Engineering Scale Demonstration of a Prospective Cast Stone Process – 15584

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ABSTRACT

A cementitious waste form is one of the alternatives being evaluated for Supplemental Immobilization of Hanford Low Activity Waste (LAW). Washington River Protection Solutions (WRPS) is collecting data on cementitious or pozzolanic waste forms such as Cast Stone. The goal of this project for WRPS is to obtain data on the performance of the Cast Stone waste form for immobilizing LAW.

As part of the data package being assembled, an engineering-scale demonstration with non-radioactive simulants was performed at the Savannah River National Laboratory (SRNL) using the Scaled Continuous Processing Facility (SCPF) to fill a container with simulated Cast Stone grout. Legacy salt solution from previous Hanford salt waste testing was trimmed to resemble the average composition generated from the Hanford Tank Waste Operation Simulator (HTWOS) used in the screening tests. The dry blend materials, ordinary portland cement (OPC), Class F fly ash, and ground granulated blast furnace slag (GGBFS or BFS) were obtained from Lafarge North America in Pasco, WA. Over three days, the SCPF was used to fill a 6,056 liter (1600 gallon) container with simulated Cast Stone grout. The tank was instrumented with x-, y-, and z-axis thermocouples to monitor curing temperature and two formed core sampling vials. The target production rate was 5.7 L/min (1.5 gpm). This required a salt solution flow rate of approximately 3.78 L/min (1 gpm) and a premix feed of approximately 263 kh/h (580 lb/h). The final surface slope at a fill height of 100 cm was 2.5-3.8 cm across the 260 cm (8.5 ft) diameter tank. During processing, grout was collected from both the mixer discharge and the discharge into the tank. These samples were stored in a humid environment either in a closed box proximal to the tank or inside the laboratory.

INTRODUCTION

Washington River Protection Solutions (WRPS) is generating and collecting data on cementitious or pozzolanic waste forms such as Cast Stone. A cementitious waste form is one of the alternatives being evaluated for Supplemental Immobilization of Hanford Low Activity Waste (LAW), along with vitrification, bulk vitrification, and fluidized bed steam reforming.

The goal of this project for WRPS was to obtain data on the performance of the Cast Stone waste form for immobilizing LAW. In support of this goal, a testing program was developed to obtain additional information on the Cast Stone option for immobilizing the LAW.[1] Screening tests to examine expected ranges in waste composition, waste concentration, dry materials sources, and free water (in the waste liquid)-to-dry blend mix ratios have been performed.[2]

As part of the data package, an engineering-scale demonstration with non-radioactive simulants was performed. The Scaled Continuous Processing Facility (SCPF) at SRNL was used to fill a container with simulated Cast Stone grout to display the ability to operate a process to immobilize a simulated LAW salt solution in a cementitious waste form.

METHODS

A Composition from the screening tests in Reference 2 was chosen to represent a possible Cast Stone formulation for the engineering scale demonstration (ES Demo). The dry blend binders for the were obtained from Lafarge North America in Pasco, Washington, a source that would be readily available at the Hanford Site. The salt solution waste simulant was prepared using salt simulants reclaimed from the fractional crystallization pilot scale test performed at SRNL.[3] The Cast Stone grout was prepared using the SCPF, an integrated grout preparation system at SRNL capable of producing up to 15 liters of grout per minute.

Dry Blend Binders
Ordinary portland cement (OPC), ground granulated blast furnace slag (BFS), and Class F fly ash were characterized using compositional analysis, X-ray diffraction analysis, particle size distribution, and Brunauer–Emmett–Teller (BET) surface area measurements. The dry materials were weighed and blended in 7 kg batches to facilitate loading the dry feeder. Table I is the baseline Cast Stone ratios used for the binder materials.

TABLE I. Dry blend mixture of binders used to prepare 7 kg batches.

Dry Blend Component	Target (wt %)	Targeted Mass (g)	Acceptable Mass (g)
OPC	8	560	555-565
BFS	47	3290	3270-3310
fly ash	45	3150	3130-3170

Salt Solution Waste Simulant
Select totes of legacy salt solution were analyzed and blended in a single tank. The blended salt solution was trimmed to approximate the targeted composition of the 7.8 M Na Overall Average from the bench scale testing.[4] Table II shows the composition of the main components of the targeted salt solution and the composition of the blended totes after trim chemicals were added. The components that were targeted for approximation were aluminum, sodium, and nitrate + nitrite. The differences between the measured values for each of the analytes were within the analytical error associated with each of the components.

TABLE II. Targeted and measured salt solution compositions.

Salt Solution Component	Concentration (mg/L)	
	Targeted Overall Average	Blended/adjusted Tank
Al	12.9	11.4
Cr	1.74	1.58
K	2.00	6.36
Na	179	169
Na (Mol/L)	7.8	7.3
Cl	2.33	4.61
PO4	7.29	2.18
SO4	12.8	9.41
NO2	40.5	33.4
NO3	157	193

Density	1.346	1.362
Wt% solids	38.4	40.5
Wt% water	61.6	59.5

Waste Form Container

The receptacle for the Cast Stone waste form is a 6,056 liter (1600 gallon), 260 cm (8.5 ft) diameter, polyethylene container. The container was staged approximately 18 m from the SCPF. The container was fitted with three grout entry points, an array of thermocouples spanning the diameter and expected fill height of the interior, and two refurbished emplaced core vials recovered from previous testing.[5] Temperature data were recorded for 14 days after processing.

Scaled Continuous Processing Facility

The SCPF uses a gravimetric feeder to supply the dry materials to a Readco-Kurimoto 2-inch continuous processor. The mixer is fitted with co-rotating, twin shafts, configured with intermeshing paddles. Clearance between intermeshing paddles and between the paddles and the barrel of approximately one-eighth inch produces a self-wiping action that minimized material buildup. A variable speed gear pump delivers the liquid feed (water or salt solution) to the grout mixer. Fresh grout is discharged from the mixer into the agitated grout pump hopper. A peristaltic pump transfers grout from the grout hopper through tubing to the grout receipt container. Figure 1 is the SCPF components configured for operation.
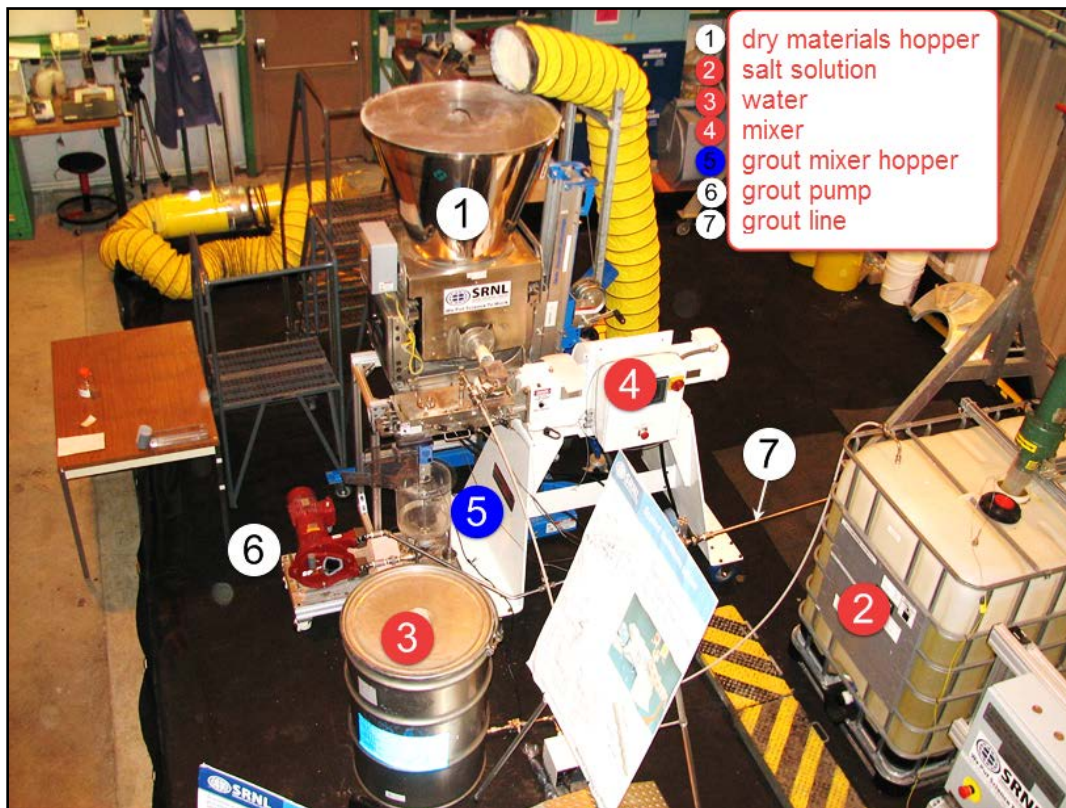

Fig.1. Labeled components of the SCPF configured for operation.

Sampling
During processing, fresh grout was sampled at two locations: at the discharge of the grout mixer and at the grout tank. A sampling chute above the grout hopper allowed sampling of fresh grout from the discharge of the mixer and a diverter valve at the container enabled sampling without discontinuing flow to the container. The density of freshly prepared grout was measured with weight per gallon sample cups using a simplified ASTM method.   A flow curve was processed for samples from both locations using a Haake VT550 rotoviscometer. 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, providing yield stress and plastic viscosity values. Additional sample material was collected in 5 x 10 cm (2 in x 4 in) cylinders, placed in zip top bags with moistened towels, and stored either in a lidded box near the grout container, or indoors behind the mixer.

DISCUSSION

Processing
The initial operating parameter for the dry materials was 270 kg/h. The salt solution feed was set at 3.4 L/m to maintain a water to dry materials ratio near 0.60. The W/DM of 0.60 was labeled as the controlling parameter and is based on the mass of the water in the salt solution and the mass of the dry blend materials, Eq. 1. The dry feed rate was selected to meet the processing rate to fill the grout container over the allotted time frame, with the assumption that there was minimal stoppage time. It is a practical goal of processing to maintain a constant dry feed rate unless that has been identified as a variable parameter. The salt solution feed rate is adjusted based on the water content of the salt solution and the dry feed rate to obtain the targeted W/DM. However, the salt solution flow meter was not able to control the flow rate precisely enough to rely on the salt solution feed values. Although the measured values are reported with two significant figures for calculation purposes, the practical flow rate is 4 L/m.

$$\frac{W}{DM} = \frac{\text{mass of water in salt solution}}{\text{mass of dry materials blend}} \qquad \text{(Eq. 1)}$$

Where the mass of the water in the salt solution is:

$$\frac{1.362 \text{ g salt solution}}{1 \text{ ml salt solution}} \times \frac{0.595 \text{ g water}}{1 \text{ g salt solution}} = \frac{0.810 \text{ g water}}{1 \text{ g salt solution}} \qquad \text{(Eq. 2)}$$

Where the g water/g salt solution is from Table II and,

$$\frac{W}{DM} = \frac{\dfrac{0.810 \text{ g water}}{1 \text{ g salt solution}} \times \dfrac{3.39 \text{ L salt solution}}{1 \text{ min}}}{\dfrac{270 \text{ kg dry materials blend}}{1 \text{ h}} \times \dfrac{1 \text{ hr}}{60 \text{ min}}} = 0.61 \qquad \text{(Eq. 3)}$$

These conditions resulted in a calculated grout density of 1.72 g/ cm^3, determined from the mass flow meter in the grout line, and a grout processing rate of 5.15 L/m into the container as calculated by the data acquisition system. Small adjustments were made to the salt solution flow to maintain a constant W/DM. The SCPF produced grout for approximately 3.9 h, resulting in ~28.5 cm of grout in the container. Processing was halted to preclude the onset of filling the lower emplaced core vial so the vial could be filled in a single lift. In previous testing, vials were filled under various processing conditions. For this testing, the

goal of filling the emplaced core vials was to produce a typical sample. Fig. 2 shows the grout surface at the conclusion of the first lift. The self-leveling ability of the grout slurry was evident at a fill rate of ~10 min/cm. Although not necessary, a self-leveling slurry improves operational flexibility by increasing the surface area serviced by a pour point.

The second lift was performed the following day using similar operating parameters as the initial lift: 270 kg/h and 3.5L/m, for a targeted W/DM of 0.63. The uncertainty of the salt solution flow meter lead to variability in the W/DM calculation. This in turn decreased the precision in the control of the salt solution feed rate and the resulting W/DM. These conditions resulted in a calculated grout density of 1.69 g/cm^3 and a grout processing rate of 5.3 L/m into the container. Small adjustments were made to the salt solution flow to maintain a stable W/DM. The SCPF produced grout for approximately 6.1 h resulting in ~36 cm of grout added to the container. The average salt solution flow rate for the second day of processing was 3.4 L/m and the average grout processing rate was 5.2 L/m. During processing, a film of excess salt solution was observed on the surface of the pour. The salt solution flow rate was subsequently reduced to 3.4 L/m – the flow rate used during the first lift. All of the excess salt solution had been incorporated into the grout slurry prior to the completion of the lift.

Fig. 2. Surface of the grout at the conclusion of the first lift. Note the opining for the emplaced core sampler.

The third and final lift was performed using similar operating parameters as the initial lift: 270 kg/h and 3.4 L/m, for a targeted W/DM of 0.61. The SCPF produced grout for approximately 2 h. To evaluate the ability to produce grout at a faster rate, the dry materials feed rate was increased to 276 kg/h. Issues with the ability to precisely measure the salt solution flow rate resulted in the salt solution feed rate reduced to 3.3 L/m. The W/DM shifted to 0.57, the calculated grout density still averaged 1.69 g/cm^3 and the grout processing rate was 5.2 L/m into the container. The SCPF was operated using these parameters for ~1.6 h. The SCPF processed the increased dry feed rate without issue and the decision was made to further increase the dry feed rate. The dry materials feed rate was increased to 283 kg/h with the salt solution flow rate increased to 3.4 L/m. The resulting W/DM was 0.58, the calculated grout density averaged 1.69 g/ cm^3 and a grout

processing rate of 5.2 L/m into the container. The SCPF was operated using these parameters for ~2.4 h. The third lift raised the final height of the container level to 100 cm. Figure 3 is the final grout surface after the third lift. The maximum difference in height across the 260 cm (8.5 ft) container was 2.5-3.8 cm.

Sampling
During processing, grout slurry samples were periodically collected from the mixer exit and at the container. Samples were evaluated for density and rheological properties. Additional samples collected in5 x 10 cm (2 in x 4 in) cylinders were stored for future analysis. It was observed during handling of the collected samples that grout slurries obtained directly from the mixer exit contained soft agglomerates of dry materials that had not yet been fully incorporated into the slurry. The inhomogeneity of the samples obtained from the mixer may have exacerbated the variability in density measurements for samples collected at the mixer exit. Conversely, after a residence time of approximately 60 s in the grout mixer hopper and transfer through 18 m of tubing, samples collected from the container appeared homogeneous. The observed transition from inhomogeneous to homogeneous grout slurry was also noted in previous testing.[6]

The plastic viscosity ranged from 0.08 – 0.13 Pa·s and Bingham plastic yield stress ranged from 1.5 – 7.3 Pa. These values were calculated from the down curve of the rheology flow curve. Specific rheology testing in Reference 11 notes the thixotropy associated with grout slurry samples collected at the mixer and shortly after the mixer. Only the down curve was considered for analysis to provide a more direct comparison and eliminate the heterogeneity associated with the less sheared samples.

Waste Form Container
After completion of the processing, the waste form container was monitored for temperature for 14 days. Thermocouple trees with three thermocouples (at bottom of container, at ~51 cm from the bottom, and at ~99 cm from the bottom—2.5 cm below the expected surface) were installed in the center and at equidistant radial positions. The maximum temperature, 69 °C, was reached at the mid-height thermocouples in the installations located 30 cm from the center. This temperature was reached 119 h after pouring was completed and the maximum temperature was maintained for ~10 h before dropping below 69 °C. Fig. 3 is a representation of the potentially different temperature zones present in the grout container as a result of the temperature profiles and the operating strategy of the SCPF. Since the temperature profile in the cylindrical grout container was symmetrical, a radial figure was used for clarity. The cold joints represent the top surface of the previous day of operation that is exposed to the atmosphere. The size of the smaller zones were artificially increased to facility the identity of cores for testing. Coring of the container occurred in April 2014 and the results of the coring and analysis will be presented in a separate report.
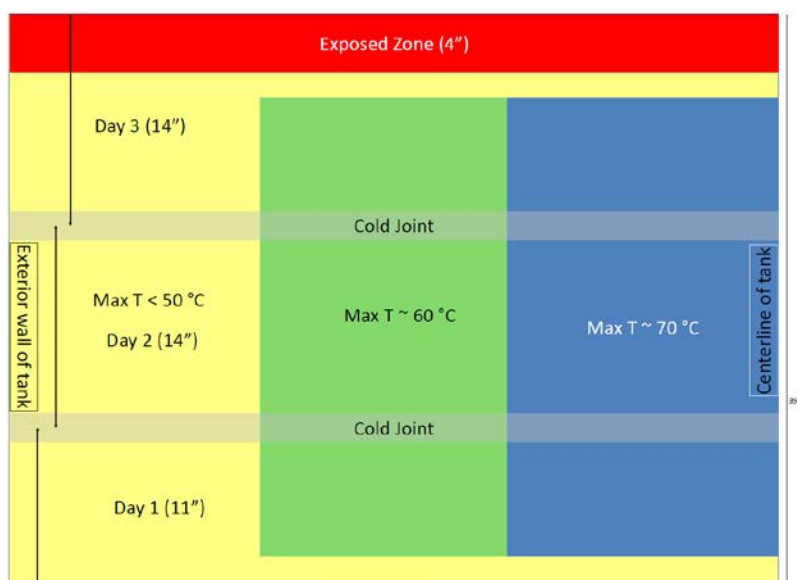
Fig.3.   Graphic representation of the radial temperature zones resulting from the Engineering Scale
          Demonstration.

CONCLUSIONS

The SRNL SCPF was successfully operated over three days to fill a6,056 liter (1600 gallon) polyethylene
container with simulated Cast Stone. Cast Stone grout was processed at a nominal rate of 4.9 L/m for over
16 h. The grout produced was self-leveling across an 260 cm (8.5 ft) diameter with a maximum height
differential across the diameter of   2.5-3.8 cm from the pour entry point to a diametrically opposed point
on the grout container. In-process adjustments of the salt solution flow were able to incorporate and further
deter the formation of excess salt solution on the surface. In-process adjustments of the dry materials feed
demonstrated the ability to process at faster rates than planned.

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