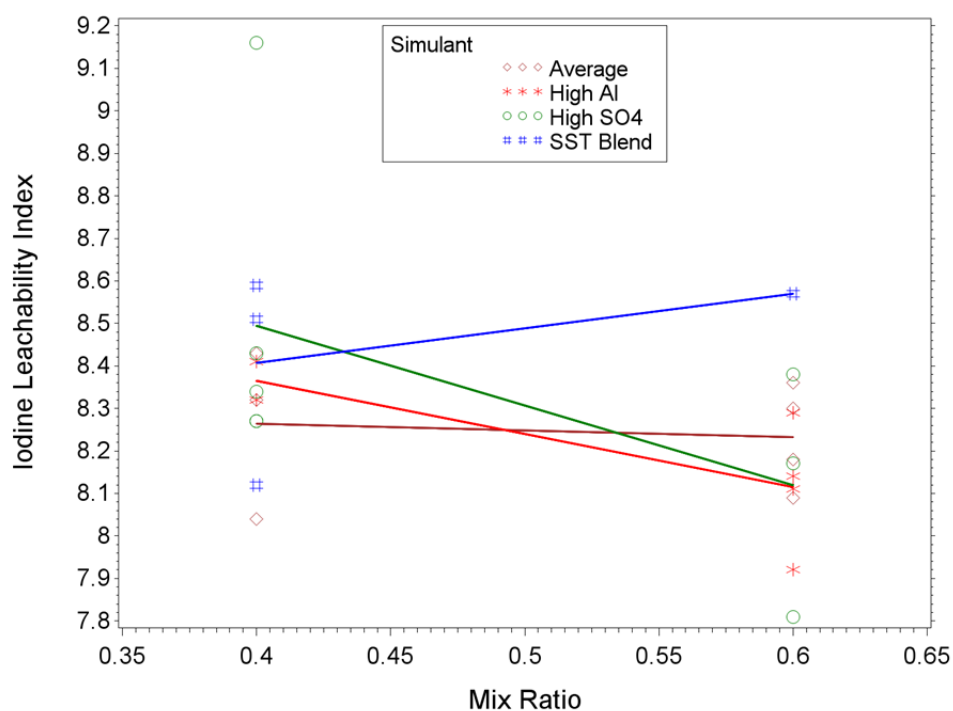


Figure E-63. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Iodine Leachability Index

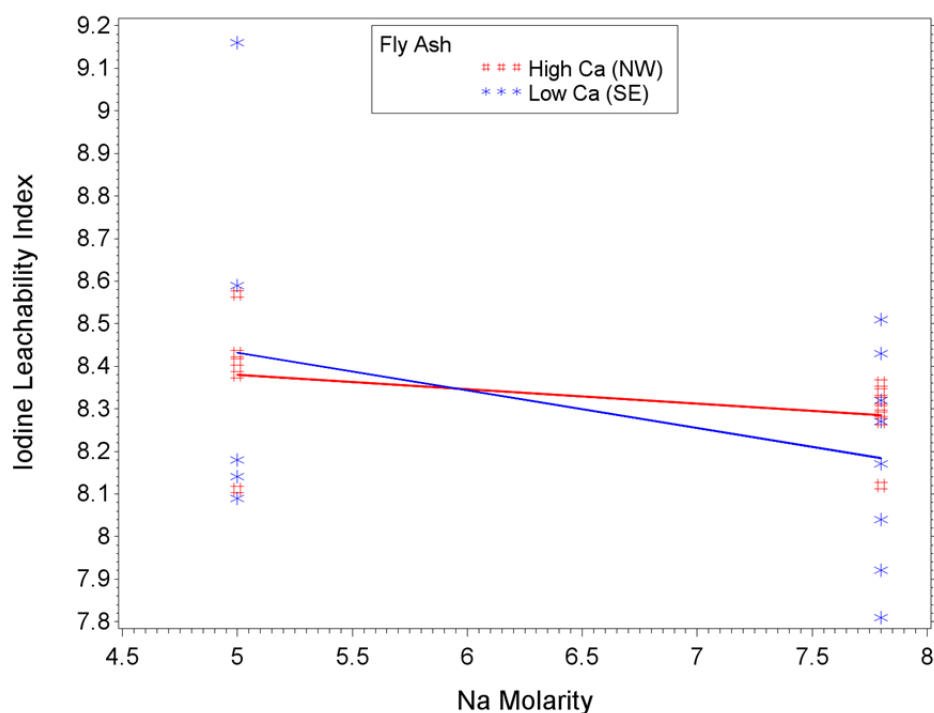


Figure E-64. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Iodine Leachability Index

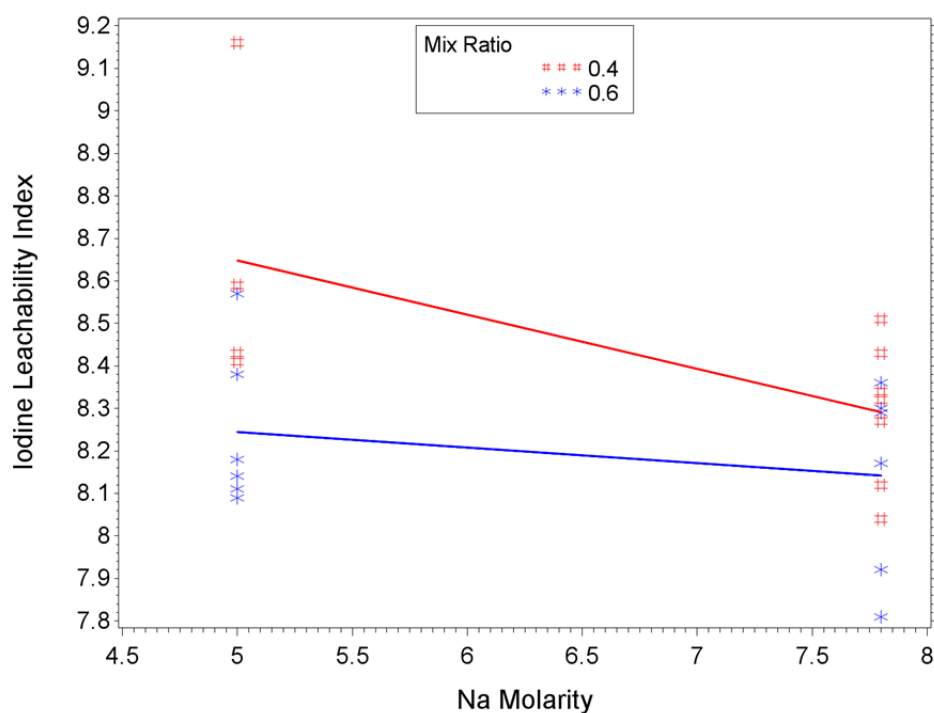


Figure E-65. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Iodine Leachability Index

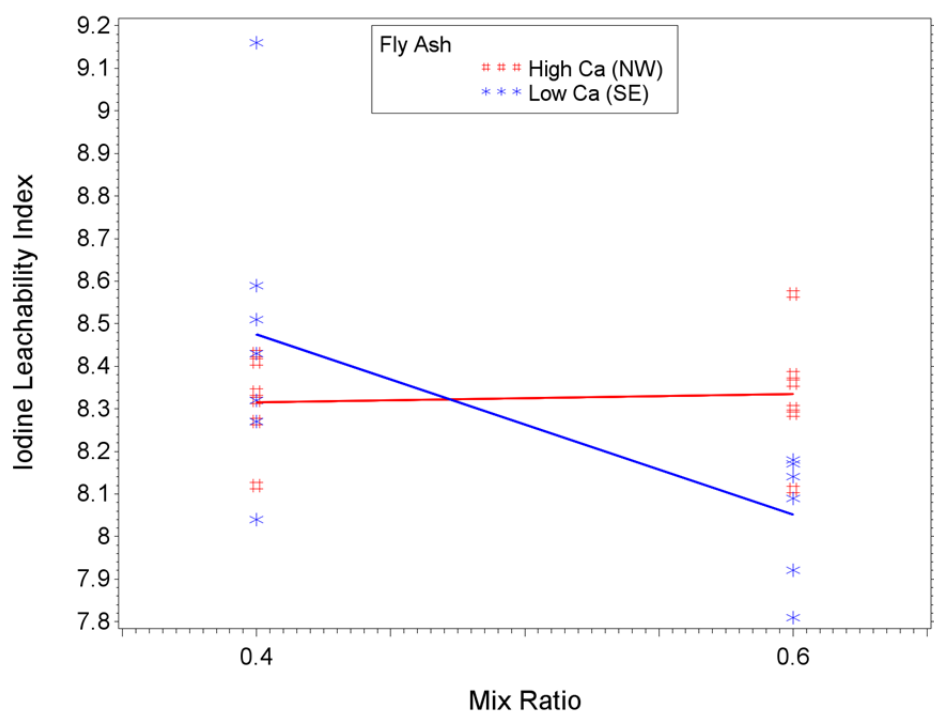


Figure E-66. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Iodine Leachability Index

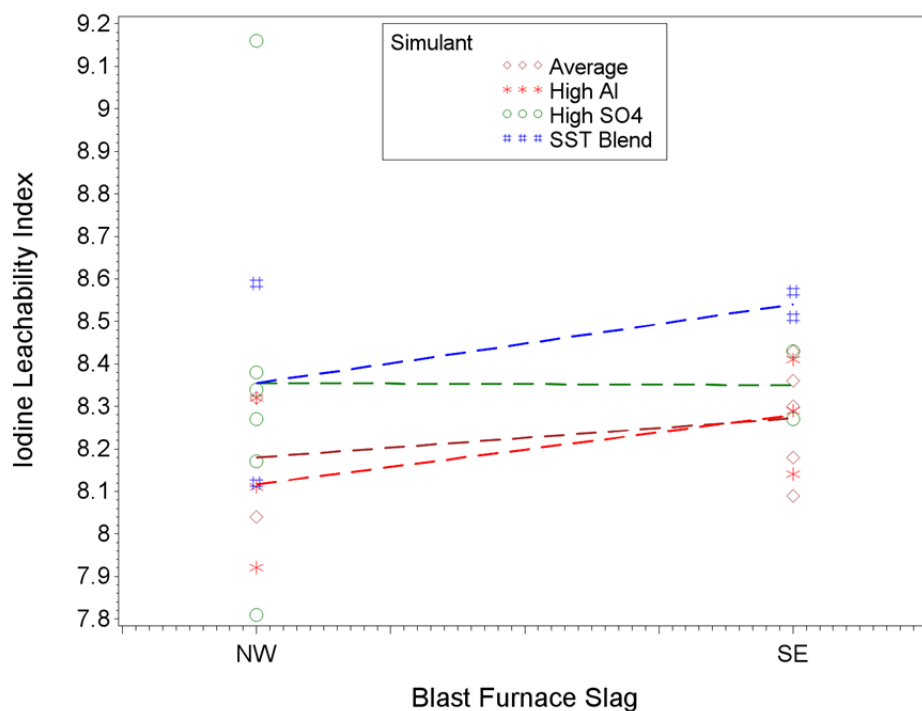


Figure E-67. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Iodine Leachability Index

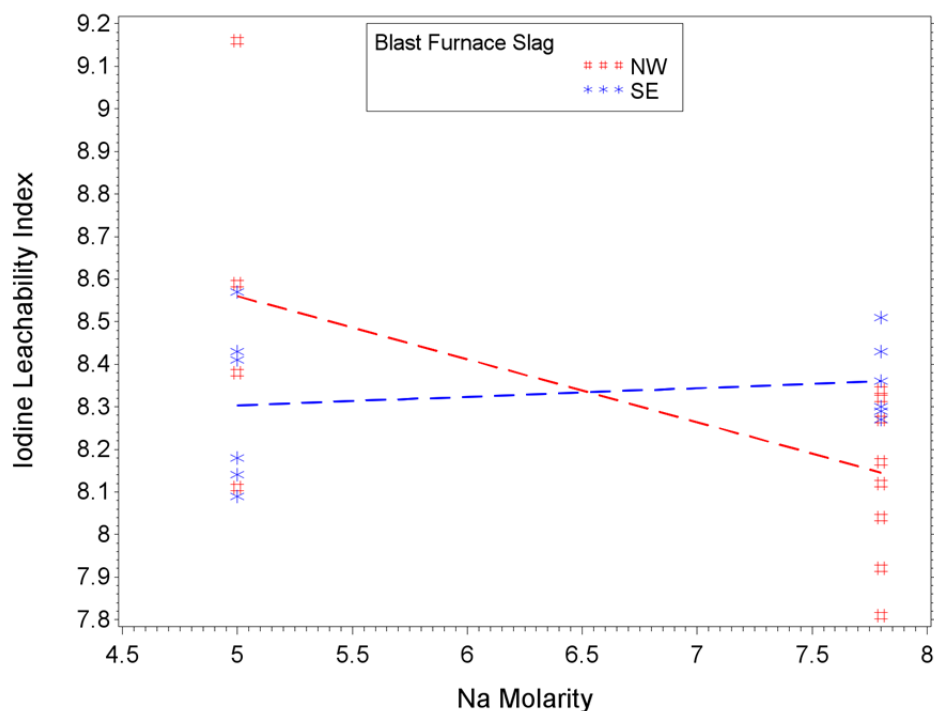


Figure E-68. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Iodine Leachability Index

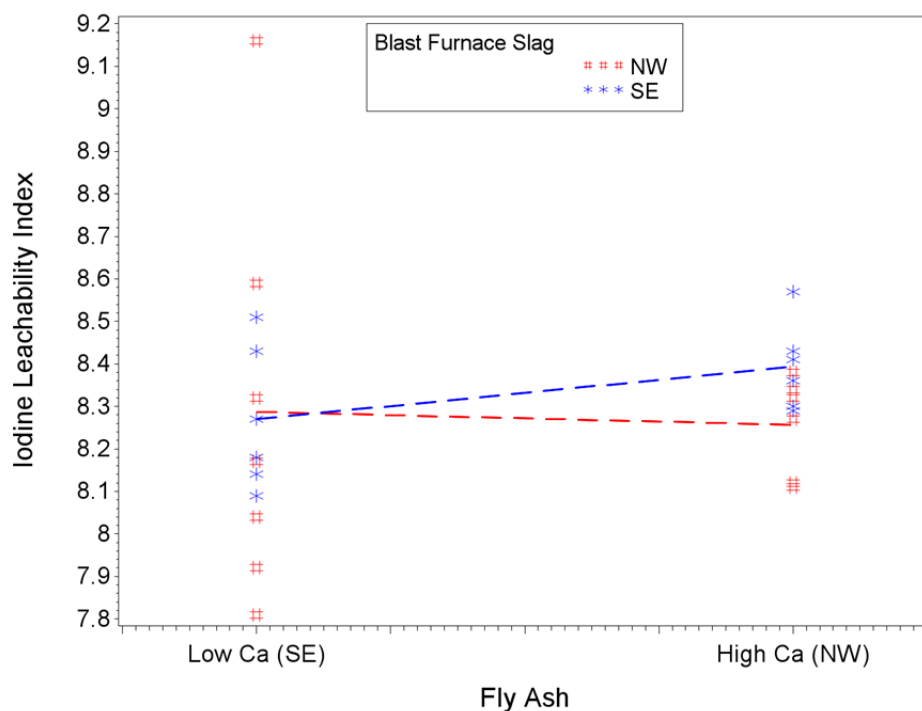


Figure E-69. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Iodine Leachability Index

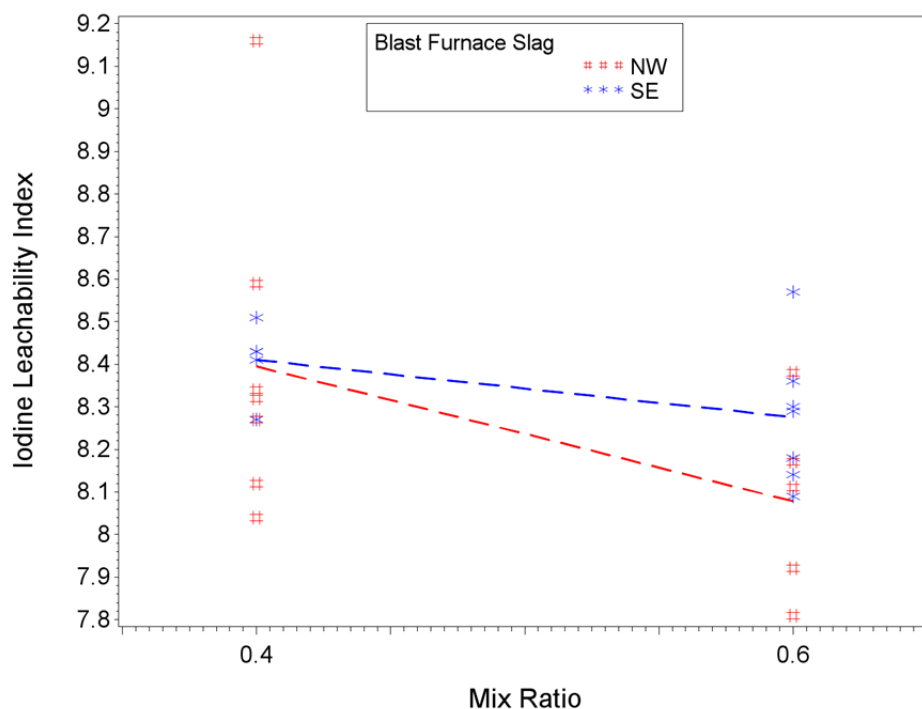


Figure E-70. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Iodine Leachability Index

E.4.4 *Technetium Leachability Index*

Figure E-71 through Figure E-80 display the data-interaction plots for the averaged Tc LI of the 26 Cast Stone mixes in the screening text matrix.

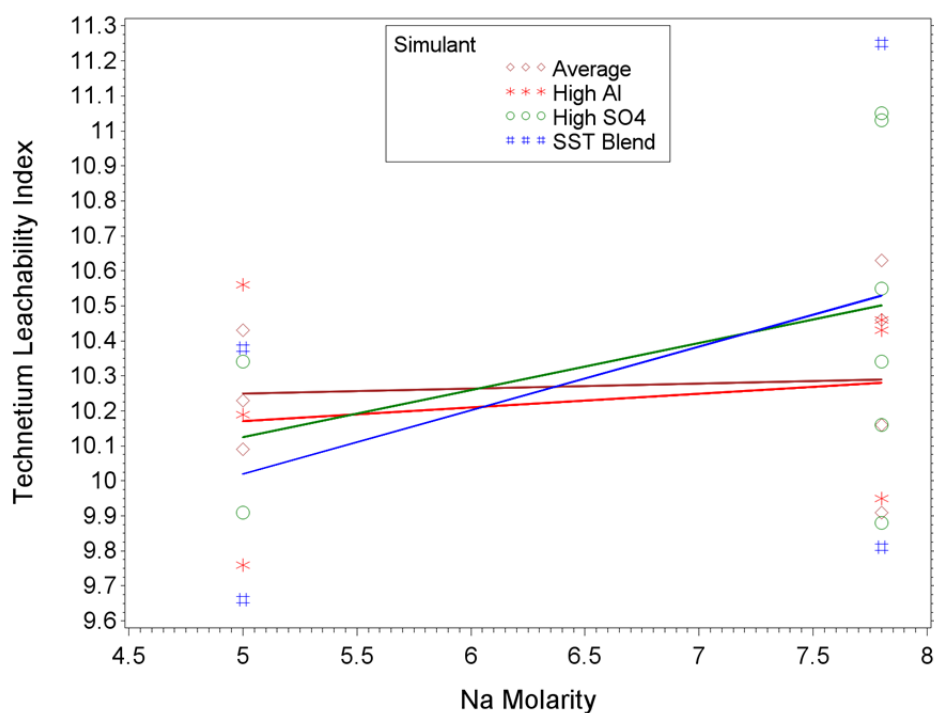


Figure E-71. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Technetium Leachability Index

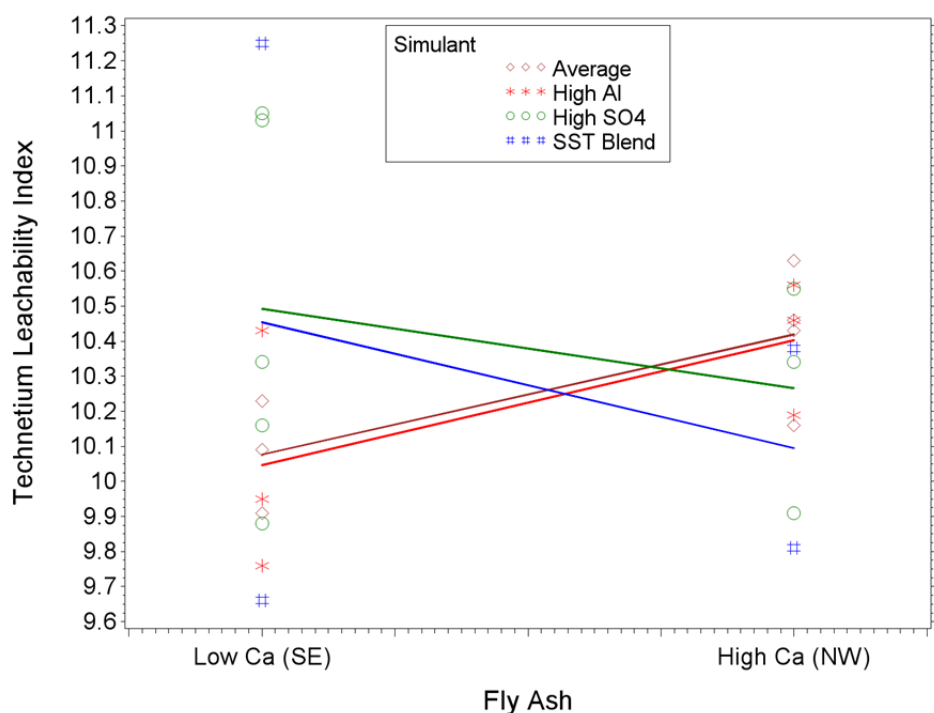


Figure E-72. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Technetium Leachability Index

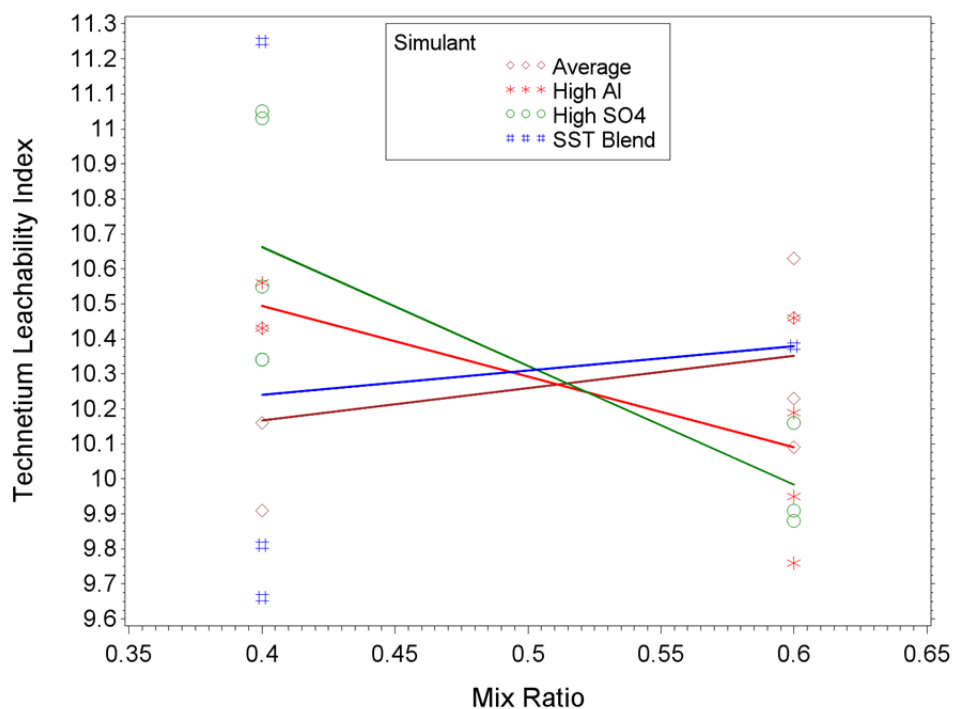


Figure E-73. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Technetium Leachability Index

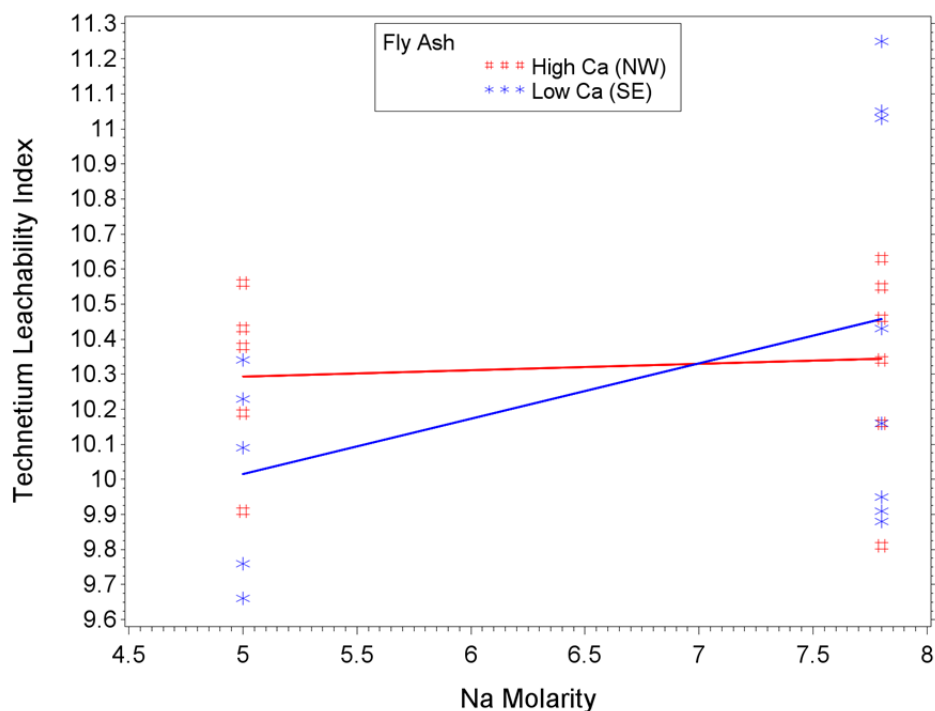


Figure E-74. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Technetium Leachability Index

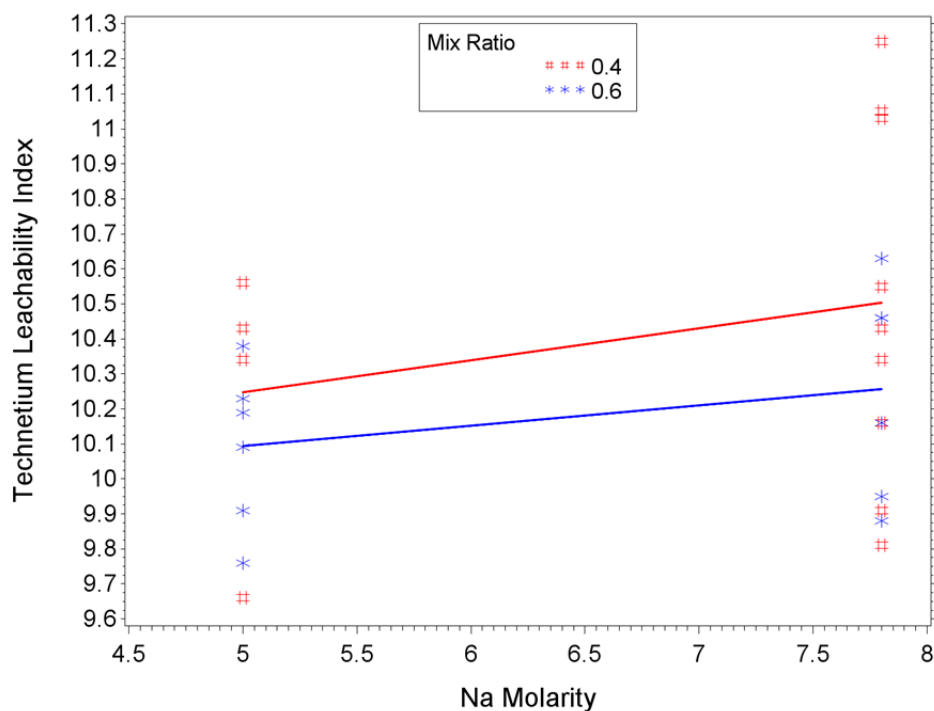


Figure E-75. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Technetium Leachability Index

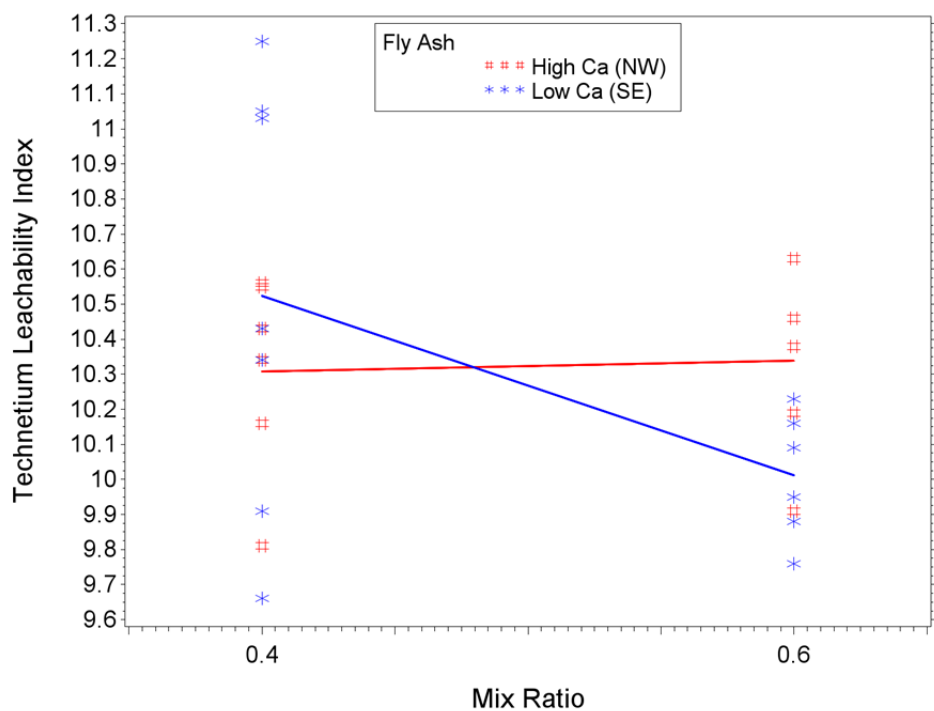


Figure E-76. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Technetium Leachability Index

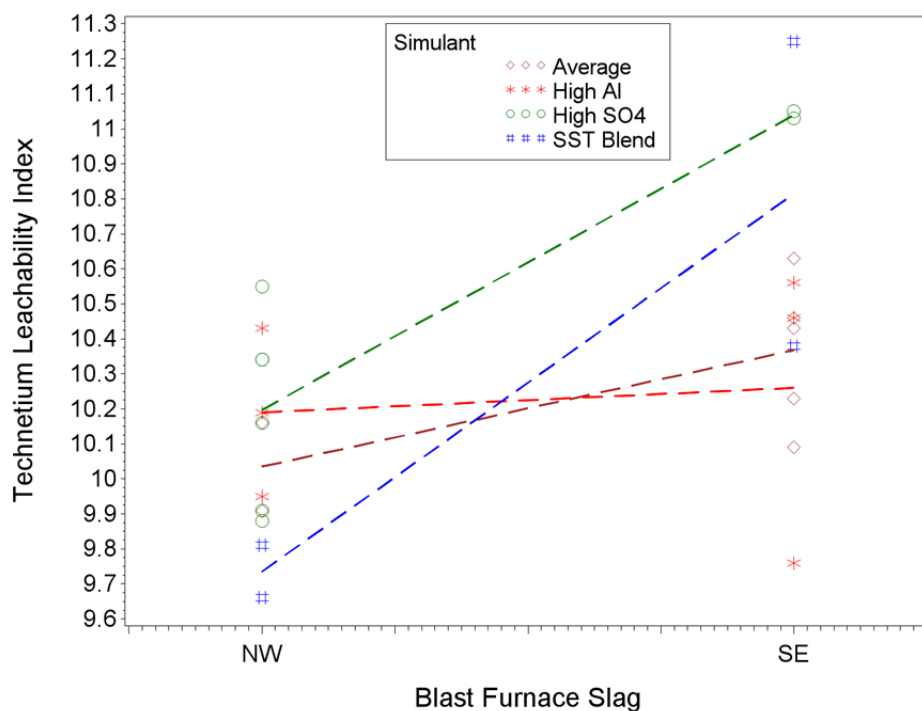


Figure E-77. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Technetium Leachability Index

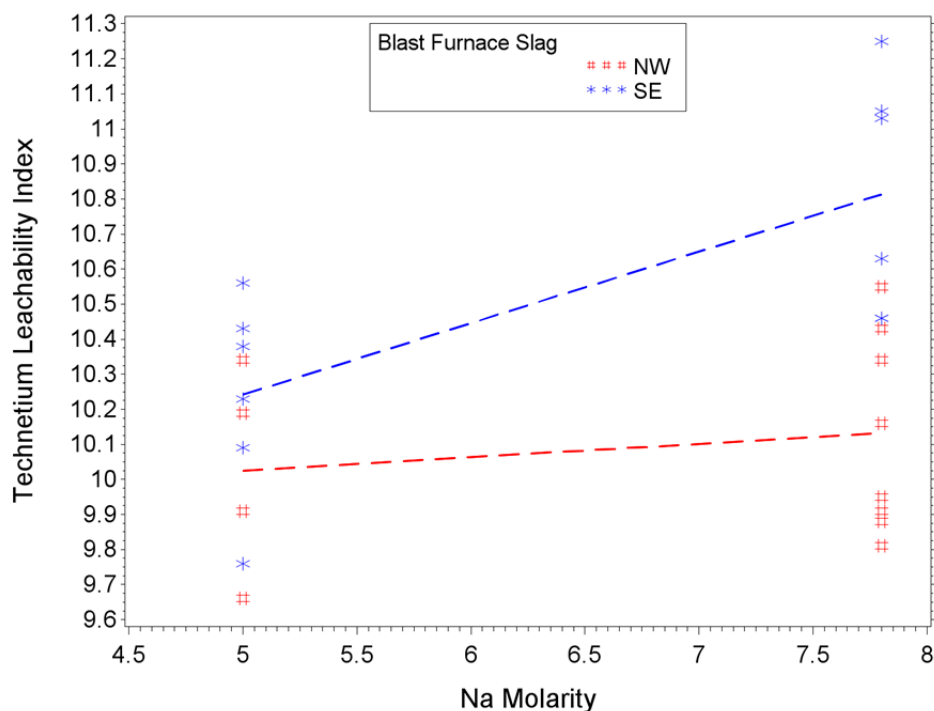


Figure E-78. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Technetium Leachability Index

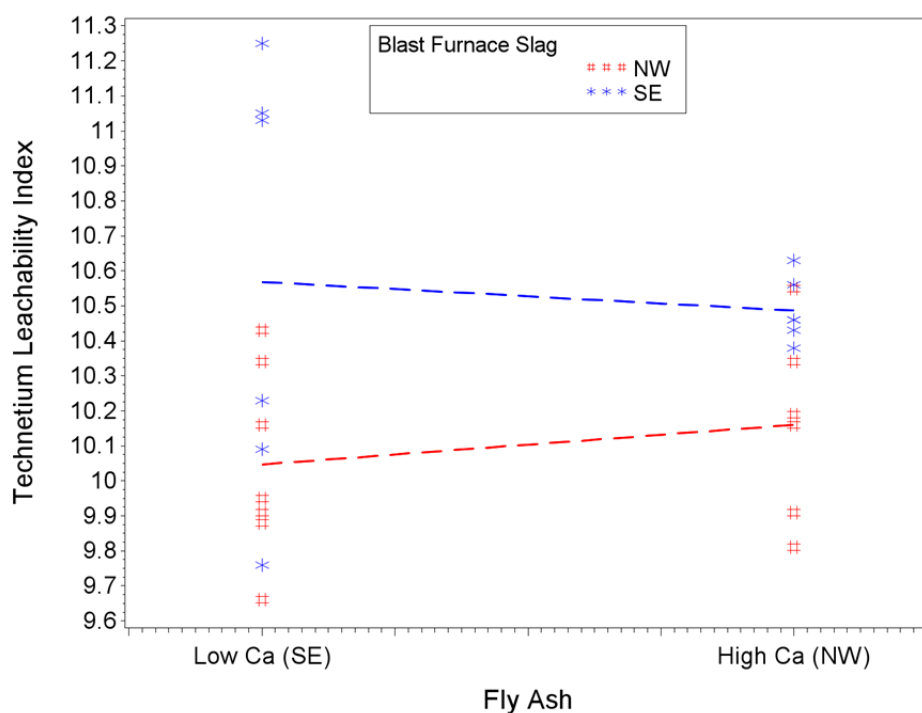


Figure E-79. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Technetium Leachability Index

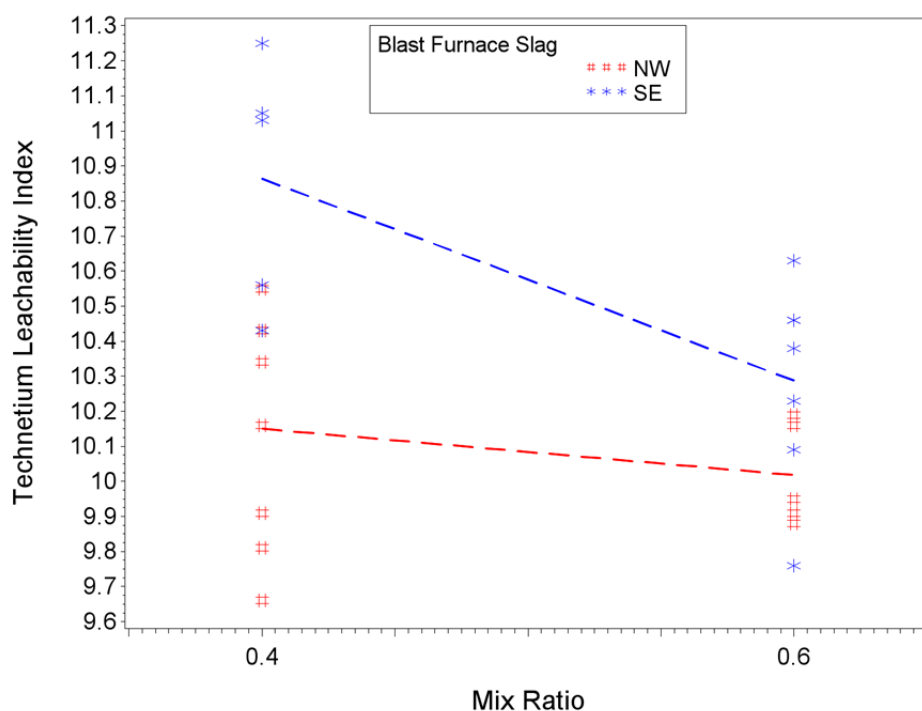


Figure E-80. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Technetium Leachability Index

E.4.5 *Chromium Leachability Index*

Figure E-81 through Figure E-90 display the data-interaction plots for the averaged Cr LI of the 26 Cast Stone mixes in the screening text matrix.

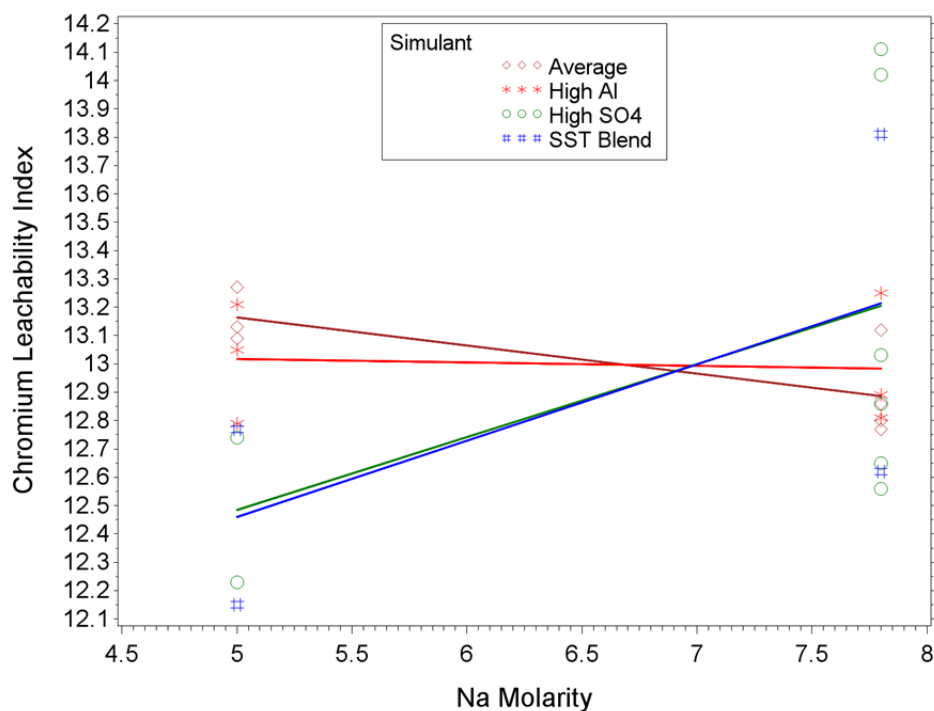


Figure E-81. Data-Interaction Plot for the Effects of Na Molarity and Simulant on Chromium Leachability Index

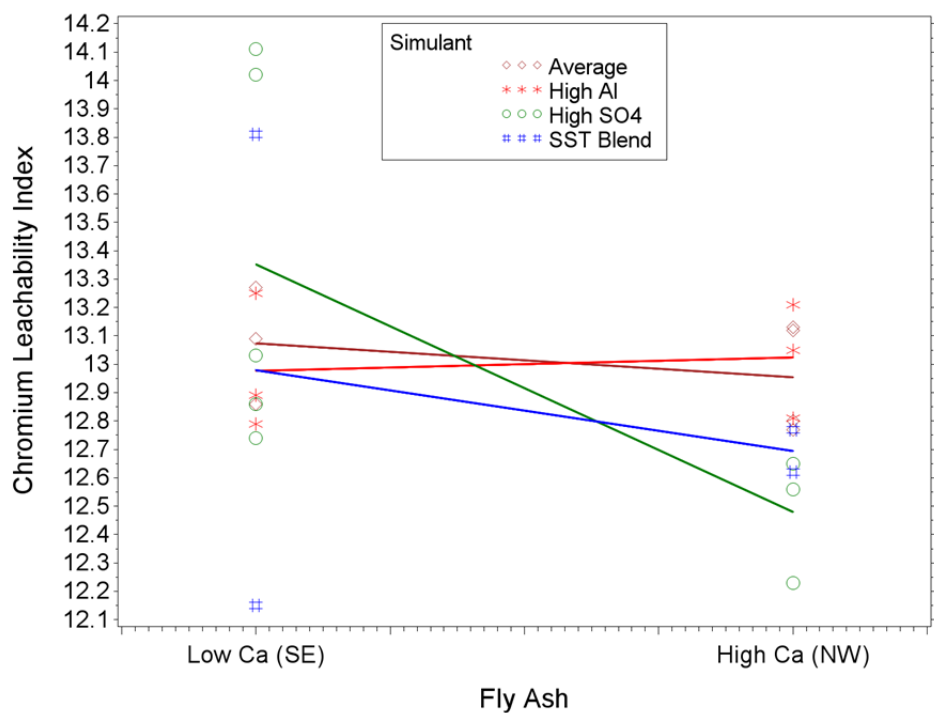


Figure E-82. Data-Interaction Plot for the Effects of Fly Ash and Simulant on Chromium Leachability Index

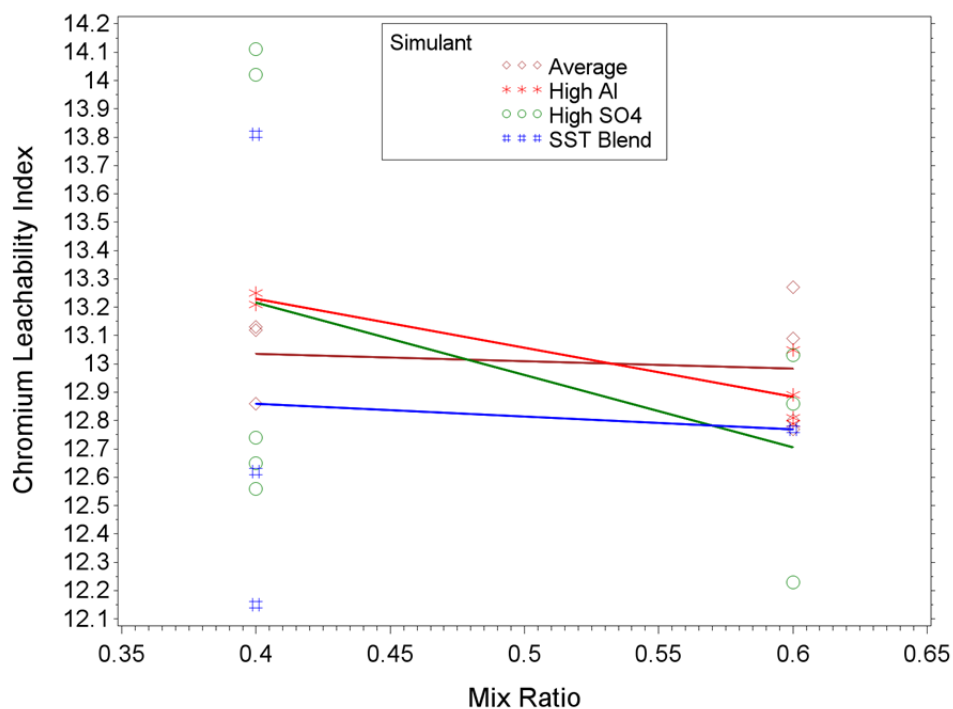


Figure E-83. Data-Interaction Plot for the Effects of Mix Ratio and Simulant on Chromium Leachability Index

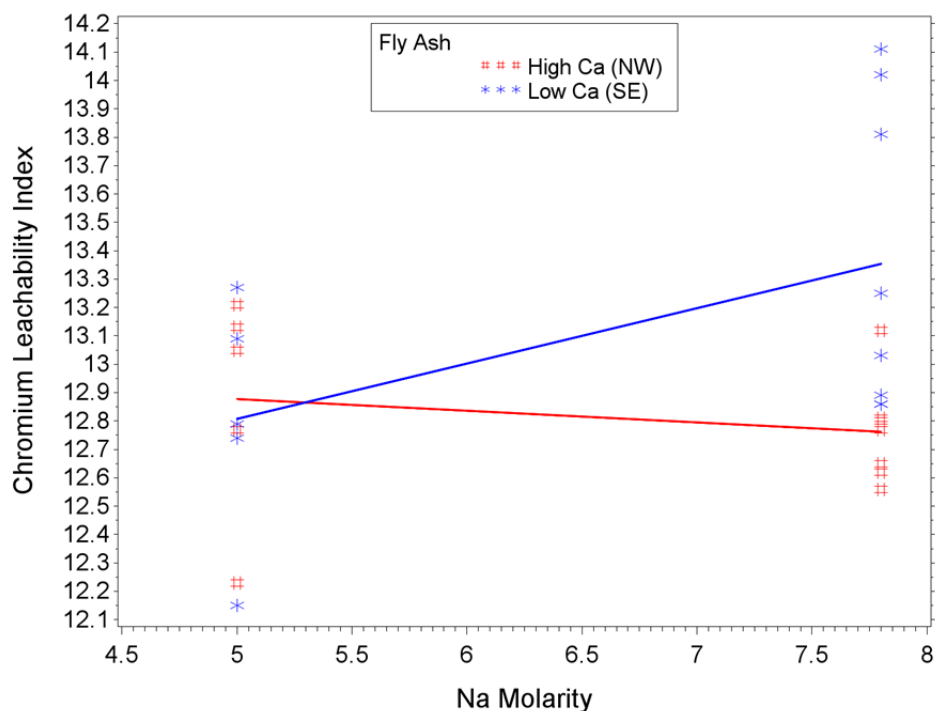


Figure E-84. Data-Interaction Plot for the Effects of Na Molarity and Fly Ash on Chromium Leachability Index

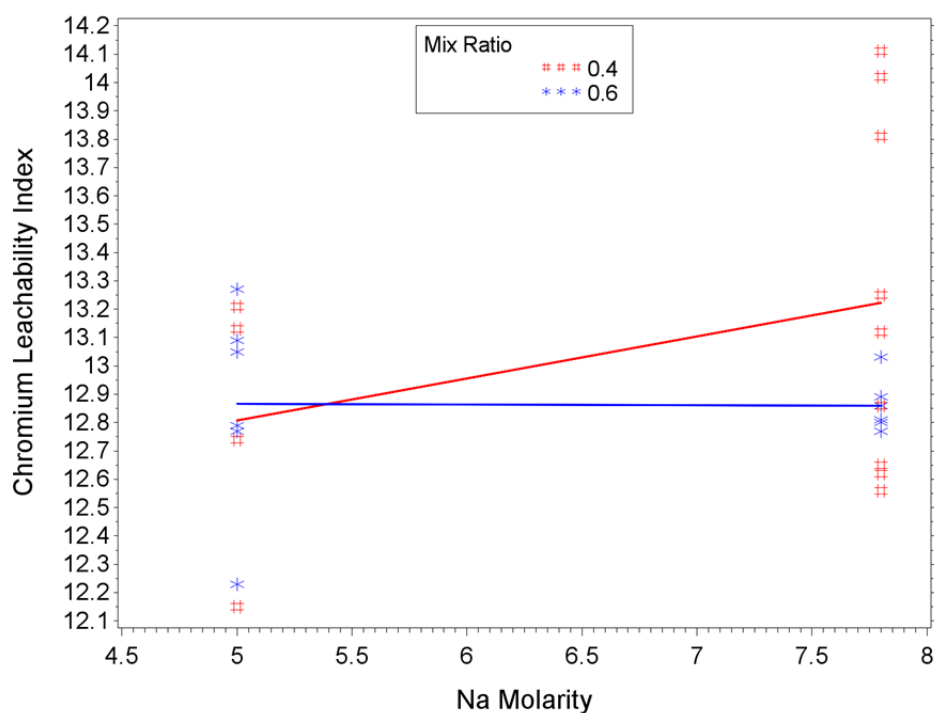


Figure E-85. Data-Interaction Plot for the Effects of Na Molarity and Mix Ratio on Chromium Leachability Index

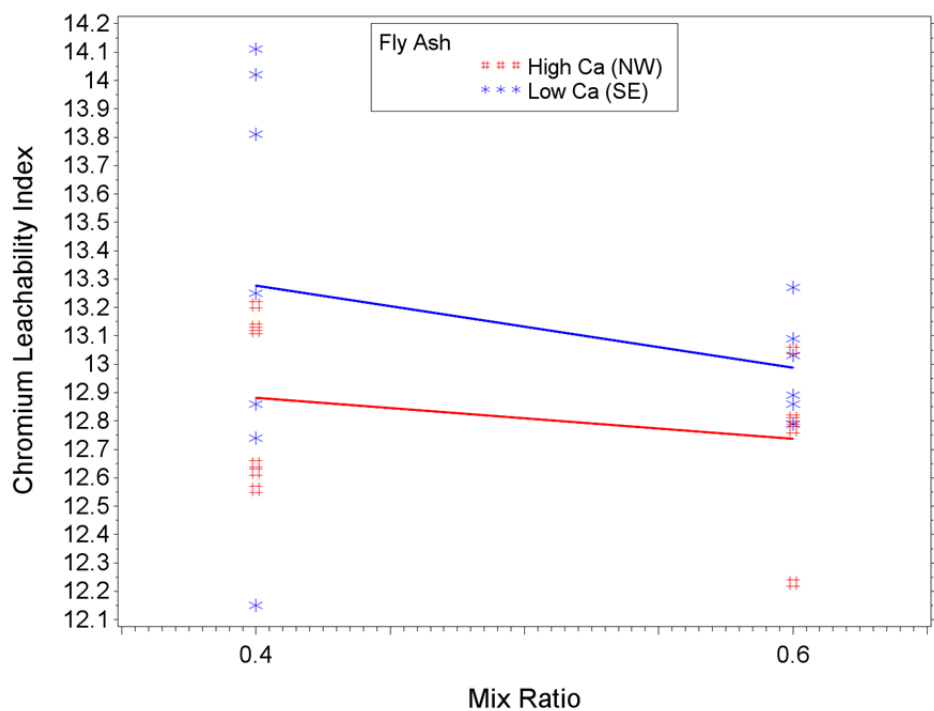


Figure E-86. Data-Interaction Plot for the Effects of Mix Ratio and Fly Ash on Chromium Leachability Index

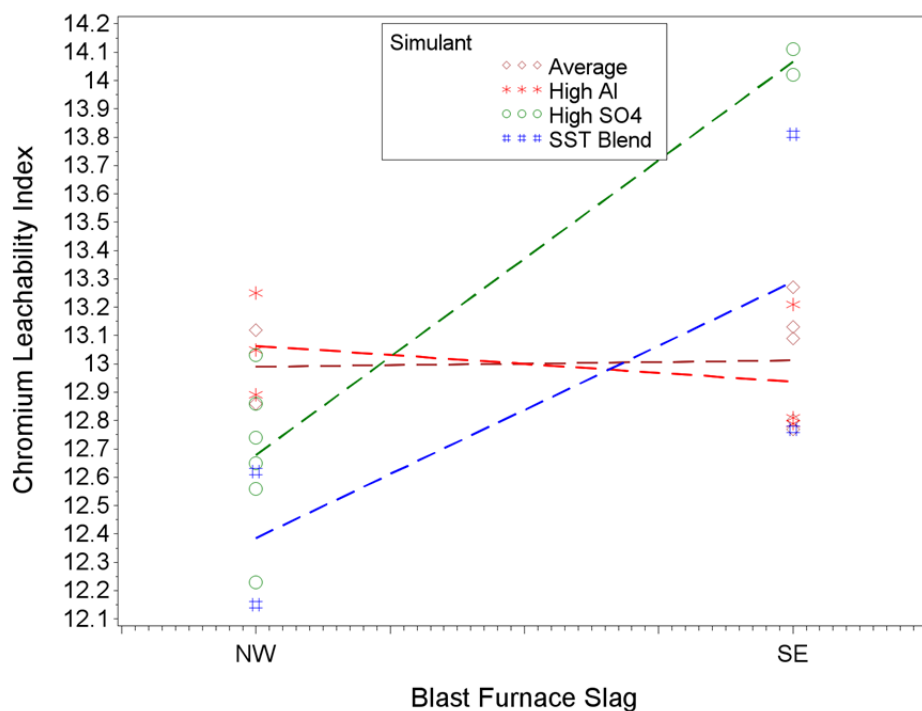


Figure E-87. Data-Interaction Plot for the Effects of Blast Furnace Slag and Simulant on Chromium Leachability Index

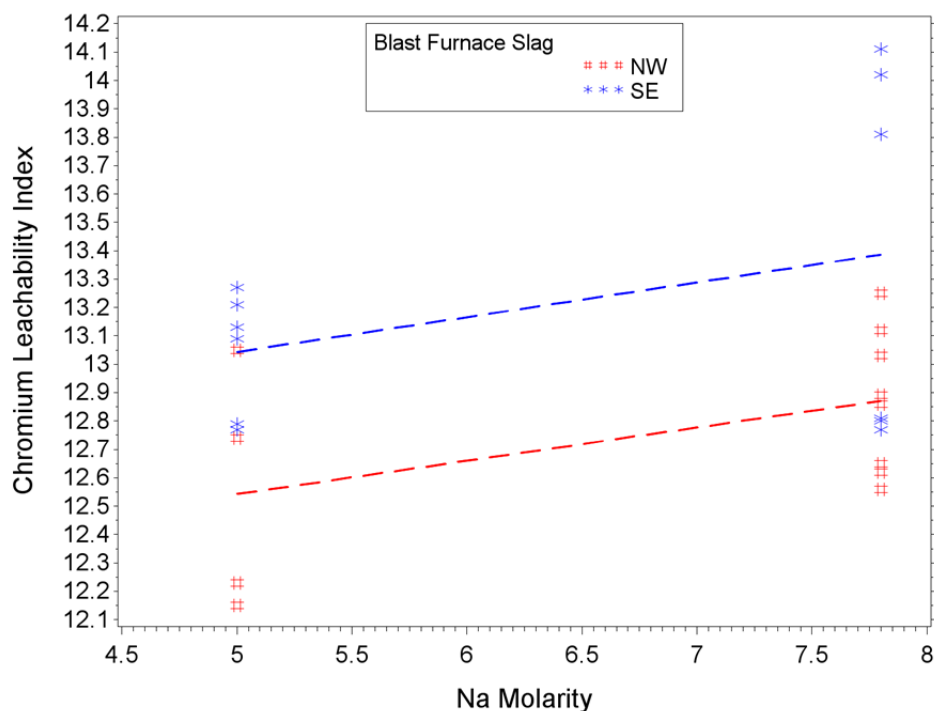


Figure E-88. Data-Interaction Plot for the Effects of Na Molarity and Blast Furnace Slag on Chromium Leachability Index

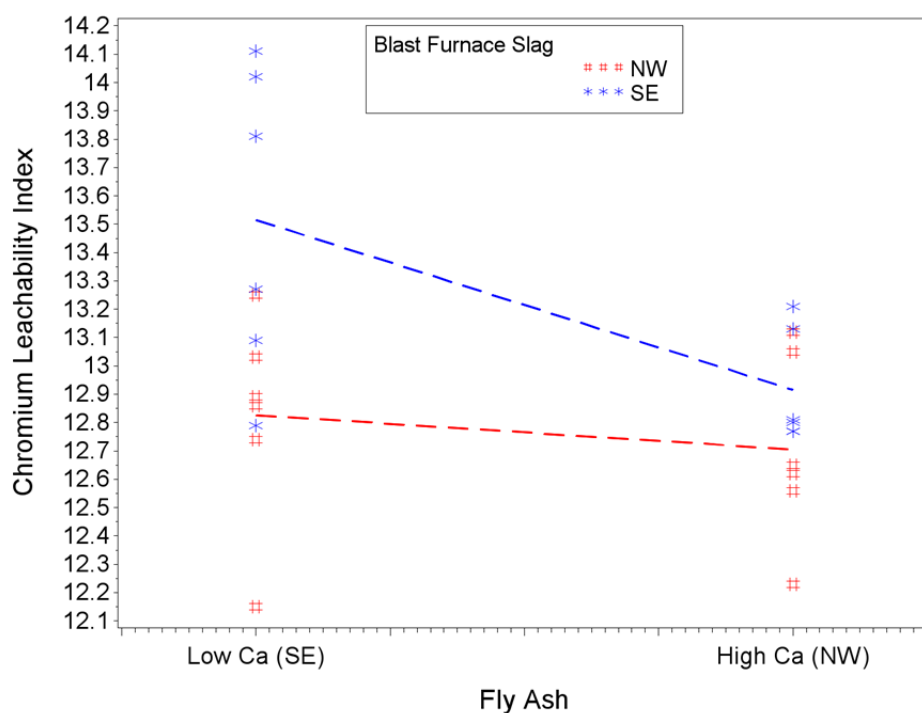


Figure E-89. Data-Interaction Plot for the Effects of Fly Ash and Blast Furnace Slag on Chromium Leachability Index

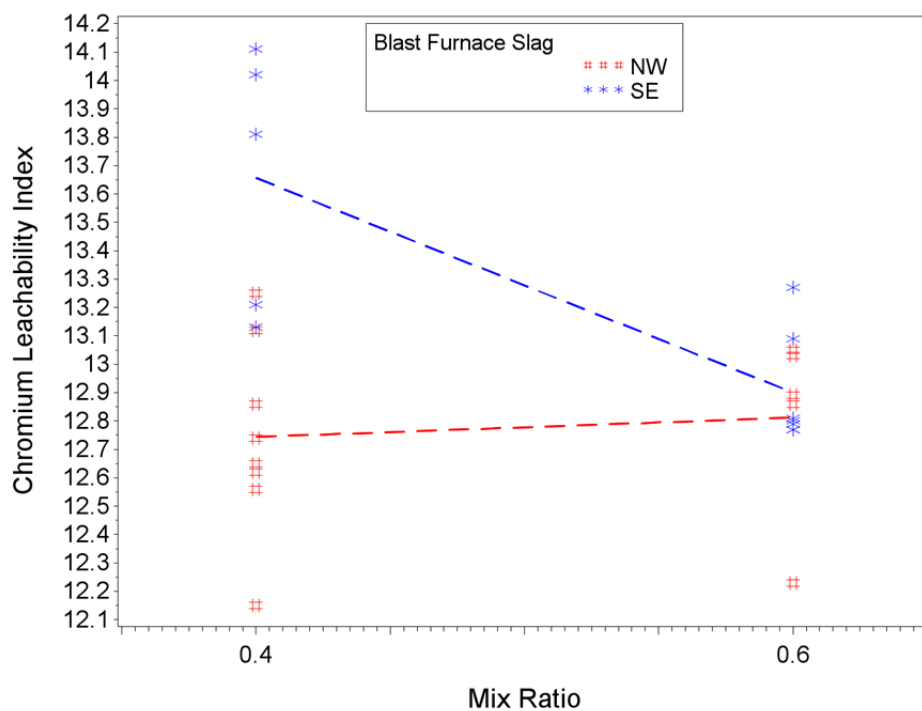



Figure E-90. Data-Interaction Plot for the Effects of Mix Ratio and Blast Furnace Slag on Chromium Leachability Index

Appendix F. Material Certification Reports

2013 10095
 Version 6.19



Material Certification Report

Brand: Envirocore™ Family of Products

Material: GranCem® Slag Cement

Grade: 100

Date Range: June 1-30, 2012

Lot Number: Multiple Lots

Certification
 This cement meets the requirements of ASTM specification C989 for Grade GranCem 100 GranCem®.

Supplier: Holcim (US) Inc.

Address: 1554 Hartman Industrial Blvd.
Birmingham, AL 35213

Telephone: Roberto Carrillo/773-768-1717 x 254

Date Issued: 01-Aug-2012

Source Location: Birmingham Slag Facility
1555 Hartman Industrial Blvd.
Birmingham, AL 35214

Contact: Lance Stephens/205-929-6810

The following information is based on average test data during the test period.
The data is typical of cement shipped by Holcim; individual shipments may vary.

Test Data on ASTM Standard Requirements

Chemical			Physical		
Item	Limit ^A	Result	Item	Limit ^A	Result
Sulfide S (%)	2.5 max	1.15	+45 µm (No. 325) Sieve (%)	20 max	0.43
Sulfate Ion - SO ₃ (%)	-	0.13	Blaine Fineness (m ² /kg)	-	602
			Air Content (%)	12 max	5.45
			Slag Activity Index (%)		
			Avg 7 Day Index	75 min	78
			Avg 28 Day Index	95 min	116
			Compressive Strength - MPa (psi):		
			Slag + Reference Cement		
			7 Day	-	23 (3360)
			28 Day	-	44 (6400)
			Reference Cement ^B		
			7 Day	-	30 (4320)
			28 Day	-	38 (5550)

Test Data on Reference Cement

Chemical			Physical		
Item	Limit ^A	Result	Item	Limit ^A	Result
Total Alkalies as Na ₂ O (%)	0.60 - 0.90	0.69	Compressive Strength - 7 day	-	29.6 (4300)
C ₃ S	-	61	Compressive Strength - 28 day	5000 psi	37 (5390)
C ₂ S	-	13			
C ₃ A	-	7	Blaine fineness (m ² /kg)	-	360
C ₄ AF	-	8.22			

Notes

^ADashes in the limits column means Not Applicable

^BReference cement results from procedure "Preparation of Specimens". Information on Reference Cement qualification available upon request.

Specific Gravity: 2.69

This data may have been reported on previous mill certificates. It is typical of the cement being currently shipped which was produced in June of 2012

BOL# 805992928

P.O.# SRR1002656-6

By Robert Carrillo, Quality Manager

2013 10095
7 9

QUALITY GRINDING AIDS, INC

Larry Hurley
626 West Alabama
Houston, Texas 77006

Phone: 713-942-7077
Fax: 713-942-7379
Mobile: 713-299-1049

Email: lhurley@qualitygrindingaids.com

Certificate of Analysis:

QGA-GL

Diethylene Glycol Grinding Aid

Physical Data:

Diethylene Glycol	+99%
Water	<1.0%
Appearance	Clear Liquid
Specific Gravity	1.10-1.12
pH	6.0- 9.0
Solubility in water	complete
Boiling Point	245C
Freezing Point	<-10C

BOL# 805992928

P.O.# SRR0002656-6

Larry Hurley

2013 8 9 10095

QUALITY GRINDING AIDS, INC

Larry Hurley
626 West Alabama
Houston, Texas 77006

Phone: 713-942-7077
Fax: 713-942-7379
Mobile: 713-299-1049

e-mail: lhurley@qualitygrindingaids.com

Certificate of Analysis:

QGA-AA

Amine-Acetate Grinding Aid (50% Amines reacted with 25% Glacial Acetic Acid)

Physical Data:

Amine-Acetate	75%
Water	25%
Appearance	Dark Liquid
Specific Gravity	1.14-1.16
pH	6.0-7.0
Solubility in water	complete
Boiling Point	116C
Freezing Point	<-20C

BOL# 805992921

P.O.# SRR0002656-6

Adrian Storey



2013 8-10-10099
Page 8 of 8

Client: Mr. Tom Hendrix
The SEFA Group
P.O. Box 6
Moncks Corner, SC 29461

Date: March 5, 2013
TEC Services I.D.: TEC 06-0509
Lab No.: 13-009-WA

REPORT OF FLY ASH TESTS				
Sample I.D. No.: WA122812		Date Sampled: December 28, 2012		
Manufacturer: Wateree Station (Thermally Beneficiated)		Date Received: January 7, 2013		
Chemical Analysis**		Results	Specification (Class F)	
			ASTM C618-08a	AASHTO M295-07
Silicon Dioxide		49.91	----	----
Aluminum Oxide		25.71	---	----
Iron Oxide		11.13	---	----
Sum of Silicon Dioxide, Iron Oxide & Aluminum Oxide		86.75	70 % min.	70 % min.
Calcium Oxide		4.15	----	----
Magnesium Oxide		1.14	----	----
Sulfur Trioxide		0.99	5 % max.	5 % max.
Loss on Ignition		1.87	6 % max.	5 % max.
Moisture Content		0.07	3 % max.	3 % max.
Total Chlorides		0.003	----	----
Available Alkalies as Na ₂ O		0.84	----	1.5 % max.
Physical Analysis		Test Date		
Fineness (Amount Retained on #325 Sieve)		01/19/13	14.0%	34 % max.
Strength Activity Index with Portland Cement				
At 7 Days:				
Control Average, psi: 4870	Test Average, psi: 4010	01/24/13	82%	75 % min. (of control)
At 28 Days:				
Control Average, psi: 6300	Test Average, psi: 5160	02/14/13	82%	75 % min. (of control)
Water Requirements (Test H ₂ O/Control H ₂ O)				
Control, mls: 242	Test, mls: 235	01/17/13	97%	105 % max. (of control)
Autoclave Expansion		01/16/13	-0.02%	± 0.8 % max.
Uniformity Requirements		Test Date	Variation	
Specific Gravity: 2.30	Average: 2.23	01/19/13	3.1%	5 % max. from average
% Retained #325 Sieve: 14.0	Average: 16.7	01/19/13	-2.7%	5 % max. from average

[†] Meeting the 7 day or 28 day strength activity index will indicate specification compliance

* Optional

**Chemical Analysis performed by Wyoming Analytical on January 25, 2013.

The results of our testing indicate that this sample complies with ASTM C618-08a and AASHTO M295-07 specifications for Class F pozzolans.

Respectfully Submitted,
Testing, Engineering & Consulting Services, Inc.

Anne Miller
Project Manager

P.O. #SRR0002657-6

T2886
4/4/13

Shawn P. McCormick
Laboratory Principal



Cement

FLY ASH TEST REPORT

Analysis by: Lafarge Seattle Concrete Lab
Sample from : Centralia Power Plant
Average Analysis: August 1st - August 31st 2012
Test Report Number 9-12 F

Chemical Analysis

	Results	Limits
Silicon Dioxide (SiO ₂)	54.0 %	
Aluminum Oxide (Al ₂ O ₃)	16.5 %	
Iron Oxide (Fe ₂ O ₃)	6.5 %	
Total (SiO ₂) + (Al ₂ O ₃) + (Fe ₂ O ₃)	77.0 %	70% Min - ASTM
Sulphur Trioxide (SO ₃)	0.8 %	5% Max - ASTM
Calcium Oxide (CaO)	12.2 %	
Magnesium Oxide	4.1 %	
Moisture Content	0.21 %	3% Max - ASTM
Loss on Ignition	0.19 %	6% Max - ASTM
Available Alkali as Equiv. Na ₂ O (<i>previous month's result</i>)	1.2 %	

Physical Analysis

Fineness Retained on 45 um (No. 325 Sieve)	17.3 %	34% Max - ASTM
Strength Activity Index with Portland Cement		
% of Control at 7 Days	80 %	75% Min - ASTM
% of Control at 28 Days (<i>previous month's result</i>)	96 %	75% Min - ASTM
Water Requirement, Percent of Control	98 %	105% Max- ASTM
Autoclave Expansion	0.02 %	0.8% Max - ASTM
Density	2.59 Mg/m ³	

Uniformity Requirements

Density, Variation from Average	0.00 %	5% Max - ASTM
Fineness 45um Sieve, Variation from Average	0.00 %	5% Max - ASTM

We hereby certify that the composite fly ash sample above meets the chemical and physical requirements of ASTM C618-08 and AASHTO M295-07 for class F and C fly ash.

Certified : 

WESTERN REGION

5400 West Marginal Way SW, Seattle, Washington 98106-1517
Office: 206.923.0098 or 800.477.0100 Fax: 206.923.0388



Mill Test Report Number: SEA_NEWCEM_AUG12	
YEAR:	2012
MONTH:	JULY
PLANT:	Seattle
CEMENT TYPE: Grade 100 NewCem	

Reference Cement		Slag			
Fineness by Air Permeability (m ² /kg; ASTM C204)	388	Fineness by Air Permeability (m ² /kg; ASTM C204)	586		
Fineness by 45 µm (No. 325) Sieve (% retain; ASTM C430)	4.5	Fineness by 45 µm (No. 325) Sieve (% retain; ASTM C430)	3.2		
Compressive Strength (ASTM C109/C109 M)	psi	Compressive Strength (ASTM C109/C109 M)	psi	SAI	Limit
7-day	4,330	7-day	4,875	89	75
28-day	6,250	28-day (Previous Month)	5,684	110	95
Total Alkalies (Na ₂ O + 0.658 K ₂ O) (%, ASTM C114)	<u>Actual</u> 0.85 <u>Max Limit</u> 0.9	Specific Gravity (Mg/m ³ ; ASTM C188)	2.87	<u>Actual</u>	<u>Max Limit</u>
		Air Content of Mortar (%, ASTM C185)	4.6	4.6	12
		Sulfide Sulfur (% S, ASTM C114)	0.7	0.7	2.5
		Sulfate Ion (% as SO ₃ , ASTM C114)	3.2	3.2	4
Slag					
CHEMICAL ANALYSIS		Percent			
Silica Dioxide (SiO ₂ ; ASTM C114)	30.5				
Ferric Oxide (Fe ₂ O ₃ ; ASTM C114)	0.8				
Aluminum Oxide (Al ₂ O ₃ ; ASTM C114)	11.9				
Calcium Oxide (CaO; ASTM C114)	43.8				
Sulfur Trioxide (SO ₃ ; ASTM C114)	4.9				
Magnesium Oxide (MgO; ASTM C114)	4.2				
Potassium Oxide (K ₂ O; ASTM C114)	0.7				
Titanium Oxide (TiO ₂ ; ASTM C114)	0.5				
Loss on Ignition (L.O.I.; ASTM C114)	2.0				
Inorganic Process Addition	9				

The ground granulated blast furnace slag complies with the current specification of the chemical physical requirement of ASTM C-989, AASHTO M-302 for grade 100 Ground Granulated Blast Furnace Slag (GGBFS) and and CSA A3001 Slag.

Certified by:

Daniel Waldron

Daniel Waldron
Quality Control Laboratory Supervisor
August 21, 2012