PNNL-16659

Pacific Northwest
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Testing Report: Littleford-Day Dryer Operation Dryer Operation Impacts of Proposed MIS Mitigation Changes

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June 2007

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

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Littleford-Day Dryer Operation
Dryer Operation Impacts of Proposed MIS Mitigation Changes

Revision 0

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June 2007

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Summary

Pacific Northwest National Laboratory performed a series of tests using the Littleford Day 22-liter dryer during investigations that evaluated changes in the melter-feed composition for the Demonstration Bulk Vitrification System. During testing, a new melter-feed formulation was developed that improved dryer performance while improving the retention of waste salts in the melter feed during vitrification. The results of this testing were:

- Finely milled glass-forming minerals (GFMs) performed well in the dryer, producing melter feed with a larger mean particle size than feeds produced with standard soil from the Horn Rapids Test Site (HRTS). This is credited to the GFMs material's capability to retain moisture better than HRTS soil-based feed and increase the agglomerate size of feed particles in the dryer. While feed produced with HRTS soil has a mean particle size between 300 to 500 microns with an ~1 percent moisture content, melter feed produced using GFMs has a mean particle size between 840 to 2000 microns with a moisture content between 2 to 5 percent.
- Alpha-cellulose could be added as a melter-feed component without being detrimental to the process. However, it did have the effect of decreasing the overall bulk density of the input charge material to the dryer by over half, forcing the batch size (mass) of the dryer to be reduced on the 22-liter scale to prevent problems with the drying operation. Overall, the maximum working volume of the 22-liter dryer was found to be 67 percent before impacts on mixing performance were noted.
- Bleed and feed operations were tested on a small scale with the 22-liter dryer, and no impact to product or operations were observed. The zirconium and boron GFMs that were used with the initial charge in the dryer did not have a significant impact on dryer scaling.
- 22-liter dryer results match those seen by the 130-liter dryer, implying that the results of both tests can be applied to the large 10,000-liter dryer.
- Aging studies of the final melter feed showed that this material will exchange moisture with the surrounding air and reach an equilibrium condition that depends on the relative humidity of the air. The melter feed tended to lose moisture when exposed to low relative humidity but quickly gained moisture when exposed to air with a relative humidity near 100 percent.
- Drying with sucrose resulted in sticky feeds that locked up the dryer mixer plows. This occurred when sugar was added as a dry powder at the end of dryer operations or was pre-dissolved and added as a part of the liquid waste.
- Clean-glass batch preparation found that cellulose was not necessary to obtain a large mean particle size in the dryer product. There was no apparent advantage to adding sodium carbonate in the form of a dry material. Using a 5-molar sodium solution of sodium carbonate is recommended for producing clean glass batches.

Acronyms

APEL	Applied Processing Engineering Laboratory
DBVS	Demonstration Bulk Vitrification System
GFM	glass-forming mineral
HRTS	Horn Rapids Test Site
ICV™	In-Container Vitrification
LOD	loss on drying
MIS	molten ionic salt
PNNL	Pacific Northwest National Laboratory
RH	relative humidity
TCLP	Toxicity Characteristic Leach Procedure
WTP	Waste Treatment Plant

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1.0 Introduction

In support of both CH2M HILL and AMEC Earth & Environmental, the Pacific Northwest National Laboratory (PNNL) has been investigating methods of eliminating the migration of molten ionic salt (MIS) during operations of the GeoMelt® In-Container Vitrification (ICV™) System. During those investigations, PNNL proposed that the composition of the melter-feed matrix be modified to reduce the effects of MIS. PNNL suggested that significantly reducing the soil particle size would improve the melter feed's capability to retain MIS within itself by increasing the available surface area for the MIS to adhere to. It was also recommended that an organic reducing agent, such as sucrose, be used as well to reduce MIS migration further by converting molten nitrate salts to oxide upon initial formation. However, these proposed changes to the melter feed composition were considered significant enough that CH2M HILL requested a new series of dryer tests to verify that these changes did not impact operations of the Demonstration Bulk Vitrification System (DBVS). To outline those activities, PNNL created test plan ST07-TP-01, "LITTLEFORD-DAY DRYER OPERATION; Dryer Operation Impacts of Proposed MIS Mitigation Changes" (Elmore 2006). The objectives of this test plan were to:

- verify that the drying behavior and product characteristics are similar for the 22-L dryer operation compared to the recent 130-L dryer tests for the "Baseline Case" using as-received Horn Rapids Test Site (HRTS) soil first mixed with the $Al(OH)_3$ and SiO_2, fed with liquid simulated S-109 waste, and then dry-mixed with B_2O_3 and $ZrSiO_2$
- determine the effects of soil particle-size distribution on the drying process. Soils to be compared are as-received HRTS soil and the ground HRTS soil to be supplied by Praxair.
- determine what effects organic compound additions to the feed matrix have on the drying process and the most efficient method of adding such compounds to the soil matrix during operations. The first organic compound to be tested was sucrose.
- determine the particle size of dryer product produced with ground soil to support CH2M HILL hazards analysis and the feasibility of reducing MIS
- determine, if possible, what the process constraints would be for a semi-continuous process (e.g., discharge mass and frequency).

After initial rounds of dryer tests, it was discovered that other organic materials needed to be tested besides sucrose because of poor dryer performance. Crucible testing also found that crushed soil was not as successful as hoped to mitigate MIS formation. During these tests, the following variants to the test materials were made:

- Cellulose was used as a substitute for sucrose to resolve issues with drying performance.
- Glass Forming Minerals (GFMs) used in the Waste Treatment Plant (WTP) were used as a substitute for ground HRTS soil to resolve issues involving producing a fine enough soil that would consistently mitigate MIS migration.

At the end of this study, melter feed was produced for AMEC's engineering scale test ES-31F to test the repeatability of the final process and characterize the feed produced. Tests were also conducted to verify that the final clean glass melter feed (a mixture of sodium carbonate and GFMs used at the end of the melt) with the desired properties could be produced using the dryer.

2.0 Testing Equipment and Operations

The ICV™ feed dryer system consists of three principal components: 1) a steam-heated Littleford-Day vacuum dryer, 2) a boiler to supply steam to the dryer, and 3) a liquid-ring vacuum pump equipped with off gas filter and condensate collection to provide vacuum to the dryer. Figure 2.1 shows a schematic of the dryer test system, and Figure 2.2 and Figure 2.3 show the experimental setup in the Applied Processing Engineering Laboratory (APEL) where the testing occurred.

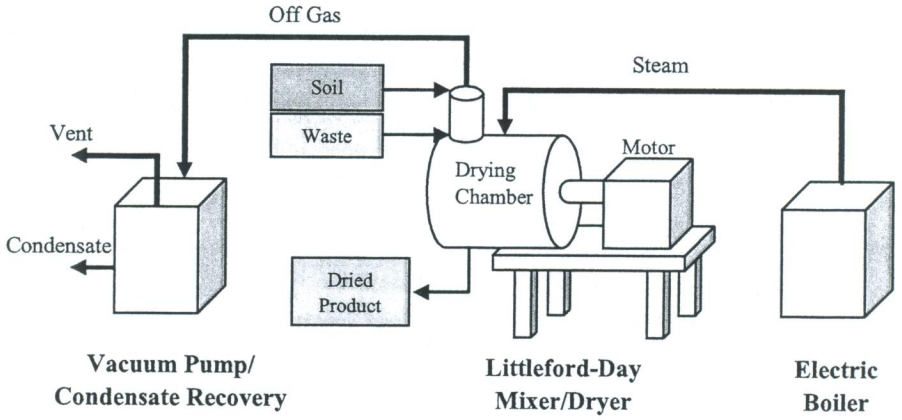

Figure 2.1. Schematic of Small-Scale Batch Soil Drying System

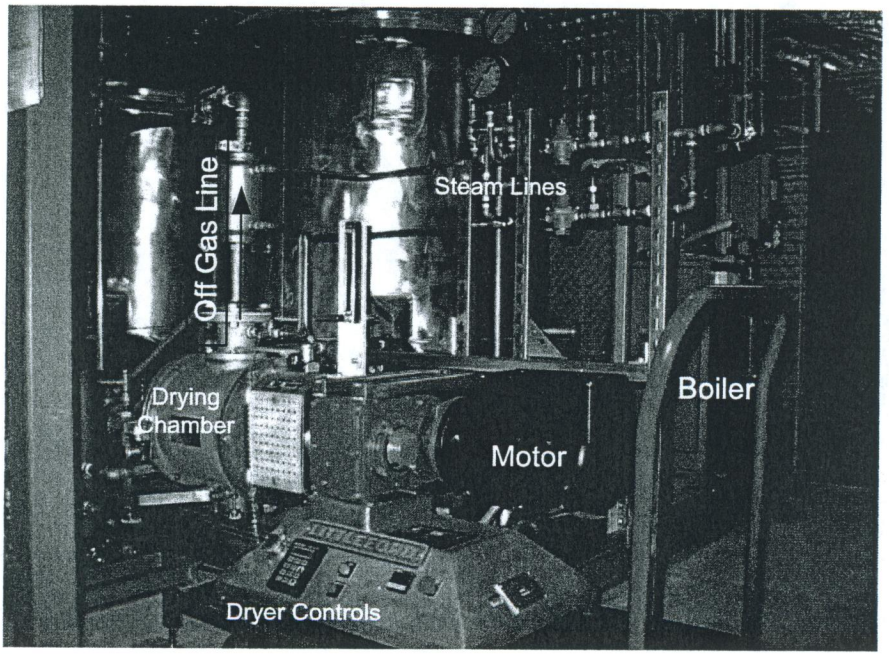

Figure 2.2. APEL Test Setup: Dryer and Boiler

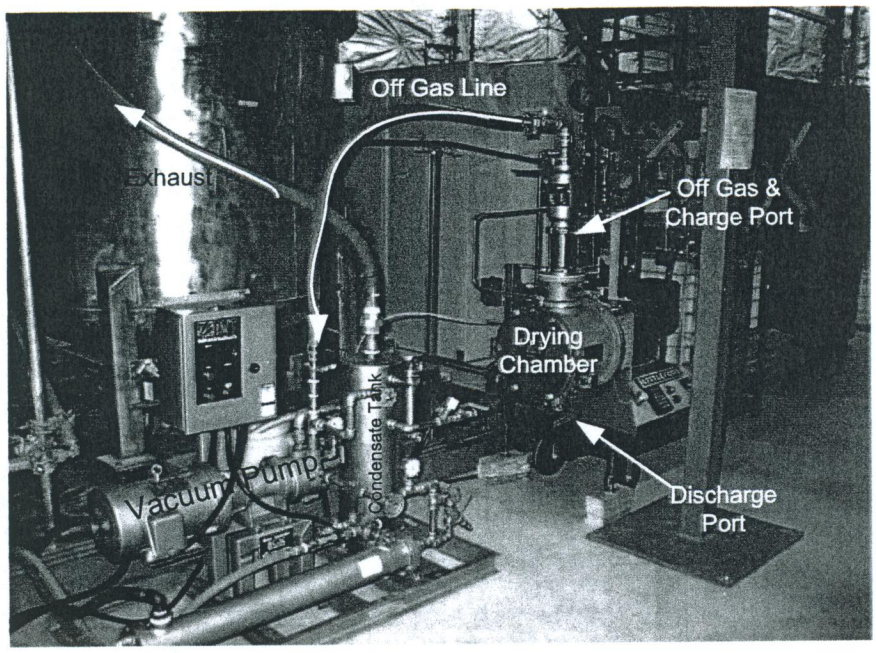

Figure 2.3. APEL Test Setup: Vacuum Pump and Dryer

The dryer has an internal volume of 22 L and is designed to be operated in a batch mode. Pre-measured amounts of soil and amendments are added to the dryer through the top port. Steam is supplied to the dryer jacket to heat the dryer contents and evaporate water. A batch quantity of liquid-waste simulant is fed into the drying chamber at a continuous rate via a metering pump over the course of the drying operation. The rotating mixer in the dryer blends and fluidizes the liquid waste, soil, and glass-enhancing minerals to produce a granular product. The mixer blade is attached to a three-phase, 5-horsepower motor that is geared down to produce mixer speeds up to 240 rpm. A variable speed frequency drive controller is used to control the mixer speed as well as monitor the amperage drawn. Typically, the drawn amperage of the motor falls between 3 to 4 amperes when the mixer speed is operating between 120 to 240 rpm. The vacuum system pulls a vacuum on the dryer to remove evaporated water. Operating the process under vacuum also lowers the drying temperature, thereby reducing the chance of decomposing components of the waste, primarily nitrates and nitrites. At the end of a dryer batch cycle, the vacuum system is isolated, and the dryer discharge valve is opened to remove the dried product, which is then staged as feed to the ICV™ melter. Typical operation limits used in dryer testing are shown in Table 2.1.

Process monitoring of the system was conducted using the following existing instrumentation:

- Dryer's Variable Speed Drive Controller. The controller provides the motor speed and motor amperage of the dryer mixer.
- Dryer's Temperature Monitor. The monitor is connected to a Type-J thermocouple that measures the internal temperature of the dryer contents.
- Pressure gauges and regulators on steam lines to control steam-jacket temperatures.
- Vacuum gauges at the inlet of the ring pump and the dryer off-gas port.

Table 2.1. Testing Operation Limits

Test Parameter	Units	Range	Control
Dryer Shell Steam Pressure	psig	15–20	Steam Regulator
Internal Dryer Temperature	°F	140–200	Waste Feed Rate
Dryer Vacuum	Inches of Hg	15–29	Vacuum Pump
Dryer Agitator Speed	RPM	128–256	Dryer Speed Control
Motor Amperage	Amps	3.1–3.9	Dryer Speed Control
Waste Addition Rate	cc/min	30–80	Metering Pump

The criteria for success or failure of the product were based on whether the material could be successfully processed in the dryer and produce a granular melter feed product.

Characterization of the final product's moisture content, particle-size distribution, and material flow capability were made. Moisture content was determined by loss on drying (LOD) testing. LOD measurements were made by heating samples for 24 hours in an oven at 105°C and determining the weight loss. Particle-size distributions were determined by screening the product through various sized sieve trays using a mechanical shaker for a 5-minute period. Table 2.2 lists the sieve trays used for the characterization.

Table 2.2. Sieves Used for Particle Sizing

Sieve Screen Size	Screen Opening (μm)
¼ in.	6350
U.S. Mesh Size	
6	3360
10	2000
20	841
45	354
325	44

The capability of the material to flow was characterized by slump testing (see Figure 2.4). This was done by pouring material through a funnel until a stable pile was formed. Angles of repose were then estimated by examining digital pictures of the stable pile of feed.

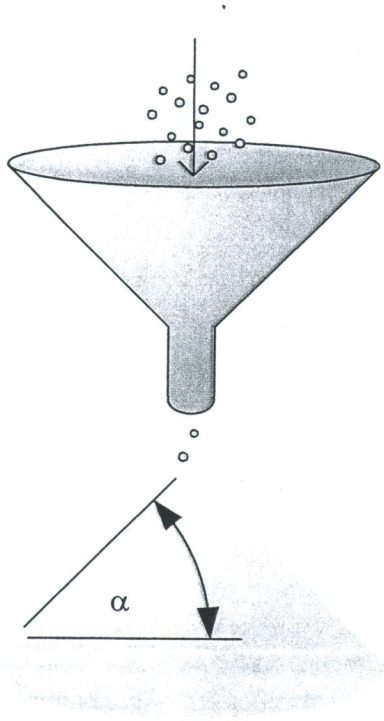

Figure 2.4. Slump Testing and Angle-of-Repose Estimation

3.0 Test Descriptions and Results

3.1 Dryer Testing Using Crushed HRTS Soil and Sucrose

3.1.1 Test Matrix and Objectives

The MIS dryer test program was initiated to verify the effects of the proposed changes to the feed matrix. The objectives of the original test matrix were to:

- verify that changes in the base-line simulant and the use of zircon as a replacement for zirconium oxide did not impact operations of the 22-Liter Dryer and was still comparable to the 130-Liter Dryer
- verify that crushed HRTS soil provided by Praxair did not negatively impact drying operations significantly
- verify that initial additions of sucrose as a de-nitration agent did not impact drying operations significantly.

Operation of the dryer was kept consistent for this series of tests. The process parameters for these tests were:

- Waste Feed rate: 40–45 cc/min
- Final Batch size: 19–20 kg
- Mixer Speed: 128 rpm
- Motor Amperage: 3.4 Amp (Max.)
- Shell steam pressure: 15–18 psig

Figure 3.1 is a flow diagram of the original test matrix provided in PNNL ST07-TP-01.

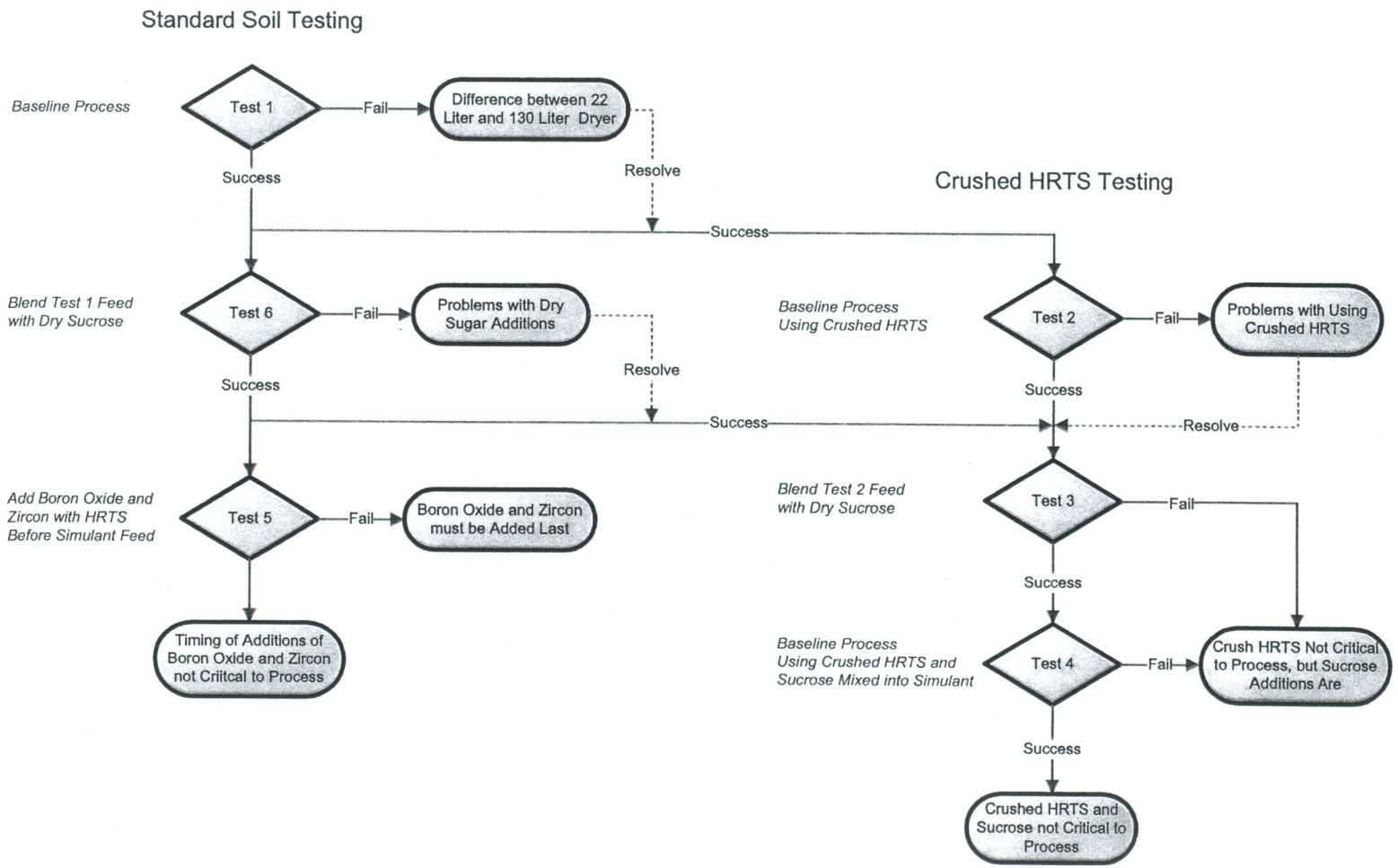

Figure 3.1. Original Test Matrix

3.1.2 Testing Results

Table 3.1 lists the results from the original test matrix.

Table 3.1. Test Results from Original Testing Matrix

Test	Description	Result
Test 1	Baseline Product	Success
Test 2	Crushed HRTS from Praxair	Success
Test 3 Test 6	Dry Sucrose Additions to Tests 1 and 2	Failure
Test 5	Addition of Zircon and B_2O_3 with Soil	Success
Test 4*	Sucrose Addition Mixed with Simulant	Failure
* Test performed with standard soil instead of crushed soil due to availability.		

Success and failure of these tests were determined by whether the produced product was similar to that found in previous testing, and it did not produce scale on the dryer walls.

The dryer testing without sucrose additions produced a product similar to that obtained from the "dry process" in the 130-liter-scale dryer as described in earlier reports by Daniel, Mann, Johnson and Mendenhall (DMJM Technology) (Nigel et al. 2004), and the 22-liter-scale dryer test at PNNL (Shimskey and Elmore 2005). The inside walls from the dryer appeared to have little build-up, and a free-flowing granular product was produced with a final moisture level between 1 to 2 percent. Figure 3.2 shows an image of the produce melter feed as well as the results from particle sieve analysis.

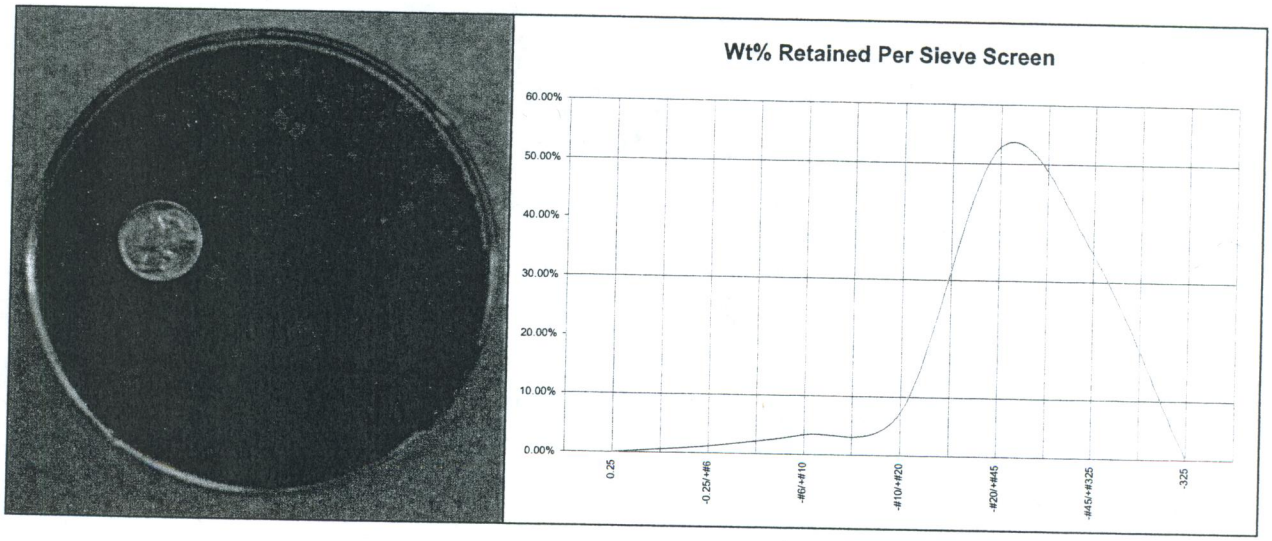

Figure 3.2. Baseline Melter Feed Results

Overall, these results match the 130-liter tests performed by CH2M Hill using HRTS soil, as reported in RPP-RPT-31688 (Tedeschi 2007). Described as Phase 1 and Phase 2 tests, operating that dryer with a continuous waste-simulant stream produced a final product with a similar size and moisture content and feed rates that were scalable to the size of the initial charge of the dryer. The internal temperature of the 130-liter dryer still is lower than the 22-liter dryer. Some of this may be a scaling issue, but the likely reason is that the 130-liter dryer has a superior vacuum system. While the 22-liter dryer achieved vacuum levels between 18 and 22 in Hg, the 130-liter dryer operated closer to 25 to 26 in Hg vacuum. This decrease in vacuum level is significant enough for the evaporation temperature of water to be lower in the 130-liter dryer, and this explains the decrease in the internal dryer temperature.

Test 2 was a repeat of Test 1, except that the standard HRTS soil was substituted with HRTS soil that was mechanically milled by Praxair to reduce the average particle size. The results of the test were similar to that found in Test 1. Figure 3.3 shows the condition of the dryer after Test 2 and the appearance of the produced melter feed.

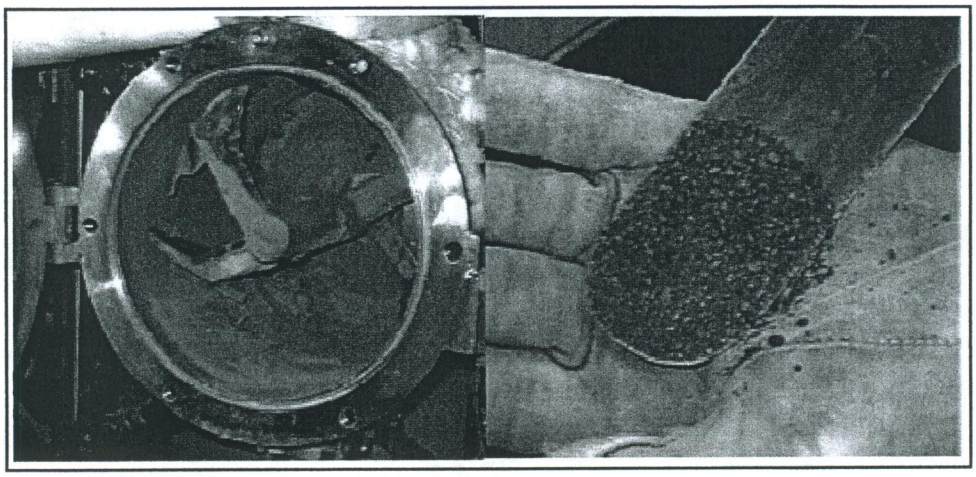

Figure 3.3. Test Results from Processing Ground HRTS Without Sucrose

The results of dryer testing involving additions of sucrose were not successful. Both batch additions of dry product and gradual additions of sucrose dissolved in the simulant proved to have impacts on the process. In the case of the dry addition of sucrose to feed (Tests 3 and 6), this was done in the dryer while the feed was still warm to simulate additions in the full-scale dryer. Because the product was moist and warm, the sugar dissolved and turned the feed into a viscous material. Once the moisture level and temperature of the feed decreased, the feed material solidified to a solid mass around the mixer blade, requiring it to be manually chipped out of the dryer (left picture in Figure 3.4). Blending the sucrose with the simulant and gradually feeding it to the soil matrix (Test 4) was also unsuccessful. The material quickly began to stick to the side walls of the dryer within the first hour of a 6-hour process, forcing the discontinuation of the test. A hard scale formed on the inside of the dryer walls, which showed visible wear markings from where the mix blades came into contact (right picture in Figure 3.4).

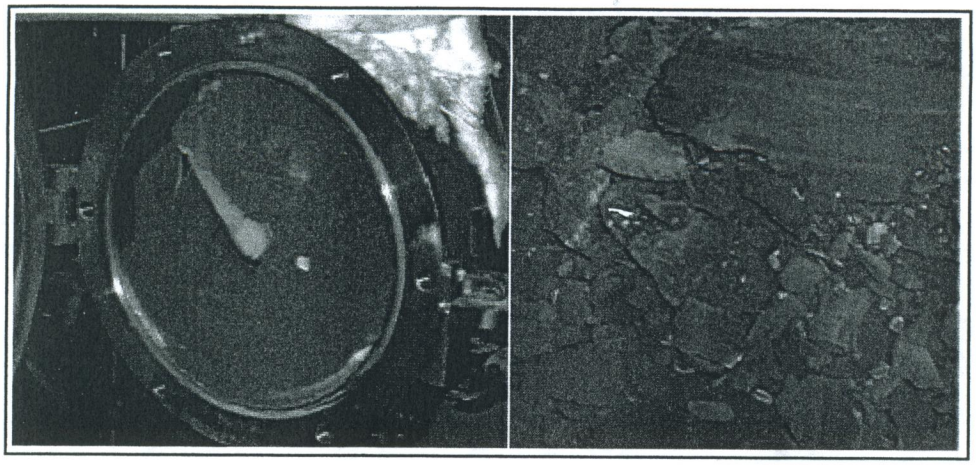

Figure 3.4. Results from Test 3 (left) and Test 4 (right)

After the sucrose proved to be problematic, Test 5 was attempted to understand the effect of adding zirconium and boron glass-enhancing minerals at the beginning of the process. Unlike the previous studies performed by DMJM Technology, the results from Test 5 showed that adding boron and zirconium minerals with the soil had no impact on the process and produced a product much like Tests 1 and 2. The size of these minerals probably had no impact on this because the boron oxide source has remained the same, and multiple sizes of zirconia were tried in the past—some as large as 1 mm in diameter. It is believed that the switch to zircon as a source of zirconium is the most likely reason why adding these minerals is no longer a problem. However, this is speculation and was not experimentally confirmed.

3.2 Testing Using WTP Glass Minerals and Cellulose

3.2.1 Test Matrix and Objectives

After the end of the initial test matrix, the MIS mitigation program had two issues requiring resolution:

- The initial dryer tests presented enough concerns about adding sucrose to the melter feed that it was deemed unacceptable.
- Crucible testing that was occurring parallel to the dryer testing showed that Praxair was not able to use production-viable grinding equipment to grind HRTS soil to the levels necessary to control MIS migration.

Because Hanford soil particle-size reduction was impractical and sucrose performed poorly in dryer operations, potential solutions for both of these issues needed to be found and verified with dryer testing. After discussions with PNNL staff and initial crucible testing, cellulose was suggested as a replacement for sucrose for dryer testing because it was less soluble than sucrose but still performed similarly as a reducing agent for the nitrates present in the MIS. While Praxair continued to look at ways to grind HRTS soil to required levels, it was also decided to purchase finely milled minerals from WTP suppliers as a viable replacement for crushed HRTS. Using WTP minerals has two advantages: 1) there is much better control of particle sizes and chemical composition than using HRTS soil and 2) using WTP allows for precise control of the final glass composition. For these tests, a blend of WTP minerals is used to

simulate the HRTS composition to produce the same glass evaluated for this project. However, if a superior performing glass is developed in the future, the use of WTP minerals would make implementing a change much easier.

Figure 3.5 outlines the test scheme used to explore the use of cellulose as a replacement for sucrose and how it behaved with different mineral/soil matrixes during dryer testing.

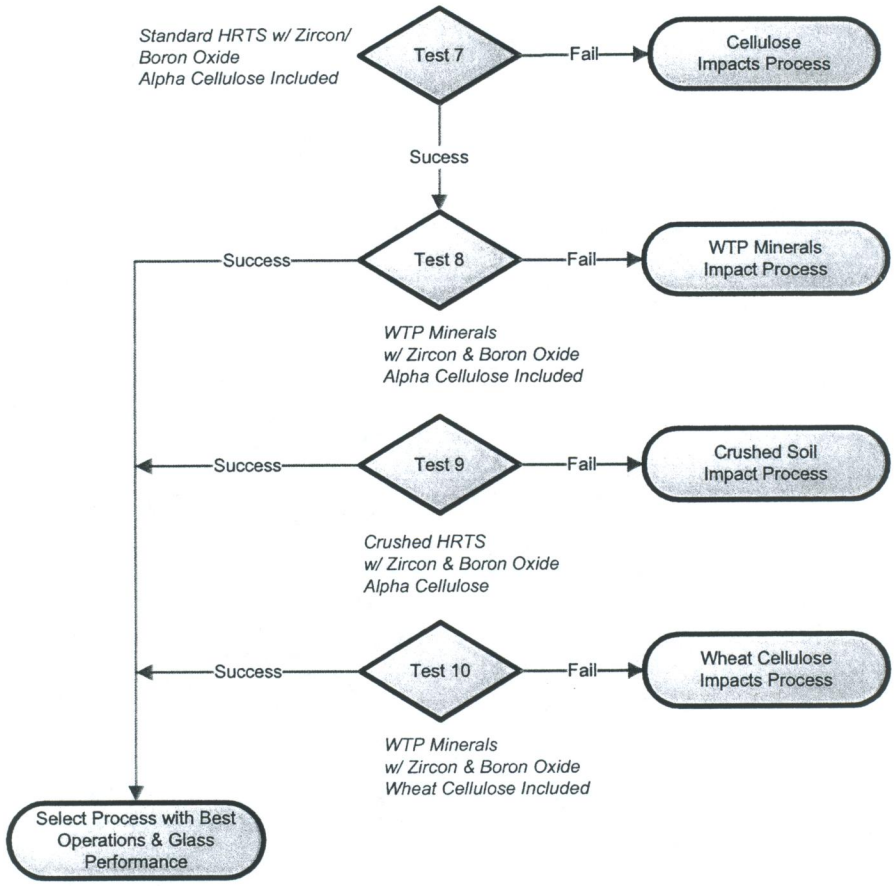

Figure 3.5. Modified Test Matrix to Resolution Issues from Initial Testing

Like before, attempts were made to maintain consistent operation of the dryer. However, batch sizes and feed rates were found to be variables that could not be fixed for these tests. Also, the mixer-blade speed was increased to 218 rpm so the mixer blade tip speed would be closer to that used on the 130-liter dryer and be more prototypic of the full-scale dryer based on a recommendation received by CH2M HILL personnel during operations at Littleford Day.

The process parameters for these tests were:

- Mixer Speed: 218 rpm
- Maximum Motor Amperage: 3.4 Amps
- Shell steam pressure: 15–18 psig.

3.2.2 Testing Results

Table 3.2 lists the results from the modified test matrix.

Table 3.2. Testing Results of the Modified MIS Mitigation Dryer Test Matrix

Test	Description	Result
Test 7	HRTS Soil Alpha Cellulose	Success
Test 8	WTP Minerals Alpha Cellulose	Success
Test 9	Crushed HRTS Soil Alpha Cellulose	Success
Test 10	WTP Minerals Wheat Cellulose	Failure

Like previous testing, success and failure of these tests were determined by the dryer product. If the product was similar to that found in previous testing and it did not produce scale on the dryer walls, the test was successful.

The initial testing of cellulose was with an alpha-cellulose-grade product (cotton based) and standard soil (Test 7). This type of cellulose was the purest and least soluble form of the chemical, which presented the best chance of success. The cellulose was batched with the soil at the beginning of the process, with 75 percent of the starting mass that has been used with the previous 22-liter dryer test (~ 9 kg). This was done only because the amount of alpha-cellulose available for a quick-turnaround screening test was limited.

Using a simulant waste-feed rate of 40 cc/min, the material processed in the dryer without much issue. The material appeared similar to product produced from previous tests with standard HRTS, and the dryer did not have any significant buildup on the dryer walls (see Figure 3.6). The only issue was that a high frequency of filter back-pulsing was necessary during operations to maintain the vacuum level. The cellulose appears to be easily carried up into the off-gas system because of its low density, which created the need for the frequent filter back-pulsing. Figure 3.6 shows areas where alpha-cellulose was deposited in the off-gas piping.

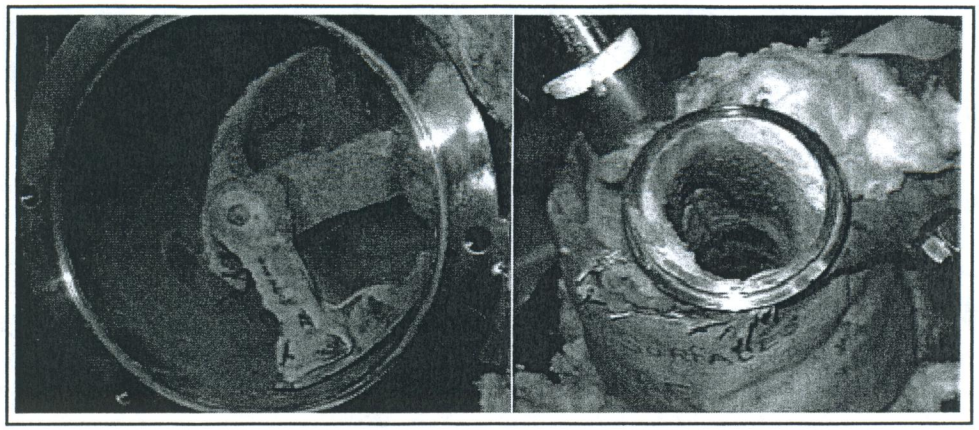

Figure 3.6. Standard HRTS Soil with Alpha Cellulose

Test 8 used alpha-cellulose again, but replaced the HRTS soil with a blend of finely milled GFMs. The blend was created to simulate HRTS soil chemically, but possesses a much smaller and controlled particle-size distribution. The initial batch mass was 80 percent of the first series of tests. However, the test was terminated when the vacuum to the system was shut down because the entire off-gas piping filled with material, including the filter.

The bulk density of the starting material was dramatically lower after adding cellulose (decreasing to 0.5 to 0.6 gm/cc), which required a reduced initial batch mass. The previous tests only filled the dryer to the top of the mixer blade shaft (11 to 12 kg filling 10 liters of space). However, an initial mass of 9.5 kg of cellulose and GFMs almost completely filled the dryer to the entrance of the off-gas pipe (~20 liter volume). Without sufficient void space in the dryer, the material cannot fluidize properly, and in the case of the 22-liter dryer, the mixer blades will force the excess material into the throat of the off-gas piping.

The test was restarted with half the original batch size. The simulant waste-feed rate was also dropped to 30 cc/min because the batch size was reduced. The reduced batch size alleviated the main vacuum system problem, but frequent back-pulsing of the off-gas filter was required.

In the past, frequent back-pulsing of the filter was a sign that the feed product was too dry and very dusty. Based on the results of the first test, it was hypothesized that the feed rate could be increased to keep the cellulose moist enough stay in the feed and not to be carried to the filter by the off-gas flow exiting the dryer. While monitoring the feed temperature and the motor amperage, the feed rate was incrementally increased to a maximum feed rate of 70 cc/min until the back-pulsing frequency began to subside, and the feed temperature began to decrease. After an hour of operation, the waste-simulant feed rate was decreased to a range of 55 to 58 cc/min, and the temperature stabilized. Samples taken of the feed showed a much more granular product (Figure 3.7) than what was produced from previous testing, and the motor amperages dropped from 3.4 amperes to 3.1 amperes. Examining the dryer once the process was completed showed a clean inner surface (Figure 3.7).

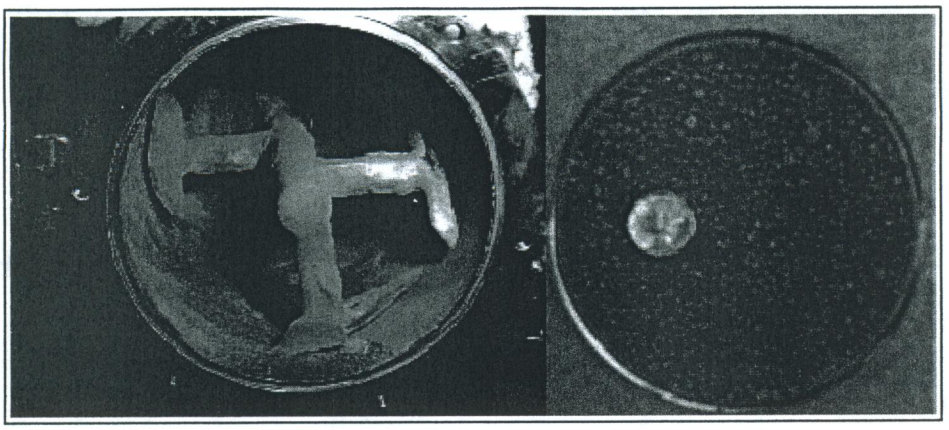

Figure 3.7. WTP Minerals with Alpha Cellulose

After sampling the feed, the dryer was reloaded with the dried product from the first batch and the other half of the dryer feed material not processed the first time. This second operation simulated "bleed and feed" operations proposed for the DBVS system. Unlike the first attempt to process the total mass, this attempt worked well. With half of the total mass in a granular form, there was sufficient void space in the dryer to fluidize the material. The feed rate was kept the same as before, and the combined batch processed similarly. With vacuum on the dryer operating close to 20 in. Hg, the process temperature of the feed stayed between 160 to 170°F with the motor amperage dropping to from 3.4 to 3.1 over time. In the end, the final product looked much like the product from the first half-batch. Figure 3.8 shows the visual results of slump testing of the feed after the entire batch was processed. An estimated angle of repose between 30 and 35 degrees was measured.

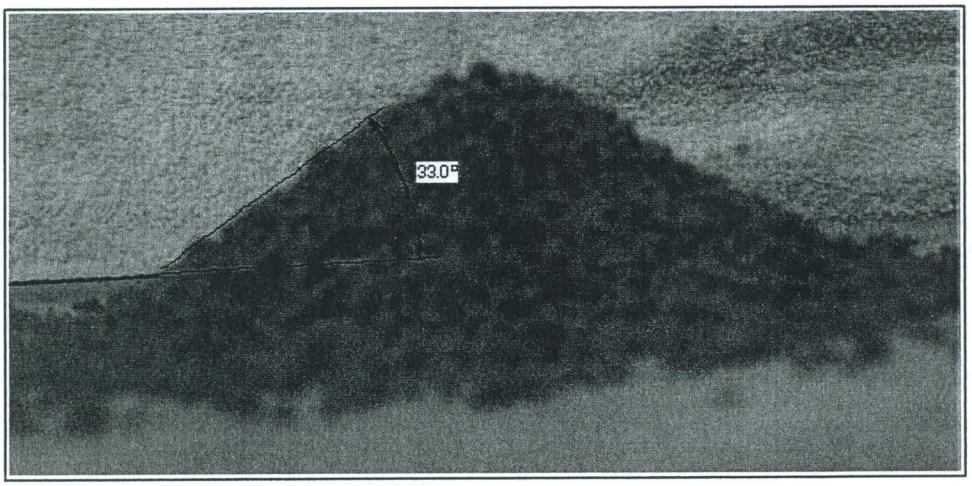

Figure 3.8. Slump Testing of GFM Feed

Once Test 8 was completed, Praxair provided more HRTS soil to PNNL, which was plate milled four times to produce a soil that was much finer than the first attempt. Visual examination of the product showed it to look more like the WTP glass mineral blend. This material was used as the mineral basis for Test 9.

The batch size for Test 9 was similar to the original batch size of Test 8. Like before, there was little void space in the dryer once all the material was added. In attempts to increase the available void space in the dryer, water was added to the material before the drying operations (~400 gm). However, the off-gas port once again plugged up after 10 minutes of operations, and the batch was split in half.

Once the batch size was reduced, the material processed very similarly to Test 8. The feed rate was established at 60 cc/min, and the motor amperage dropped about 1 hour into the process. At this point, in-process sampling showed that the feed matrix was very granular. Once the first half was processed, the un-processed half of the batch was loaded into the dryer and processed as a "feed and bleed" operation. As before, the material produced was granular, and the inside of the dryer had very little buildup (Figure 3.9).

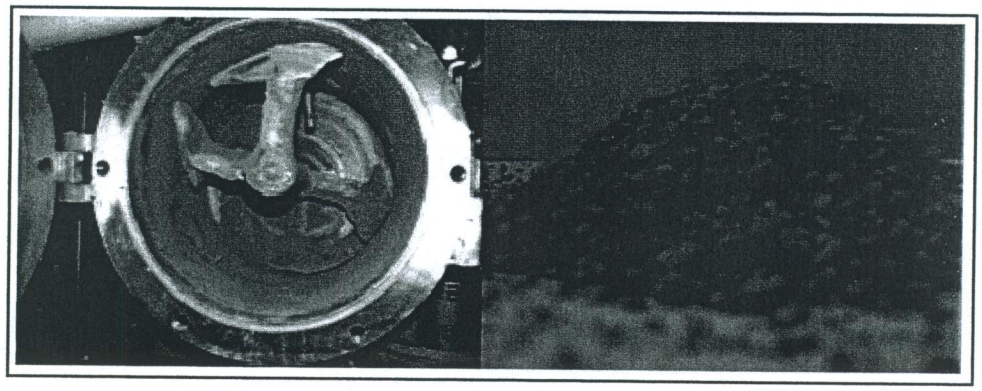

Figure 3.9. Ground HRTS Soil with Alpha Cellulose

Slump testing was also performed to understand the flow ability of the final product, as shown in Figure 3.10. The material appeared to have a higher angle of repose than what was seen in Test 8. It was estimated to be between 35 and 40 degrees.

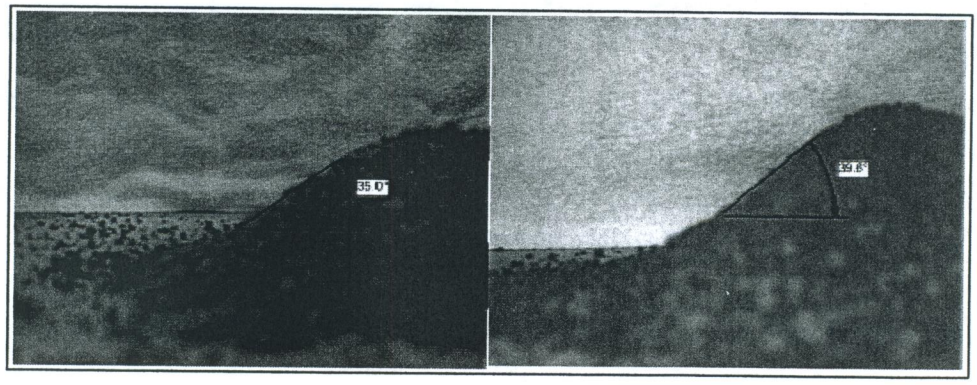

Figure 3.10. Slump Testing of Test 9 (ground HRTS and cellulose)

Test 10 was performed to examine the behavior of cellulose from a different source to see how its quality impacts this process. A wheat-based cellulose that contained trace amounts of proteins and starch was used for this test, using GFMs as the mineral base for the feed. Based on the previous testing, the batch size and feed rate were kept the same as in the previous tests.

Initially, the material processed like the previous tests using alpha cellulose. However, the motor amperage began to increase about an hour into the process. Once the motor amperage climbed past 3.9 amperes, the test was terminated. Examining the inside of the dryer showed a layer of build-up at the base of the dryer, which the blades were rubbing against. While this is indicative of the material becoming too wet, the product inside the dryer was about the same as that in Tests 8 and 9. Also, the feed temperature of the batch was well over 170°F when this behavior began, indicating that the process was not getting too moist. It is possible that the starch and proteins present in the wheat cellulose may have behaved like the sucrose and began sticking to the walls. Figure 3.11 shows the buildup inside the dryer from this test. Another issue was that the feed did not form the same granular product as the alpha cellulose. Even though the test was finished early, the feed should have shown some sign of this behavior. Further tests might have resolved the dryer issues, but the decision was made to proceed with the alpha cellulose for near-term engineering and full-scale proof-of-concept tests.

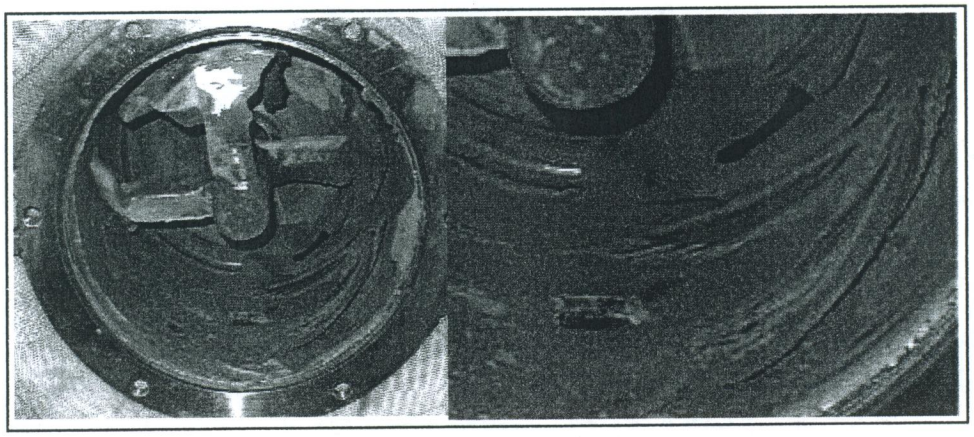

Figure 3.11. WTP Minerals with Wheat Cellulose

3.3 Processing ES-31F Results

3.3.1 Operational Results

Once the second test matrix was completed, a decision was made on the makeup of engineering-scale tests of ES-30K and ES-31F for AMEC.[(a)] Crucible testing indicated that the 22-liter dryer feed produced from GFMs and alpha-cellulose (Test 8) performed the best. With the success of the dryer and crucible tests from Test 8, it was decided that GFMs and alpha-cellulose would be the basis of the vitrified feed. It was decided that the feed for ES-31F would be made with the 22-liter dryer.

For the test, two variants of the S-109 simulant were used and blended together for the test. Table 3.3 lists the composition of each of these simulants.

(a) K. G. Finucane, and K. S. Witwer (AMEC), and D.-S. Kim, M. J. Schweiger, C. P. Rodriquez, J. Matyas, B. J. Riley, and L. M. Bagaasen (PNNL). May 2007. Demonstration Bulk Vitrification System (DBVS) Series 30 and 31 Testing—ES-30J, ES-30K, and ES-31F Test Report. 30686-RT-0001, Rev. 0A (DRAFT). Prepared for CH2M Hill Hanford Group, Inc., Richland, Washington

Table 3.3. Composition of S-109 Simulants

	Component Mass (gm/kg of Simulant)	
Chemical	**Old Simulant**	**New Simulant**
$Al(OH)_3$	2.76	0.69
$Ca(OH)_2$	0.07	-
NaCl	0.33	0.59
Na_2CrO_4	0.75	-
Cr_2O_3	0.50	
$Na_2CrO_4 \cdot 4H_2O$	-	2.63
NaF	0.18	-
Fe_2O_3	0.29	-
KNO_3	0.34	0.34
$NaNO_2$	3.42	3.43
$NaNO_3$	281.06	281.74
NaOH	6.39	6.23
$Na_3PO_4.12H_2O$	16.71	16.75
Na_2SO_4	4.20	4.21
$Na_2CO_3.H2O$	16.83	16.87
$NaCH_3COO$	0.10	0.82
$Na_2C_2O_4$	1.17	-
H_2O	664.91	665.71
Total	1,000.0	1,000.0

Equivalent fractions of simulant were to be used for ES-31F. Because approximately 80 gallons of prepared simulant was required, a 40-gallon batch of each simulant was prepared in separate 55 gallon drums, using de-ionized water. No significant temperature deviations or mixing problems occurred, although some residual material did not go into solution. For each dryer batch, equivalent fractions of each simulant were then mixed together, along with a small concentrated solution containing rhenium and other chemicals of potential concern used to evaluate Toxicity Characteristic Leach Procedure (TCLP) performance of the glass. As before, no unusual behavior was noticed from blending the simulants together. While some of the salts did not dissolve completely, no additional precipitation of solids occurred after blending, and material passed through the system's metering pump without issue.

The quantity of feed produced for ES-31F was 352 kg, which was done in 27 batches. This process was originally scheduled to be performed as 22 batches, each with a final mass of 16 kg. However, this meant that the mass loaded into the dryer was going to be approximately 9.5 kg. From the previous testing, it was expected that this quantity of starting material would be a problem. While the first batch was completed, the effort to keep the off-gas throat unplugged made operations twice as long as needed. The batch size was then reduced by 20 percent mass (7.5 kg), which was approximately 67 percent of the internal dryer volume. This initial charge seemed to work fine for the remaining 26 batches.

However, it should be noted that feed control still seems to be the most critical aspect of this process. During operations, a problem was discovered with the simulant waste metering pump that could double the pump's flow rate at the same setting. This problem was not resolved until the 13th batch, when it was discovered that accidentally dead heading the pump could increase the stroke length. For over half of the first 12 batches, the process went "wet," and the dryer contents turned into mud. While it is not clear how high the flow-rates were for some of those batches, the last flow-rate measured showed that the flow was as high as 120 cc/min. In this case, the granular texture of the feed turned into 1-inch diameter balls, and a ¼-in. wet layer of clay needed to be hand scraped off the dryer walls. This material was later crushed to verify that a majority of it would pass through a ¼-in. screen. As a result, much of this feed material was reduced to a fine powder. To prevent this from re-occurring, the calibration of the metering pumps was verified during operations by placing the waste simulant on a scale and tracking its weight loss over time.

Batches 14 through 27 behaved identically to Test 8 and were trouble free once the feed-rate issues were resolved. Overall operations of these batches were as follows:

- Waste Simulant Feed rate: 50–70 cc/min
- Final Batch size: 13 kg
- Startup Mixer Speed: 128 rpm
- Final Mixer Speed: 218 rpm
- Motor Amperage: 3.1–3.4 amperes
- Shell steam pressure: 20 psig.

To prevent buildup of material at the ends of the 22-liter dryer, the initial temperature of the dryer was allowed to heat-up to 200°F before the meter pump was started. Typically, the ends of this dryer experiences some buildup because they are not heated, and condensation occurs there. Allowing the internal dryer temperature rise to 200°F was an attempt to pre-heat the ends of the dryer to reduce this buildup. Once waste simulant was introduced to the dryer, the internal temperature rapidly decreased between 150 to 170°F. No problems during operations (e.g., cellulose reactions) were experienced when the dryer temperature reached 200°F.

Once the pump feed rate was established, it was not changed for the entire batch. The initial mixer speed was started at 128 rpm, which significantly reduced back-pulsing of the filter system. After 30 to 45 minutes of operations, the mixer speed was increased to 218 rpm so the mixer blade tip speed would be closer to that seen at the 130-liter dryer, which operated at 160 rpm. The steam jacket pressure was also increased to 20 psig, but this was done to improve performance of the steam trap, which was sized for a higher pressure system. Despite the slight changes in mixer speed and steam pressure, all 14 batches performed similarly to Test 8, producing a granular feed with minimal scale buildup forming on the inside walls (Figure 3.12).

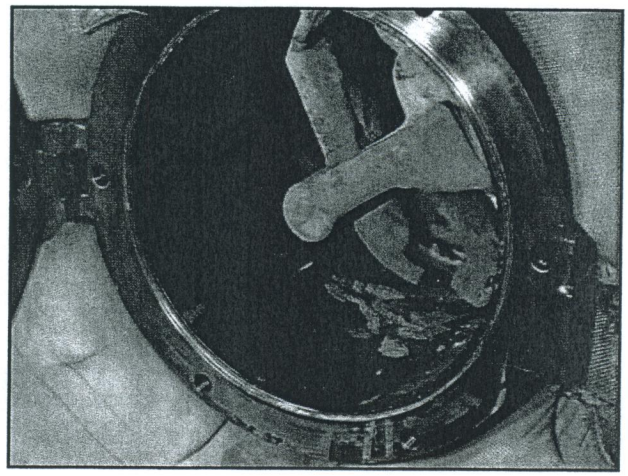

Figure 3.12. Final Dryer Condition after 27th Batch for ES-31F

3.3.2 ES-31F Feed Characterization

To understand the effects of feed flow rate on final moisture level and particle size, the feed rate was incrementally increased from batches 14 to 24. Samples from each dryer batch were tested for LOD and particle-size analysis. Table 3.4 and Figure 3.13 show the results of this testing. The results generally show that higher feed rates result in a dryer product that has higher moisture content and a larger particle-size distribution.

Table 3.4. ES-31F LOD and Sieve Screen Analysis

Batch No.	Feed Rate (cc/min)	Percent Weight Loss due to Dryness (LOD)	Weight Percent of Feed Sample Captured by Sieve Tray						
			0.25	-0.25/+#6	-#6/+#10	-#10/+#20	-#20/+#45	-#45/+#325	-325
14	55	2.5	0.0	7.6	53.7	34.7	3.0	0.7	0.2
15	50	1.3	0.0	1.3	15.0	54.8	24.4	4.0	0.4
16	55	2.0	0.0	3.4	35.4	55.1	5.3	0.6	0.2
17	58	3.4	0.0	3.4	33.9	53.8	8.1	0.7	0.1
18	58	2.9	0.0	2.1	30.6	54.5	11.7	1.0	0.1
19	59	2.1	0.0	1.9	26.1	56.5	14.1	1.3	0.1
20	60	2.5	0.0	3.8	43.3	48.4	3.6	0.7	0.3
21	63	3.4	0.0	5.1	36.0	49.2	8.7	0.9	0.0
22	65	2.6	0.1	2.8	24.6	51.6	18.2	2.5	0.1
23	68	3.4	0.0	3.1	34.1	54.5	7.5	0.8	0.1
24	70	4.3	0.0	9.5	38.4	39.0	9.9	2.8	0.3
25	70	3.4	0.7	9.2	40.6	38.2	9.5	1.5	0.3
26	70	3.1	0.0	1.8	33.3	49.9	12.9	1.9	0.1
27	70	4.7	0.1	8.8	41.9	38.8	8.8	1.6	0.0

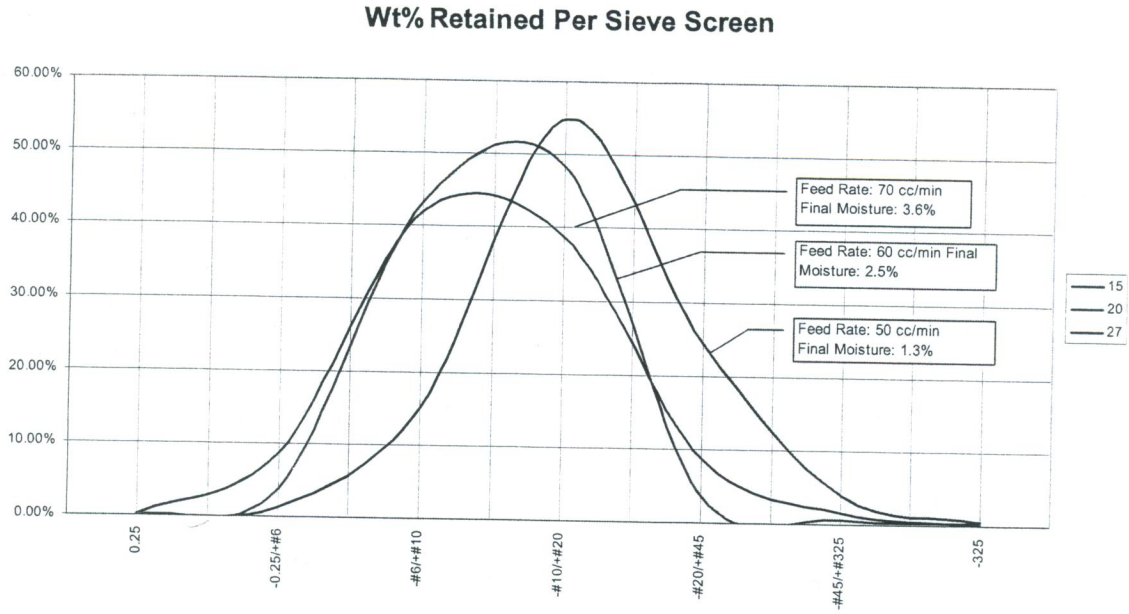

Figure 3.13. Particle Distribution of ES-31F at Different Feed Rates

Examining the Phase 3 results in RPP-RPT-31688 shows similar particle-size distributions and moisture levels for the first half of their testing. During this period, they also produced a granular product with moisture levels between 2 to 4 weight percent. However, the feed did not retain these characteristics for the second half of the testing. Despite increasing the feed rates to a point where the moisture level was greater than 5 weight percent, the feed had a very fine texture. While this was not experienced while processing ES-31F, this result was seen in later testing.

3.3.3 ES-31F Moisture Aging Study

Because of interest in how the melter feed's moisture level changes outside of the dryer, an aging study of this material at two different humidity levels was performed. An initial sample was taken from batch 27 immediately after being discharged from the dryer after processing, and was called the "as-processed" sample. Next, half of this sample was dried in an oven to complete dryness, and was called the "dried" sample. Each of the two samples were then split again and aged in two different humidity environments. The first test was to place an as-processed sample and dried sample in a sealed container with an open vial of water to humidify the air inside to 100 percent relative humidity (P_{H20} = 18 torr). The second test was to leave an as-processed sample and a dried sample to age on the bench-top exposed to ambient air, which was approximately 30 percent relative humidity (RH) at room temperature (P_{H20} = 5 torr).

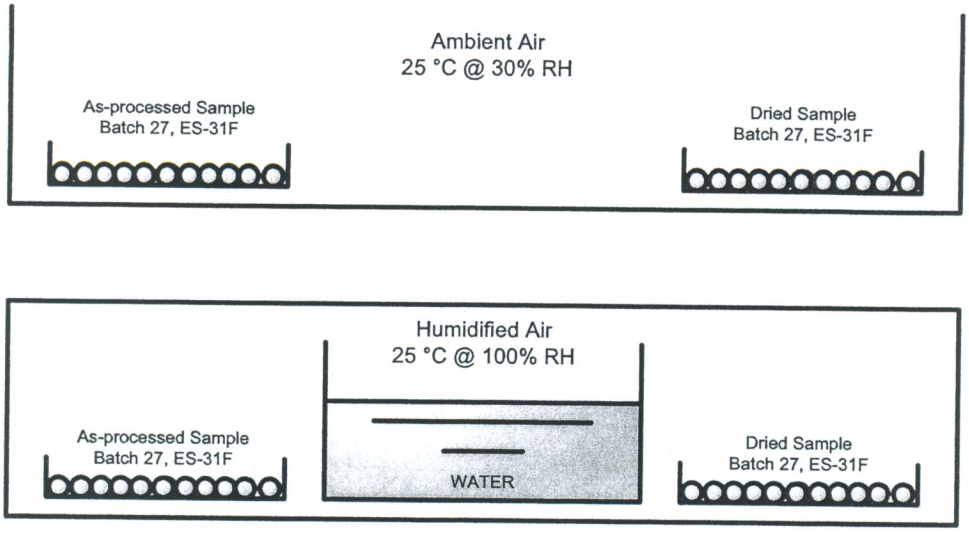

Figure 3.14. Feed Moisture Aging Study

Over time, the mass change was measured of each sample, allowing the change in moisture level to be calculated. Figure 3.15 shows the results of this testing.

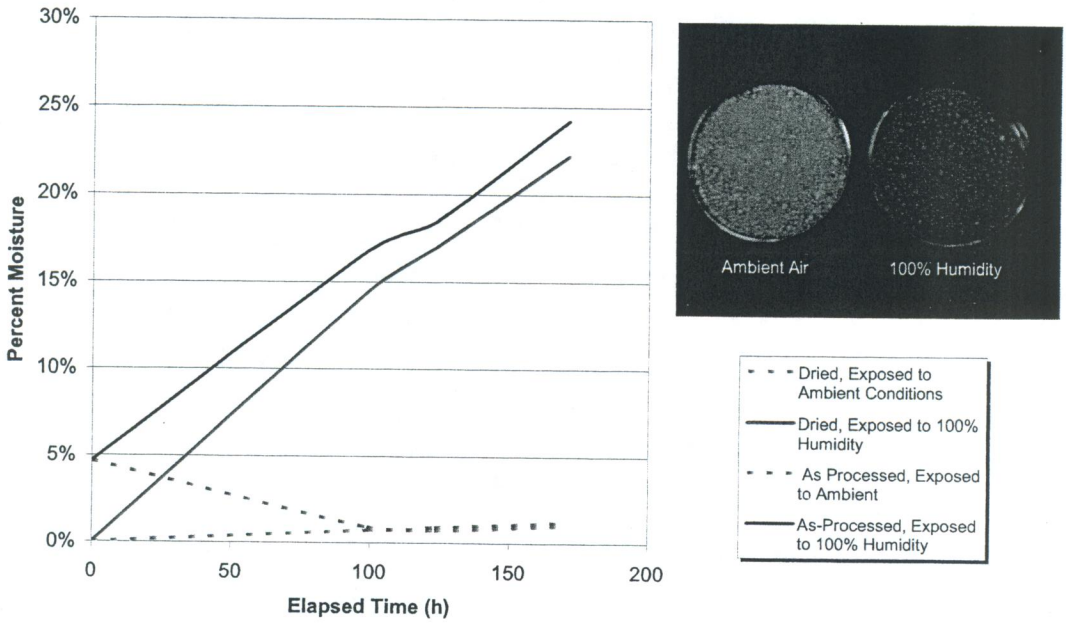

Figure 3.15. Moisture Testing of ES-31F

Over-time, both the as-processed sample and the dried sample approach the same moisture level, indicating that the feed moisture level is a function of the ambient partial pressure of water. The 100 percent RH test is of some interest because the water absorption rate did not slow down after 200 hours. The material began to lose strength as it picked up moisture and stick to itself. Eventually, the test was stopped at the 200-hour mark once liquid was seen forming at the base of the melter-feed

particle. This liquid appeared to be coming from the feed particles themselves and looked very much like the original waste simulant used.

While the humidified samples turned into mud, the samples aged in ambient air conditions maintained their structure over time. Both the dried sample and the original as-processed sample had the same moisture levels of about 1 weight percent at the end of the test.

The test indicates that the partial pressure of water present in air directly affects the residual moisture level of the melter feed over time. At lower partial pressures, these salts will maintain a stable moisture level in the feed that does not affect the feed particles' physical integrity. At higher partial pressure, the hygroscopic nature of the waste simulant salts can create a run-away condition where water will continually be absorbed into the melter feed. Eventually, the particle can no longer absorb moisture, and it turns into a liquid solution.

PNNL examined this phenomenon in the past with tank-waste solutions. Various liquid tank wastes and simulants were examined under different partial pressures of water (Scheele et al. 1996). Sodium hydroxide, sodium nitrate, and sodium nitrite were major components of these wastes and were found to be major contributors to this effect. The study found that at typical humidity conditions at the Hanford site (our ambient condition), material tended to stay in the moisture level of our feed (below 5 percent weight). However, once the partial pressure of water was greater than 10 torr, these wastes began to absorb water significantly, creating liquid solutions from dry salts. While literature exists to predict the hygroscopic behavior of tank-waste chemicals, various interactions were found to exist that increased these materials' absorbance of water from the air.

3.4 Test Matrix for Clean Glass Feed

3.4.1 Test Plan and Objectives

Once material for ES-31F was completed, testing was proposed to investigate how to process the clean glass feed used at the end of each melt. Figure 3.16 outlines the test matrix used for this study.

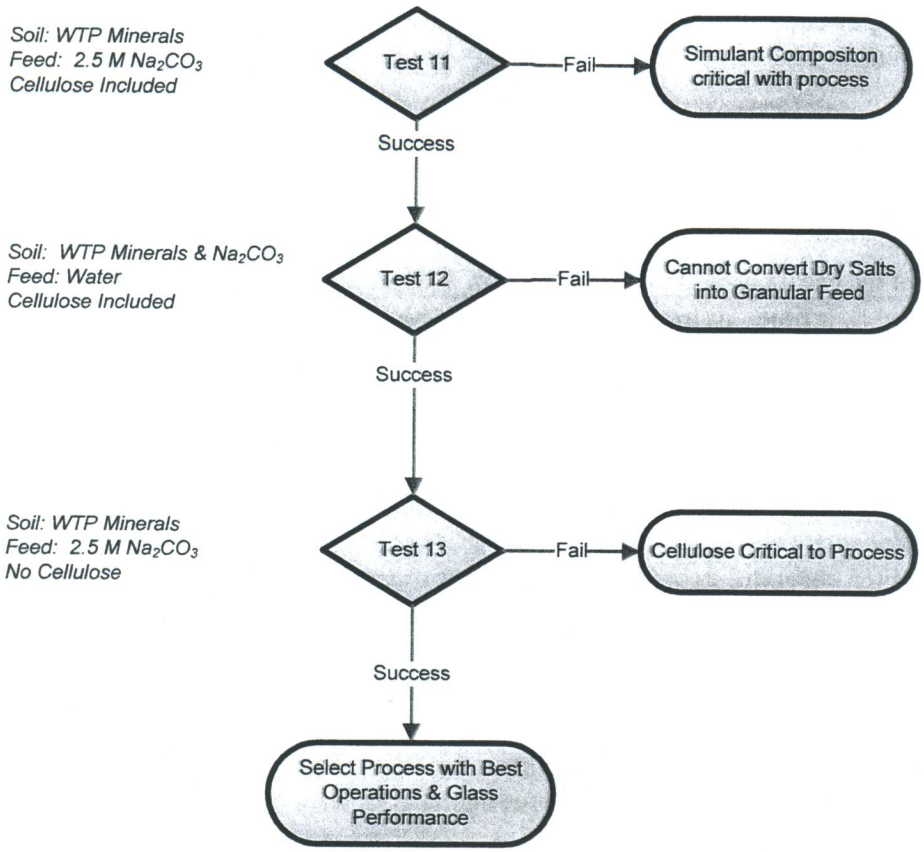

Figure 3.16. Test Matrix for Clean Glass Feed

Overall operations of these batches were as follows:

- Waste Feed rate: 50–70 cc/min
- Startup Batch Size: 8.5–11.5 kg
- Final Batch Size: 12 kg
- Mixer Speed: 128 rpm
- Motor Amperage: 3.4 amperes (Max)
- Shell steam pressure: 20 psig.

3.4.2 Testing Results

Table 3.5 lists the results from the modified test matrix.

Table 3.5. Testing Results of the Modified MIS Mitigation Dryer Test Matrix

Test	Description	Result
Test 11	WTP Minerals and Alpha Cellulose 5-M Na Solution of Sodium Carbonate	Success
Test 12	WTP Minerals and Alpha Cellulose Solid Addition of Sodium Carbonate Water Feed	Success
Test 13	WTP Minerals/No Cellulose 5-M Na Solution of Sodium Carbonate	Success

Success or failure of these tests were determined if the feed produced was similar to that produced by ES-31F and could be easily conveyed.

Tests 11 and 13 processed much like the Test 8 and ES-31F batches, producing a granular product without much difficulty. Even without alpha-cellulose present, the feed from Test 13 appeared to be much like the dryer product for ES-31F. Figure 3.17, Figure 3.18, and Figure 3.19 show the visual results of Test 13 and the particle-size analysis performed.

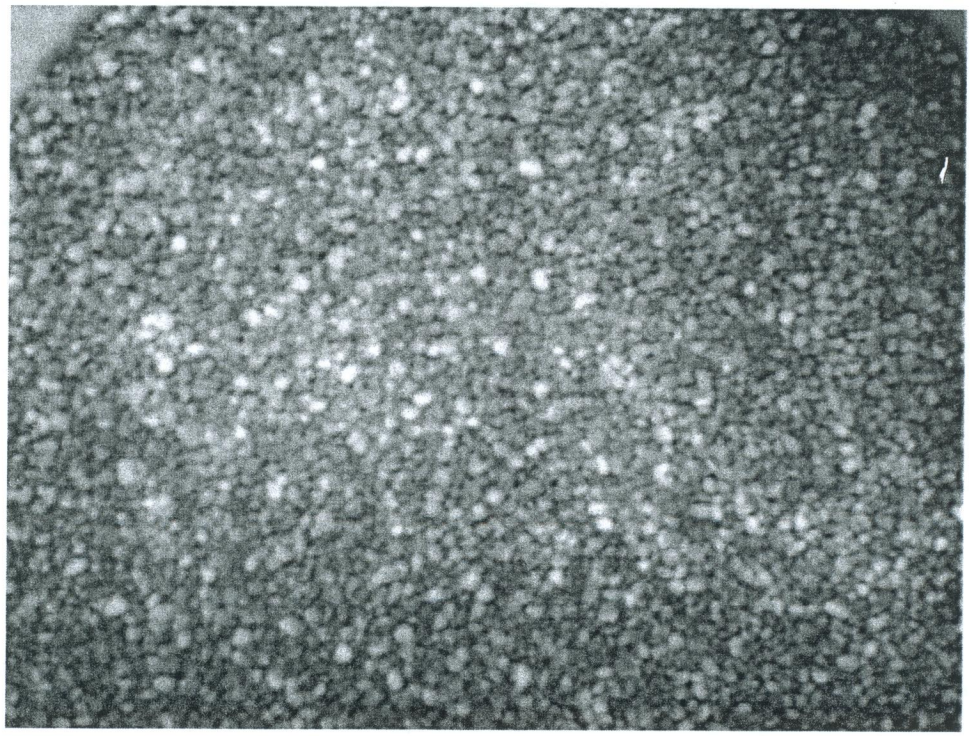

Figure 3.17. Clean Glass Feed Using WTP Glass Minerals and No Cellulose

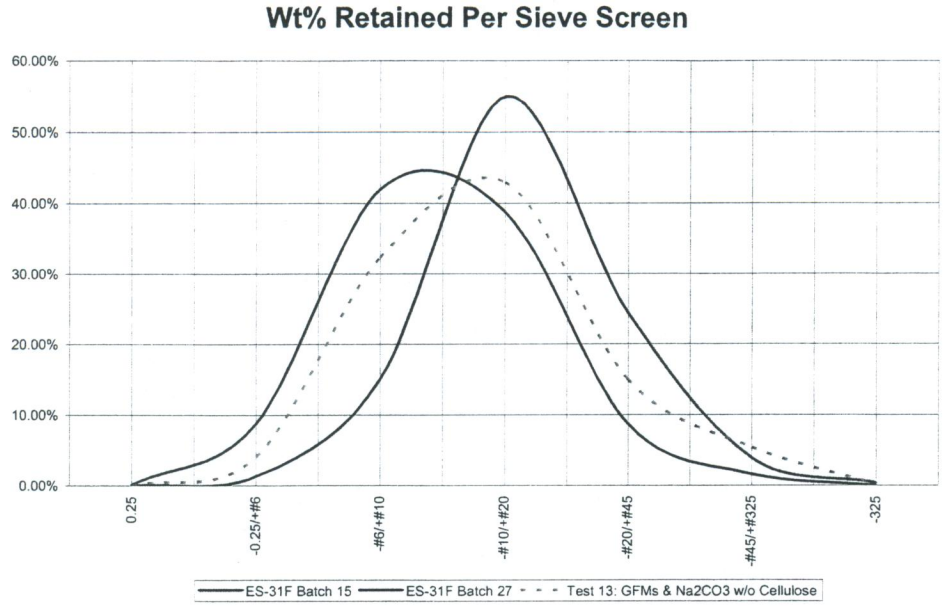

Figure 3.18. Particle Size Distribution of Clean Glass Test

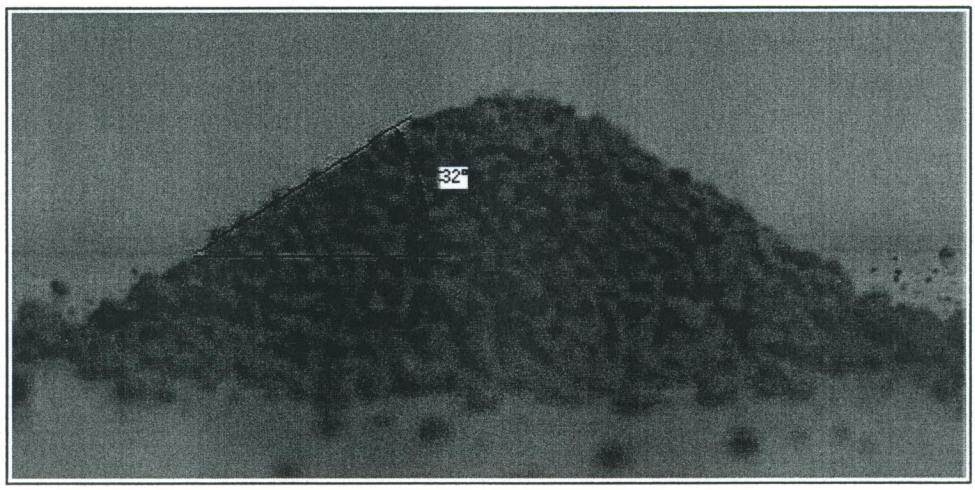

Figure 3.19. Slump Testing Of Test 13

Waste simulant feed rates for all three tests were similar to rates experienced with the melter feed produced with GFMs, despite the fact that the simulant was switched to a sodium carbonate solution for Tests 11 and 13 and to water for Test 12. This implies that the drying process is not significantly affected by changes in dissolved components in the liquid waste stream.

Although Test 12 was considered successful, there were initial difficulties. Adding 3 kilograms of sodium carbonate directly with the feed matrix pushed the start-up mass envelope for this dryer. Like before, the off-gas port had difficulty staying open, and the off-gas filter required frequent filter back-pulsing. During back-pulsing, the motor amperage would jump up to 5 to 6 amperes for a short period of time, and the feed did not become granular as expected, even after increasing the flow rate to 70 cc/min. After the duration of the test extended past the process time of Test 11 (2 hours), the batch was stopped,

and the material was removed. The feed material was examined after 2 hours, and the results were similar to that seen in the second half of the Phase 3 testing report in RPP-RPT-31688.

It was then decided that about $^{2}/_{3}$ of the material would be placed back into the dryer and processed again with water using a feed rate of 60 cc/min. Within 90 minutes, the material in the dryer was granular, and the test was stopped.

While this proved that water alone can produce a granular material, the processing time was the same as Test 11. Also, Test 11 could process more material in a single batch because the sodium carbonate was added gradually while the bulk density of the dryer product increases as it becomes granular. Adding the sodium carbonate as a dry material did not appear to have any major advantages.

The results from Test 12 also indicate that overfilling the dryer may interfere with pellet formation. The jumps in the motor amperes may indicate that too much mass in the dryer can increase the shear forces and prevent pellet formation of an agglomerated granular product. This issue is a problem that was created with the introduction of cellulose because it significantly reduces the bulk density of the charge material unless it becomes incorporated with the GFMs. This could be a problem for bleed and feed operations as well if the cellulose has not had time to be incorporated into the feed. Batch operations show that the GFMs and cellulose change form between 60 to 90 minutes. Bleed-and-feed operations need to account for this process time between exchanges to verify that the maximum working volume in the dryer is not exceeded. Unfortunately, the working volume of larger scale dryers will not necessarily be scaleable to the 22-liter dryer.

Test 13 is also of interest because while it did not have cellulose present in the feed matrix, it still produced melter feed with the same physical characteristics as melter feed produced from Test 8 and ES-31F. This implies that the GFMs, not cellulose, may be the reason for the size increase of the melter-feed particles witnessed for tests using both GFMs and cellulose. In every case, melter feed produced using GFMs appears to have a higher absorption potential of water than the standard HRTS. This is understandable because the GFMs also have a higher retention of MIS during the melting operation than HRTS soil. Comparing the baseline process to the feed produced using GFMs shows a correlation between moisture level present in the melter feed and mean particle size. This implies that water is the primary binding agent in the dryer and that using material with higher absorption rates of water will increase the mean particle size of the feed.

4.0 Conclusions

- Chemical differences in the liquid waste simulants tested (six-tank composite, different S-109 simulants, and blends of different simulants) had no impact on dryer performance or on the final physical characteristics of the feed. This was also true when waste simulant was replaced with a sodium carbonate solution while preparing clean glass melter feed.
- A change in the zirconium mineral from zirconium oxide to zircon had no adverse impact to dryer performance and the final product.
- Adding boron oxide and zircon to the initial dryer load had no impact on dryer performance.
- Reducing the average particle size of the soil/glass mineral matrix was not detrimental to the dryer performance.
- Using sucrose as a feed component in drying operations is not recommended due to the soluble nature of sucrose and the moist environment inside the dryer, causing the feed material to stick to the side walls and/or set up into a solid mass.
- Using alpha cellulose as a feed component in drying operations was not detrimental to dryer performance.
- Changes in the feed matrix increased the average waste flow and overall drying rate by 50 percent and doubled the moisture level present in the feed and final dryer product.
- Reducing the average particle size of glass minerals appeared to increase the final agglomerate size of the feed material by increasing the moisture level of the material.
- Cellulose had the effect of decreasing the bulk density of the starting material in the dryer from 1.0 to 1.2 gm/cm^3 to 0.5 to 0.6 gm/cm^3, decreasing the batch capacity of the dryer. However, higher processing rates combined with a semi-continuous operation can actually increase the dryer throughput rates.
- Over-filling the dryer with material can reduce dryer fluidization and increase the shear forces between particles, reducing its effectiveness to produce a granular feed product. During 22-liter testing, the maximum working volume of the dryer was approximately 67 percent.
- The 22-liter dryer tests produced similar results to what was experienced in the 130-liter dryer tests described in RPP-RPT-31688, *Supplemental 130L Scale Dryer Development Test Report.*
    - A continuous liquid-waste feed rate was used by both dryers during operations, and the internal drying temperature was found to remain stable throughout the process.
    - Initial charge masses and simulant waste feed rates were scaleable when processing the baseline feed product and when processing melter feed using GFMs and cellulose. When switching to GFMs, the same relative increase in feed rate was experienced.
    - Similar operations and temperatures were found during processing with the 130-liter dryer (140 to 150°F) running slightly cooler than the 22-liter dryer (150 to 170°F). This may be a scaling issue with the 22-liter dryer, or it may be because the superior off-gas system used with the 130-liter dryer could achieve better vacuum levels.

- Melter feed generated from GFMs and cellulose was found to be granular in nature when the moisture level was found to be roughly between 2 to 5 percent weight, with mean diameters greater than 840 microns. This compares to the baseline product, whose mean diameter exists between 300 to 500 microns.

- Both systems experienced an upset condition while using GFMs and cellulose where material did not become granular. Even with higher feed rates, the dryer charge stayed a powder and did not behave like other tests.
  - In the 130-liter tests, this occurred at the end of bleed-and-feed operations where the maximum waste simulant feed rate was being evaluated.
  - The 22-liter dryer experienced this when trying to evaluate different methods of trying to produce a clean batch feed and optimize the charge size. In the case of the 22-liter dryer, the problem was resolved by decreasing the input charge size to improve fluidization of the dryer.
- An aging study of feed from ES-31F confirmed past suspicions about the hygroscopic nature of the simulant salts in the feed. While no significant water-absorption effect is expected at typical Hanford conditions (P_{H2O} = 5 torr), melter feed material aged in a 100 percent relative humidity environment at room temperature (P_{H2O} = 18 torr) for 200 hours eventually absorbed enough water to break down the feed material into mud.
- The 22-liter dryer produced a desirable product for the second half of the test for ES-31F while operating in the following range:
  - Initial charge mass of 7.5 kg, producing a final batch size of 12.9 kg
  - Shell steam pressures of 18 to 20 psig (256 to 260°F)
  - Mixer speeds between 120 to 218 rpm.
  - Vacuum pressures of 20 to 22 in. Hg
  - Internal dryer temperature between 150 to 170°F
  - Simulant feed rates of 50 to 70 cc/min (62 to 88 gm/min), which produced a product with moisture levels between 1.7 to 4.8 wt percent.

5.0 References

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