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Crystal Accumulation in the Hanford Waste Treatment Plant High Level Waste Melter: Summary of 2017 Experiments

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January 2018

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EXECUTIVE SUMMARY

A full-scale, transparent mock-up of the Hanford Tank Waste Treatment and Immobilization Project High Level Waste glass melter riser and pour spout has been constructed to allow for testing with visual feedback of particle settling, accumulation, and resuspension when operating with a controlled fraction of crystals in the glass melt. Room temperature operation with silicone oil and magnetite particles simulating molten glass and spinel crystals, respectively, allows for direct observation of flow patterns and settling patterns. The fluid and particle mixture is recycled within the system for each test. In this report, use of the system for three objectives is described:

- Develop a better understanding of repeatability of the results provided by the system
- Determine whether the bulk density of the fluid and particle mixture in the feed tank influenced settling behavior in the throat during idle periods
- Determine whether lowering the height of the air lance would be useful in resuspending the layer of settled particles at the bottom of the riser.

In general, changes in the thickness of the accumulated layer at the bottom of the riser from the start to the finish of each pouring cycle and each idling cycle were negligible, and therefore repeatable. Small changes were not observable after an individual run of the system, but did have a cumulative effect over time. The degree of resuspension of particles in the throat during each pouring cycle lessened as the experiments progressed, indicating a lack of repeatability. This may have been due to settling or compaction over time, making particle resuspension more difficult. Other potential causes include repeated use of the solids within the system, curvature of the cylindrical throat, and compounding measurement uncertainty. The rate of particle accumulation in the throat during each idle period appeared to approach a consistent value as the experiments progressed.

The feed tank agitator was left running during the idle period for a series of experiments to determine whether the bulk density of material in the feed tank influences flow and settling patterns in the throat. There were no visible differences in particle flow patterns when compared to recordings of earlier experiments where the feed tank agitator was turned off during idling.

Reducing the height of the air lance in the riser appeared to redistribute the accumulated particles at the bottom of the riser, but did not appear to resuspend the particles.

Future work will include lowering the air lance further to determine whether particles accumulated in the riser can be resuspended; measuring particle size distributions of the accumulated particles; intentionally increasing the thickness of the accumulated layers to observe impacts to pouring and resuspension; and adding neutrally buoyant tracer particles to support quantification of the flow patterns and fluid dynamic modeling.

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LIST OF ABBREVIATIONS

DOE	U.S. Department of Energy
HLW	High-Level Waste
LAW	Low-Activity Waste
ORP	Office of River Protection
$T_{1\%}$	Temperature where one volume percent of spinel crystals are present in a glass melt
vol %	Volume percent
WTP	Hanford Tank Waste Treatment and Immobilization Plant

1.0 Introduction

The U.S. Department of Energy (DOE) Office of River Protection (ORP) is building the Tank Waste Treatment and Immobilization Plant (WTP) at the Hanford Site in Washington to remediate 56 million gallons of radioactive waste that is being temporarily stored in 177 underground tanks. Radioactive waste will be separated into high-level waste (HLW) and low-activity waste (LAW) fractions that will be vitrified in stable borosilicate glass with Joule-heated, ceramic refractory lined melters. Efforts are being made to increase the loading of Hanford tank wastes in glass while maintaining an adequate ability to meet process, regulatory, and product quality requirements.

Glass formulation and melter testing data have suggested that significant increases in waste loading in HLW and LAW glasses are possible over current system planning estimates.[1] Belsher and Meinert identified five constraints that were most influential on the estimated Hanford HLW glass volumes,[2] and by extension, most restricting to waste loading. One of those constraints was the limit of no more than one volume percent spinel crystals in the melt ($T_{1\%}$) at a temperature of 950 °C.

Historically, crystallization constraints are placed in process control systems to prevent premature or catastrophic failure of the melter through bulk devitrification (also described as volume crystallization) or crystal accumulation and, thus, to mitigate negative impacts of crystals as glass is produced.[a] The baseline method of controlling crystallization in the WTP HLW melter uses a model that predicts the temperature, $T_{1\%}$, at which the equilibrium fraction of spinel crystals in the melt is 1 volume percent (vol %).[4] An alternative crystal-tolerant glass approach[5] may allow higher waste loading for WTP processing while maintaining a chemically durable glass product. Some crystalline phases, such as spinel, do not impact the durability of the waste form[6] but may accumulate in the melter or riser and restrict or prevent its operation. However, prediction of spinel precipitation and accumulation could potentially allow for formulating higher waste loading, durable glasses if an alternative strategy for operating and idling a melter with some amount of tolerable crystals can be developed and implemented.

Given the identification of the $T_{1\%}$ constraint as one of the most influential constraints for estimated Hanford HLW glass volumes, ORP has initiated a program to evaluate whether this constraint can be relaxed or whether new constraints could be developed to replace the current $T_{1\%}$ approach.[7,8] A road map was developed to guide research and development efforts for a crystal tolerant glass processing strategy for WTP.[9] The basis of this potential, alternative approach will be an empirical model predicting the crystal accumulation in the WTP glass discharge riser and melter bottom as a function of glass composition, time, and temperature.[5,10] When coupled with an associated operating limit, this model could then be integrated into the process control algorithms to formulate crystal tolerant HLW glasses targeting higher waste loadings while still meeting other process related limits and melter lifetime expectancies.

Actual melter operation is likely to involve situations where accumulation of spinel crystals can occur. Unexpected events may hamper the use of a crystal accumulation process control model. Methods of recovering from such an event will make the crystal tolerant approach more robust, and allow for continued use of a melter in the event of excessive crystal accumulation.

To better understand crystal settling, accumulation, and resuspension in critical areas of the WTP HLW melter, a full-scale, room temperature test system has been designed and constructed.[11] The road map for development of crystal-tolerant HLW glasses noted that an accumulation of crystals in the melter riser could prevent discharge of the molten glass into canisters, especially when considering frequent and periodic idling.[9] Therefore, the test system focuses on the throat and riser of the WTP HLW melter. The system uses transparent materials to allow for the observation of particle behavior under a variety of process

[a] Jantzen and Brown provide a brief review of the potential, negative effects of crystallization within a melter.[3]

conditions. Data collected will support the development and implementation of a crystal accumulation process control model. The system will also be used to develop and demonstrate potential methods for recovery in the event of an unacceptable amount of crystal accumulation.

A series of experiments was completed with the room temperature system in fiscal year 2016.[12] In those experiments, a prototypic pour rate was maintained based on the volumetric flow rate. Accumulation of particles was observed at the bottom of the riser and along the bottom of the throat after each experiment. Measurements of the accumulated layer thicknesses showed that the settled particles at the bottom of the riser did not vary in thickness during pouring cycles or idle periods. Some of the settled particles at the bottom of the throat were re-suspended during subsequent pouring cycles, and settled back to approximately the same thickness after each idle period. The cause of the consistency of the accumulated layer thicknesses was not year clear, but was hypothesized to be related to particle flow back to the feed tank. Additional experiments reinforced the observation of particle flow along a considerable portion of the throat during idle periods.

This report describes the results of experiments completed in 2017 with the full-scale, room temperature WTP HLW melter riser system. Observations from the experiments are provided and discussed, and recommendations are made for future testing. This work was performed following a Task Technical and Quality Assurance Plan,[13] as well as an experimental plan for 2018 studies.[14]

2.0 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Savannah River Site Manual E7, Procedure 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2. Laboratory data for this study were recorded in the SRNL Electronic Laboratory Notebook system, experiment L0008-00162.

3.0 Experimental Runs

3.1 Test Conditions and System Operation

The design, materials selection, and operation of the full-scale, room temperature WTP HLW melter riser test system have been described in an earlier report.[11] Three sets of experimental runs were completed in 2017 using the system:

- The first set of experiments was run to develop a better understanding of repeatability of the results provided by the system. Test conditions used in 2016 were repeated, four times at the high flow rate and three times at the low flow rate.
- The second set of experiments was run with the feed tank agitator left on during the idle periods. The objective was to determine whether the bulk density of the fluid and particle mixture in the feed tank influenced settling behavior in the throat during idle periods.
- The third set of experiments was run to determine whether lowering the height of the air lance would be useful in resuspending the layer of settled particles at the bottom of the riser.

The solids were allowed to settle for at least 40 hours (and typically longer) between each of the experimental runs. A particle loading of 0.1 vol % in the fluid was selected and used to maintain visibility. A nominal pouring rate of 3.18 lpm (0.84 gpm) was selected and used as the high rate for testing. This pouring rate is the volume of glass planned to be poured per unit time in the actual melter. The rate was calculated using a nominal WTP HLW melter pour rate of 520 lbs of glass in a period of 29 minutes (8.13

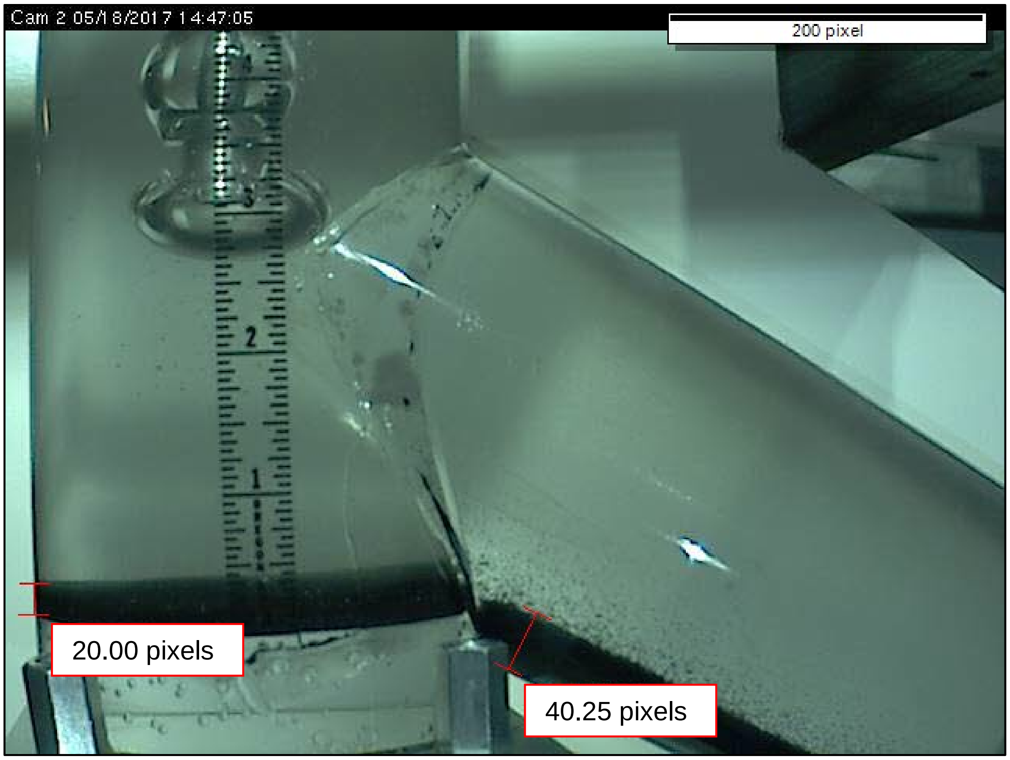
kg/min),[15,a] and an arbitrary glass density of 2.56 g/cm^3. A reduced, or low pouring rate was also selected to reflect potential changes in operation of the WTP HLW melter that might impact particle re-suspension. The low pouring rate was set at approximately 2/3 of the high rate (2.3 lpm or 0.60 gpm).

During idle periods, a continuous purge (approximately 0.2 scfh) of the air lance in the riser was maintained to better simulate planned operation of the WTP HLW melter. The idle purge was observed to produce a complex particle and fluid flow pattern in the riser during idle periods.

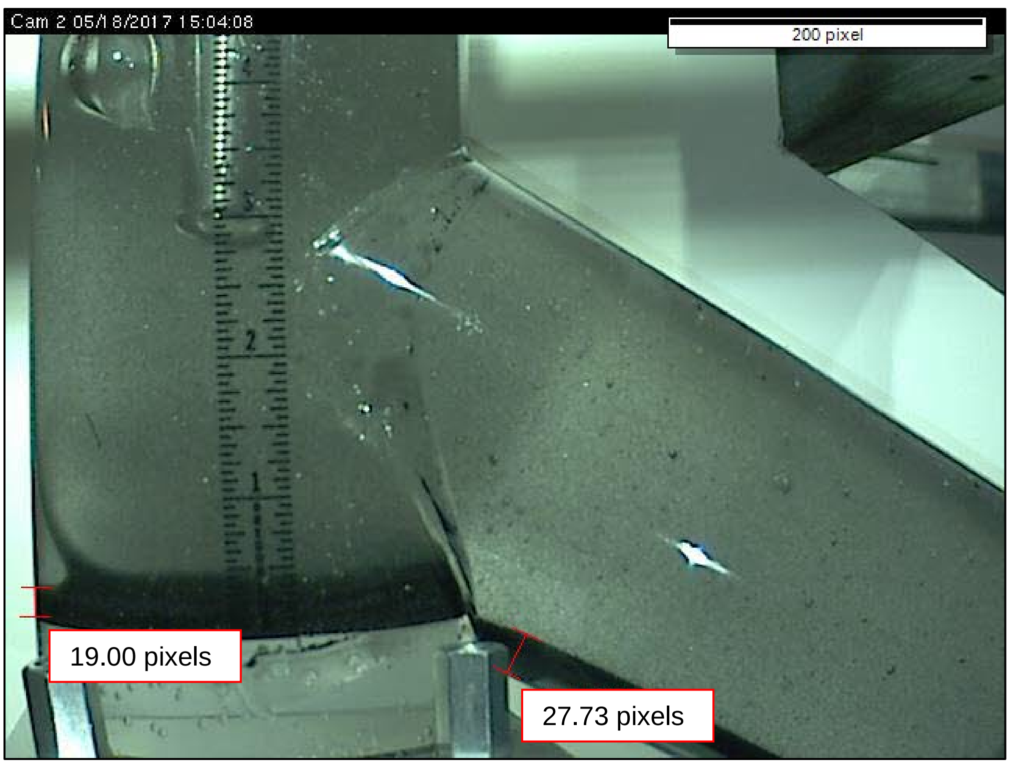
The ambient temperature in the laboratory varied from day to day, resulting in changes to the viscosity of the silicone oil. Qualitatively, this did not appear to impact particle settling behavior among the experiments. The temperature of the fluid during each experiment was recorded to allow for calculation[11] of the viscosity of the silicone oil.

Changes in thickness of the accumulated particle layers during pouring and idle periods were measured using still images from the video recordings. Images of the junction between the throat and the riser were used for reference since the position of the camera was not changed during the series of experiments. Image analysis software was used to measure the thickness of the accumulated layers at the bottom of the riser and throat before and after the pouring cycles, and at the beginning and end of the idle periods. Example measurements are shown in Figure 3-1 for the beginning and end of a pouring cycle from one of the high flow rate tests. The thickness of the accumulated layer at the bottom of the riser was measured from a reference point on the bottom left of the riser, as shown in the figure. In this example, the difference in the measurements at the beginning (20 pixels) and end of the pouring cycle (19 pixels) is considered insignificant, as the resolution of the measurements is no less than one pixel. The thickness of the accumulated layer at the bottom of the throat was measured from a reference point where the bottom of the throat visually intersects one of the stand-offs supporting the riser, as shown at the bottom center of the images in Figure 3-1. The change in accumulated thickness at this location was considered significant (40.25 pixels versus 27.73 pixels). Similar measurements were made for each of the test runs in this report.

[a] Note that the production rate of 4 MT/day given in Reference 15 is higher than the design capacity production rate of 3 MT/day given in the IHLW Waste Form Qualification Report.[16.] The higher production rate, and therefore higher pouring rate, was used in this study and considered to be an upper bound.



(a)



(b)

Figure 3-1. Example accumulated layer thickness measurements at the beginning (a) and end (b) of a pouring cycle

3.2 Repeatability Testing

A total of 17 pouring and settling experiments were completed at two flow rates using the full-scale, room temperature WTP HLW melter riser test system. Measurements from these experiments are summarized in Table 3-1 and Table 3-2. Note that the data from experiments 1-4 were included in the 2016 report.[12] Also note that experiments 11-17 were conducted with the feed tank agitator left running during the idle period. Additional observations from experiments 11-17 related to particle flow with the feed tank agitator running are included in the next section. The full set of experiments is included here for discussion of repeatability of the results.

Table 3-1 provides thickness measurements of the accumulated layers in the riser and throat at the start and finish of each pouring cycle. The percent change is calculated to support comparisons among the experiments. In general, changes in the thickness of the accumulated layer at the bottom of the riser from the start to the finish of each pouring cycle were negligible, and therefore repeatable. The exception was Experiment 5, where a significant increase in the thickness of the accumulated layer occurred. This is due to unusual uncertainty in the measurement of the layer thickness after the pouring cycle, as shown in Figure 3-2. Suspended particles in the area of the measurement made identification of the settled layer difficult; thus, this data point will be disregarded.

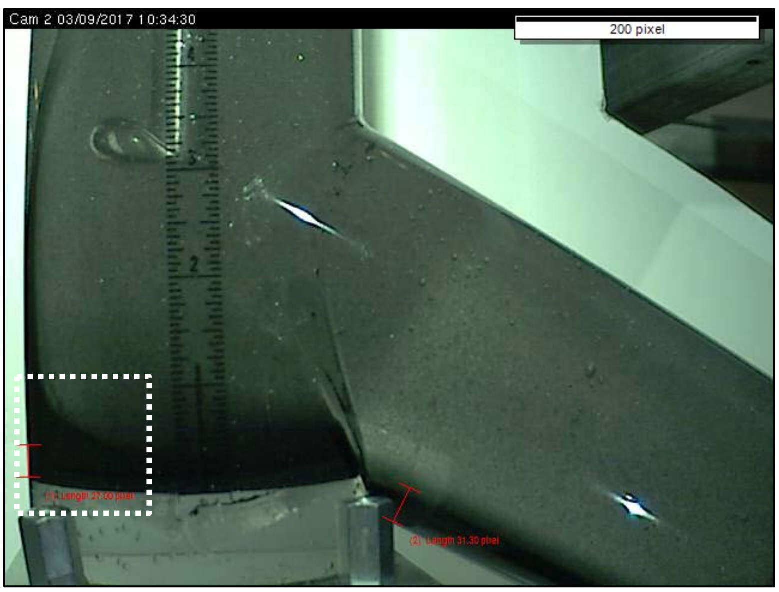


Figure 3-2. Obfuscation of the settled layer thickness measurement after pouring Experiment 5

Table 3-1. Accumulated Layer Measurements for the Pouring Cycles

Particle Settling Experiment	Targeted Flow Rate (gpm)	Average Actual Flow Rate (gpm)	Fluid Temperature (°C)	Start Time	Finish Time	Riser Layer Thickness			Throat Layer Thickness		
						Start (pixels)	Finish (pixels)	Change	Start (pixels)	Finish (pixels)	Change
1	0.84	0.84	16.7	2/10/2016 10:19:05	2/10/2016 10:36:06	11	10	-9%	29.79	20.12	-32%
2	0.84	0.98	24.8	4/25/2016 09:38:27	4/25/2016 09:52:02	15	14	-7%	32.65	23.71	-27%
3	0.84	0.99	26.0	4/27/2016 12:58:41	4/27/2016 13:14:15	15	14	-7%	33.54	22.36	-33%
4	0.60	0.65	21.3	5/5/2016 10:05:18	5/5/2016 10:25:27	15	16	7%	31.3	25.5	-19%
5	0.60	0.64	23.2	3/9/2017 10:12:36	3/9/2017 10:34:11	16	23	44%	44.72	31.3	-30%
6	0.60	0.59	19.5	3/13/2017 12:53:22	3/13/2017 13:16:33	20	20	0%	40.25	27.73	-31%
7	0.84	0.91	26.1	5/10/2017 13:27:40	5/10/2017 13:42:56	23	20	-13%	41.15	30.41	-26%
8	0.84	0.83	23.9	5/18/2017 14:47:05	5/18/2017 15:04:08	20	19	-5%	40.25	27.73	-31%
9	0.60	0.58	23.0	5/23/2017 14:21:18	5/23/2017 14:45:10	20	20	0%	38.01	29.07	-24%
10	0.84	0.65	23.0	6/1/2017 9:04:45	6/1/2017 9:28:30	21	21	0%	40.25	29.97	-26%
11	0.60	0.57	22.0	6/19/2017 10:19:56	6/19/2017 10:43:00	20	22	10%	39.36	31.3	-20%
12	0.84	0.87	23.5	6/21/2017 10:14:53	6/21/2017 10:30:05	21	21	0%	42.49	34.44	-19%
13	0.84	0.89	22.9	7/24/2017 10:49:35	7/24/2017 11:05:24	22	24	9%	41.59	33.54	-19%
14	0.60	0.58	21.6	7/26/2017 14:53:40	7/26/2017 15:18:20	21	22	5%	43.83	33.54	-23%
15	0.60	0.53	23.5	10/4/2017 9:33:07	10/4/2017 10:00:23	23	22	-4%	43.38	37.12	-14%
16	0.84	0.87	23.2	10/9/2017 9:52:56	10/9/2017 10:09:16	23	23	0%	46.07	37.12	-19%
17	0.84	0.85	22.3	10/11/2017 8:55:18	10/11/2017 9:11:14	22	22	0%	44.72	37.12	-17%

Table 3-2. Accumulated Layer Measurements for the Idle Periods

Particle Settling Experiment	Start Time	Finish Time	Feed Tank Agitator On	Riser Layer Thickness			Throat Layer Thickness			
				Start (pixels)	Finish (pixels)	Change	Start (pixels)	Finish (pixels)	Change	Accumulation Rate (pixels/minute x10^2)
1	2/10/2016 10:35	2/11/2016 02:50	No	11	12	9%	20.12	37.12	84%	1.744
2	4/25/2016 09:52	4/26/2016 05:00	No	15	15	0%	22.36	37.12	66%	1.286
3	4/27/2016 13:13	4/28/2016 11:50	No	15	15	0%	21.02	33.54	60%	0.923
4	5/5/2016 10:25	5/6/2016 04:30	No	16	16	0%	24.6	39.36	60%	1.360
5	3/9/2017 10:34	3/10/2017 9:19	No	27	20	-26%	31.3	34.89	11%	0.263
6	3/13/2017 13:16	3/14/2017 12:49	No	20	19	-5%	27.73	40.25	45%	0.886
7	5/10/2017 13:43	5/11/2017 15:56	No	20	18	-10%	29.07	38.01	31%	0.568
8	5/18/2017 15:04	5/19/2017 14:29	No	20	18	-10%	29.07	40.25	38%	0.796
9	5/23/2017 14:45	5/24/2017 16:12	No	20	20	0%	29.97	42.49	42%	0.820
10	6/1/2017 9:28	6/2/2017 8:05	No	21	22	5%	29.97	41.15	37%	0.824
11	6/19/2017 10:43	6/20/2017 11:20	Yes	21	20	-5%	33.54	42.49	27%	0.606
12	6/21/2017 10:30	6/22/2017 12:42	Yes	22	21	-5%	33.54	42.49	27%	0.569
13	7/24/2017 11:05	7/25/2017 13:02	Yes	25	22	-12%	32.65	42.49	30%	0.632
14	7/26/2017 15:18	7/27/2017 15:47	Yes	22	20	-9%	34.44	43.83	27%	0.639
15	10/4/2017 10:00	10/5/2017 10:39	Yes	24	23	-4%	37.12	45.07	21%	0.538
16	10/9/2017 10:09	10/10/2017 10:16	Yes	22	22	0%	35.78	45.52	27%	0.673
17	10/11/2017 9:11	10/12/2017 10:23	Yes	22	24	9%	37.12	45.52	23%	0.556

As shown in Figure 3-3, thickness measurements of the accumulated layer at the bottom of the riser gradually increased as the experiments progressed. Error bars in this plot reflect an uncertainty of +/- 1 pixel for riser layer thickness measurements, and +/- 2 pixels for throat layer thickness measurements.

Changes in the thickness of the accumulated layer along the bottom of the throat occurred during each pouring cycle, representing some degree of resuspension of the settled particles during each experiment. The data in Table 3-1 again show a gradual increase in the thickness of the accumulated layer in the throat as the experiments progressed. This is also shown graphically in Figure 3-3. The change in throat layer thickness (i.e., amount of particle resuspension in the throat during pouring) was somewhat repeatable among the experiments, with an average reduction of 24% (without consideration of the pouring rates or pouring times) for each pouring cycle.

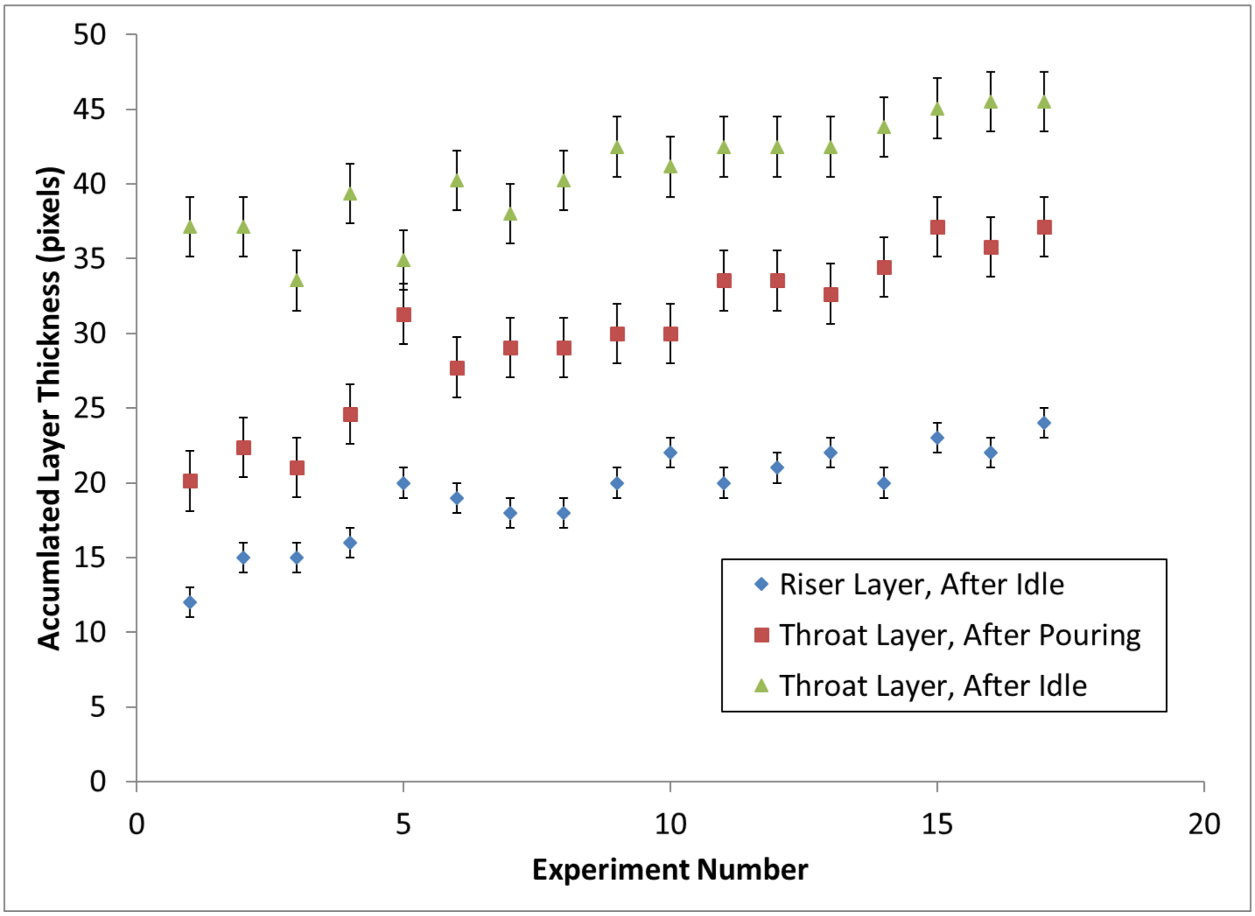


Figure 3-3. Thickness of the accumulated particle layers in the throat and riser for the 17 pouring cycle experiments

Changes in the conditions under which each experiment in Table 3-1 was performed necessitate normalization of the data to allow for more meaningful comparisons of the degree of particle resuspension during a pouring cycle. Likely of most significance are the changes in targeted flow rate for operation of the system, and the differences between the targeted and measured flow rates. As discussed in Section 3.1, the experiments targeted high and low flow rates of 0.80 and 0.60 gpm, respectively. The flow rate of the system is controlled by monitoring the change in mass of material poured as a function of time and manually adjusting the air flow to the lance in the riser to meet the targeted value. Feedback for this control loop occurs on the order of minutes, thus an average flow rate is calculated for each experiment at its completion. These data are included in Table 3-1. The temperature of the laboratory during an experiment, included in Table 3-1, influences the viscosity of the silicone oil in the system. The viscosity of the oil was previously measured as a function of temperature, with the resulting equation provided in an earlier report:

$$\eta = 13.275\, e^{1798.7\left(\frac{1}{T}\right)}$$

where η is the viscosity of the oil (cP) and T is the temperature of the oil (Kelvin).[11] Finally, the length of time that each pouring cycle operated varied somewhat among the experiments, as shown in Table 3-1. An equation was developed to calculate a normalized change in the thickness of the accumulated particle layers during a pouring operation and enable more meaningful comparisons among the experiments:

$$normalized\ change = \frac{\Delta_{accumulated\ layer\ thickness,pixels}}{(flow\ rate, gpm)(pouring\ time, min)(viscosity, cP)} \times 10^{-6}$$

This equation accounts for the actual flow rate during each experiment, the amount of time over which pouring occurred, and the viscosity of the silicone oil. A factor of 10^{-6} is applied for ease of plotting the results.

The calculated, normalized reductions in layer thickness for the settled particles in the throat are plotted as a function of experiment number in Figure 3-4. Although there are a few outliers, the general trend (linear fit) appears to be that less of the settled layer is resuspended as the experiments progressed. This indicates a lack of repeatability in the outcome of the experiments. This behavior is hypothesized to be due to one of two possible mechanisms. First, the accumulated layer in the throat may be settling or compacting over time, making particle resuspension more difficult (i.e., the shear stress imparted by the fluid is no longer sufficient to mobilize the settled particles). Second, the surface area of the accumulated layer in the circular cross section of the throat is increasing as the experiments progress, which may impact the relative change in accumulated layer thickness in that the above equation is not capturing changes in geometry. The second mechanism is considered less likely, as the changes in overall thickness of the accumulated layer in the throat are relatively small; therefore, geometry is not expected to have a significant impact.

Note also that the experiments performed to date have all used the original inventory of solids added to the system. Non-uniform accumulation of particles as a function of size, if occurring, my skew interpretation of the results. As will be mentioned in the discussion of future work (Section 5.0), samples of the accumulated particles have been collected and will be analyzed to determine whether their particle size distribution differs from that of the original system inventory.

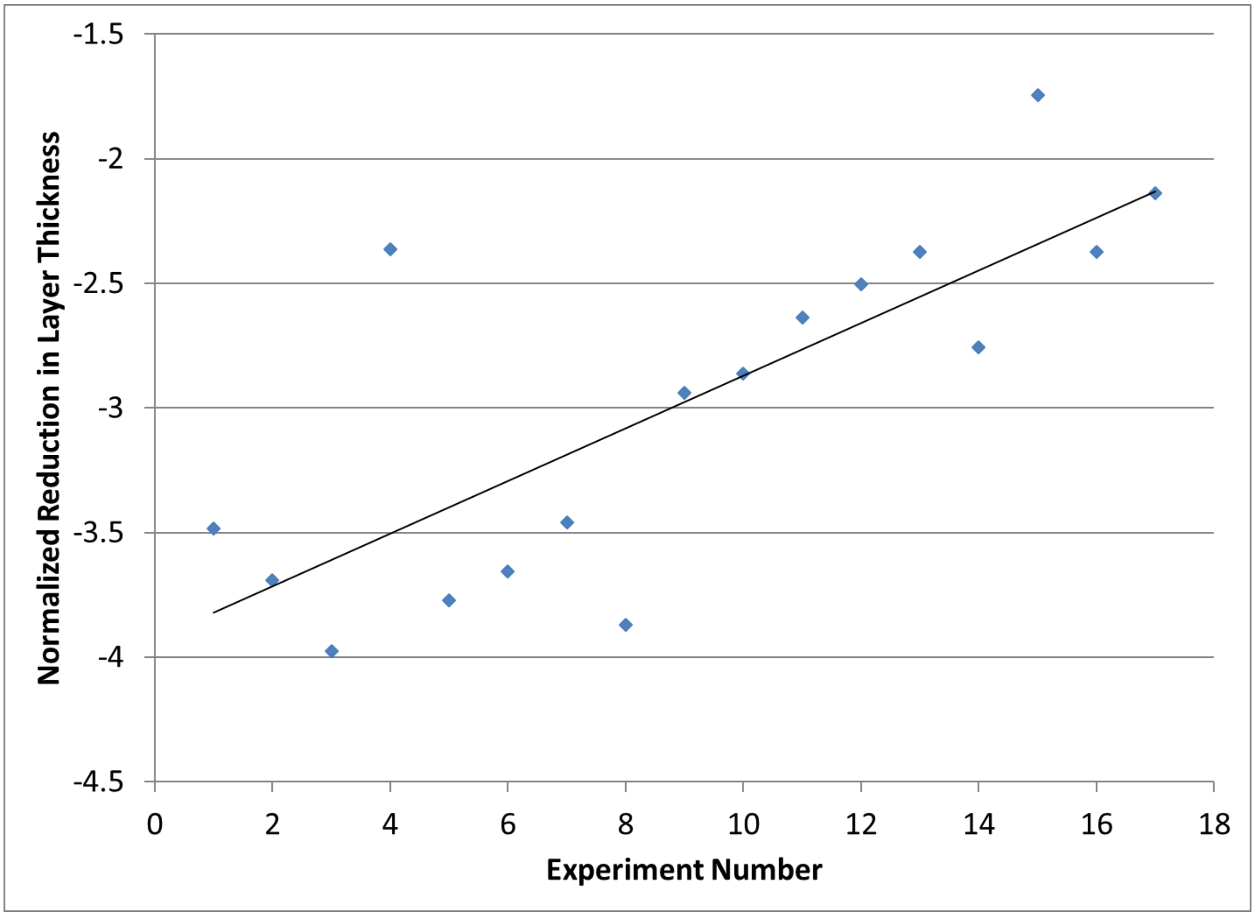


Figure 3-4. Normalized reduction in accumulated layer thickness in the throat for the 17 pouring cycle experiments

Table 3-2 provides thickness measurements of the accumulated layers in the riser and throat at the start and finish of each idle period, corresponding to the ends of the pouring cycles listed in Table 3-1.[a] Again, measured differences in the thickness of the accumulated layer at the bottom of the riser were generally negligible, indicating little if any measurable change between the start and end of each idle period. The exception was Experiment 5, where a reduction in thickness of the settled layer in the riser appeared to occur over the idle period. This again is due to the difficulty in determining the thickness of the layer at the completion of the pouring cycle (refer to Figure 3-2), and therefore this data point will be ignored.

The accumulation rate in the throat was calculated by normalizing the change in layer thickness to the time of the idle period. These data are provided in the right most column of Table 3-2. A plot of these values over the series of experiments is shown in Figure 3-5. The 2-point moving average of the accumulation rates is included on the plot, and appears to show that the accumulation rate is becoming more consistent as the experiments have progressed. Also of note is that the thickness of the settled layer in the throat after each idle period increases by only a small amount after each experiment (Table 3-2 and Figure 3-3). Neither

[a] Note that there were slight differences in timing when transitioning the video recording system from real-time to time-lapse modes, which resulted in small (1-2 pixel) differences in the thickness measurements of the accumulated layer thickness in the throat. Thus, the throat layer thickness measurements at the finish of each pouring cycle do not necessarily match those at the start of the subsequent idle period. These differences were considered insignificant relative to the magnitude of the changes measured over the course of each idle period.

the accumulated layer in the riser nor the accumulated layer in the throat appear to increase in thickness in a step-wise manner with each pouring and settling cycle, as was first anticipated. The reason for this observed behavior is not yet clear.

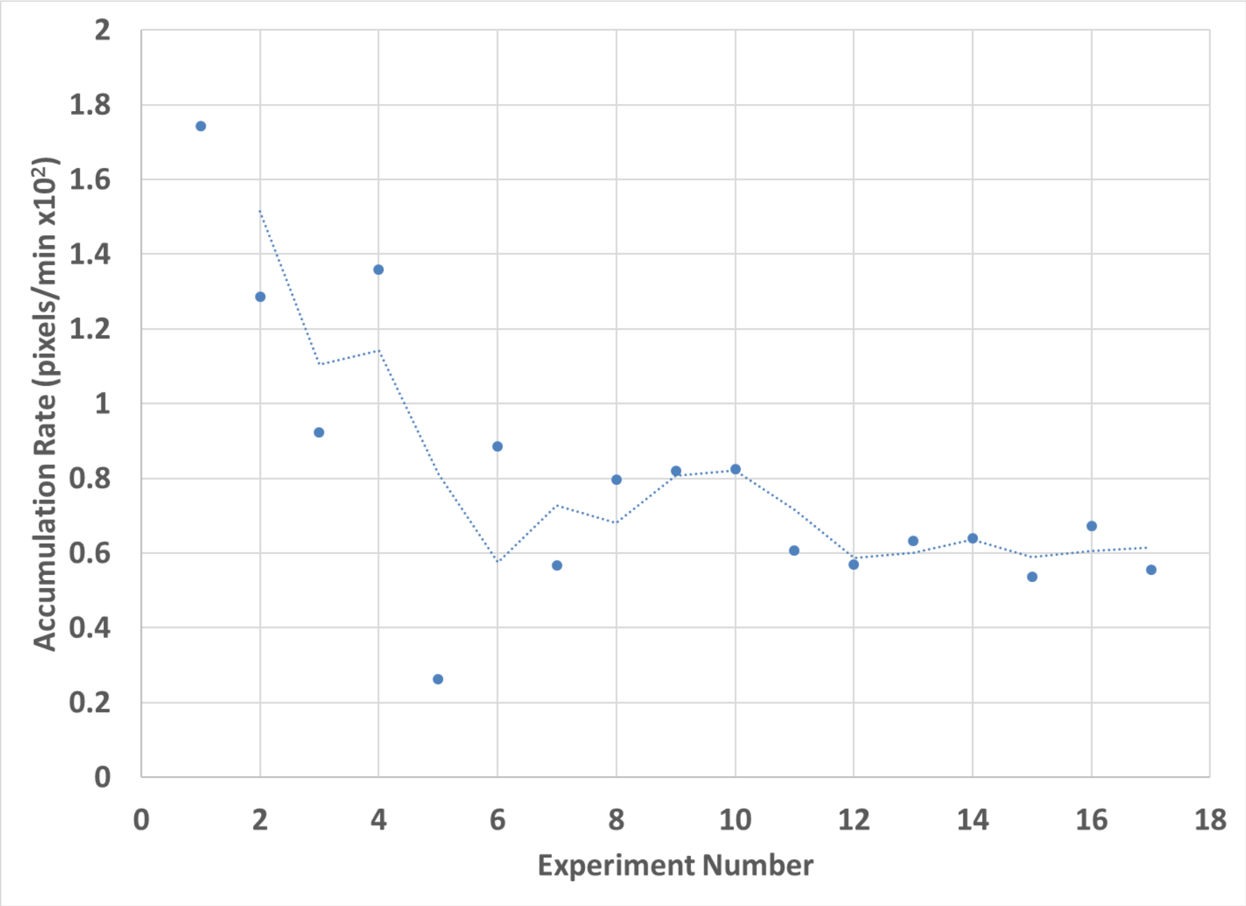


Figure 3-5. Accumulation rate of settled particles in throat and moving average

3.3 <u>Particle Flow During Idle Periods</u>

It was hypothesized that the lack of incremental increases in the thickness of the accumulated layers may be due to flow of the particles through both the riser and throat back to the feed tank during idle periods, as outlined in Figure 3-6. A review of the time-lapse recordings from earlier experiments identified flow of the suspended particles through both the riser and throat toward the feed tank, as well as recirculation of the suspended particles in the opposite direction. Settling of particles in the feed tank may result in a lower bulk density of the particle and oil mixture, providing some driving force for the particles to move from the riser and throat back to the feed tank. To test this hypothesis, a series of experiments was run where the feed tank agitator was left running during the idle period. This kept the particles suspended in the feed tank to avoid the potential reduction in bulk density.

Experiments 11 through 17 in Table 3-1 and Table 3-2 were run with the feed tank agitator left running during the idle period. Reviews of the time lapse video recordings of the idle periods for these experiments did not show any visible differences in particle flow patterns when compared to recordings of the experiments where the feed tank agitator was turned off after pouring. The data in Table 3-2 do not show any obvious differences between the experiments run with and without the feed tank agitator left on during the idle periods. Additional work is needed to identify the mechanisms controlling the flow patterns within the system during idling, potentially with the assistance of fluid dynamic modeling.

Figure 3-6. Hypothesized path of particle flow during idle periods

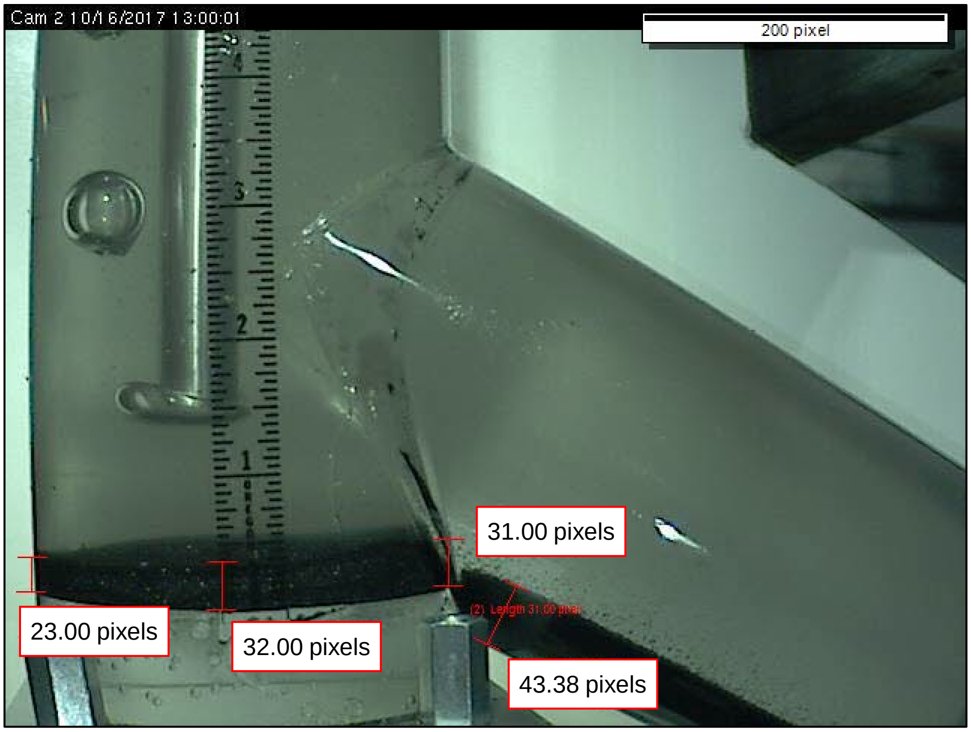
3.4 Air Lance Height Testing

A series of experiments was run to determine whether lowering the height of the air lance would be useful in resuspending the layer of settled particles at the bottom of the riser. The air lance was lowered from the nominal height of 3.25 inches above the bottom of the riser to 1.5 inches above the bottom of the riser. The idle air purge did not have an observable impact on the accumulated layer with the lower position of the air lance. Three pouring and idling experiments were run in this configuration.

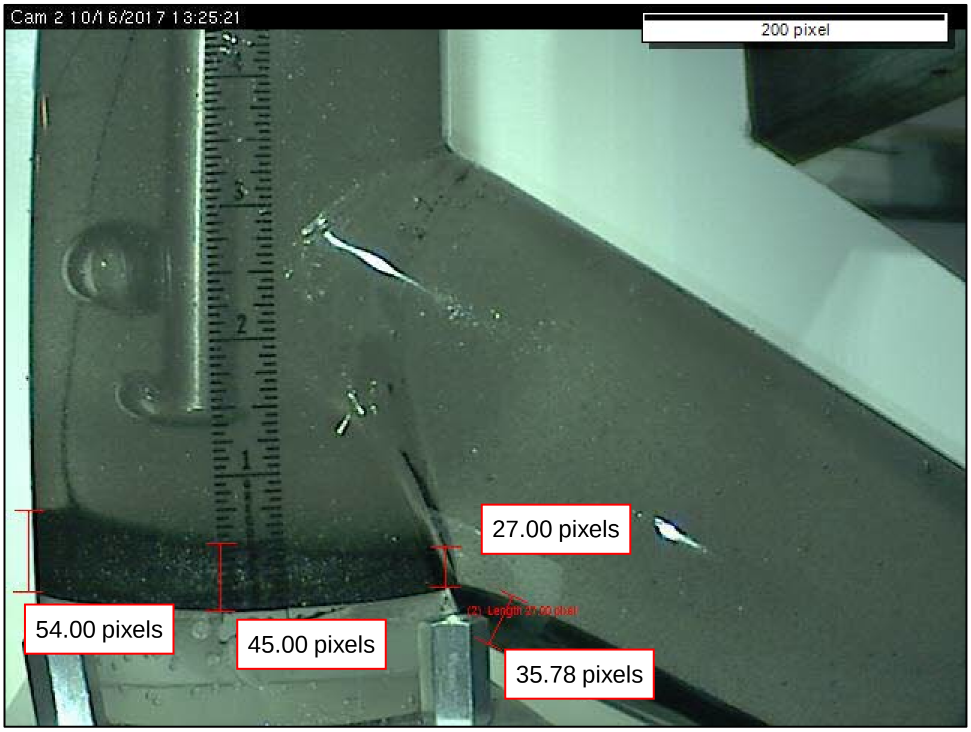
Operation at the high pouring rate (targeting 0.84 gpm) resulted in redistribution of the settled material at the bottom of the riser, as shown in Figure 3-7, where the accumulated particles have shifted to the left in the photograph. Because of this, an average of three measurements taken at the locations shown in Figure 3-7 was used to represent the thickness of the accumulated layer in the riser after lowering the air lance. The redistribution of the accumulated layer occurred over the course of minutes. A review of the real time video recordings showed no obvious resuspension of the accumulated particles at the bottom of the riser as a result of lowering the air lance.

Table 3-3 provides thickness measurements of the accumulated layers in the riser and throat at the start and finish of each pouring cycle with the air lance lowered. Table 3-4 provides thickness measurements of the accumulated layers in the riser and throat at the start and finish of each idle period, corresponding to the ends of the pouring cycles listed in Table 3-3. A review of these tables provides further insight into the experiments with the air lance lowered. The riser layer thickness is larger after each pouring cycle, particularly after the first experiment. This is posited to be the result of the accumulated particles being pushed from the center of the riser bottom out toward its circumference. Some degree of settling then appears to occur in the riser during the idle periods. Restarting the pouring cycle again pushes the settled particles back out toward the circumference of the riser, resulting in a measured increase in the thickness of the accumulated layer. Optical effects of the round tube prevented visual confirmation of this effect. There was no obvious resuspension of the accumulated particles in the riser with the air lance lowered as the thickness of the accumulated layer in the riser returned to approximately the same value after each pouring cycle.

The accumulated particles in the throat show consistent resuspension during pouring cycles followed by settling to about the same height during idle periods. There was no obvious impact of reducing the height of the air lance on the behavior of the particles in this region. Note that the calculated accumulation rate in the throat during idle periods was lower for the experiments with the lower air lance, as compared to the earlier experiments (see Table 3-2). The cause of this is yet not known.



(a)



(b)

Figure 3-7. Lowering the air lance height in the riser redistributed the settled layer of particles. The distribution of particles at the bottom of the riser and associated measurements are shown before (a) and after the pouring cycle (b).

Table 3-3. Accumulated Layer Measurements for the Pouring Cycles with the Air Lance Lowered

Particle Settling Experiment	Targeted Flow Rate (gpm)	Average Actual Flow Rate (gpm)	Fluid Temperature (°C)	Start Time	Finish Time	Riser Layer Thickness			Throat Layer Thickness		
						Start (pixels)	Finish (pixels)	Change	Start (pixels)	Finish (pixels)	Change
18	0.84	0.87	22.3	10/16/2017 13:09:00	10/16/2017 13:25:21	28.67	42	47%	43.38	35.78	-18%
19	0.84	0.81	20.0	10/18/2017 09:05:15	10/18/2017 09:22:00	34.33	40.67	18%	43.38	35.78	-18%
20	0.84	0.85	23.9	10/23/2017 09:13:53	10/23/2017 09:29:51	36	43.33	20%	43.38	35.78	-18%

Table 3-4. Accumulated Layer Measurements for the Idle Periods with the Air Lance Lowered

Particle Settling Experiment	Start Time	Finish Time	Riser Layer Thickness			Throat Layer Thickness			
			Start (pixels)	Finish (pixels)	Change	Start (pixels)	Finish (pixels)	Change	Accumulation Rate (pixels/minute x10^2)
18	10/16/2017 13:26	10/17/2017 12:38	40.33	33.67	-17%	35.78	40.25	12%	0.321
19	10/18/2017 09:22	10/19/2017 10:37	40.67	34.67	-15%	35.78	40.25	12%	0.295
20	10/23/2017 09:30	10/24/2017 09:35	39.67	36	-9%	38.01	41.15	8%	0.217

4.0 Summary

Three sets of experimental runs were completed in 2017 using the full-scale, room temperature WTP HLW melter riser test system:

- The first set of experiments was run to develop a better understanding of repeatability of the results provided by the system. Test conditions used in 2016 were repeated, four times at the high flow rate and three times at the low flow rate.
- The second set of experiments was run with the feed tank agitator running during the idle periods. The objective was to determine whether the bulk density of the fluid and particle mixture in the feed tank influenced settling behavior in the throat during idle periods.
- The third set of experiments was run to determine whether lowering the height of the air lance would be useful in resuspending the layer of settled particles at the bottom of the riser.

The accumulation and resuspension of particles in the riser was shown to be repeatable over the short term, mainly because the change in thickness of the accumulated layer was negligible in each pouring and idling cycle. A longer term view over multiple experiments showed a gradual increase in thickness of the accumulated layer. There was a slight increase in the thickness of the accumulated layer in the riser as the experiments progressed. The degree of resuspension of particles in the throat during a pouring cycle (when the data were normalized to account for flow rate, pouring time, and oil viscosity) was reduced as the experiments progressed. This is hypothesized to be due to settling or compacting of the accumulated layer over time. The accumulation rate of particles in the throat became more consistent as the experiments progressed. Note that the observed changes in the accumulated layers are based on height measurements alone; future observations based on mass and exposed area of the accumulated layers may yield further insight.

Operation of the system with the feed tank agitator left running during the idle periods was unsuccessful in explaining the lack of incremental increases in accumulated layer thicknesses after each pouring cycle. Lowering the air lance in the riser redistributed the accumulated particles at the bottom of the riser, but did not appear to resuspend the particles.

5.0 Future Work

Topics for future experiments are listed in this section. A more detailed experimental plan for 2018 will be developed, reviewed, and executed.

- The position of the air lance will be lowered further to determine whether it can effectively re-suspend particles accumulated at the bottom of the riser. The air lance will then be returned to its nominal height, and experiments will be run to determine whether accumulated layers with "equilibrium" thicknesses form again.
- A method for sampling the accumulated particles was developed and executed in 2017. A method must now be developed for dissolution of the silicone oil without impacting the magnetite particles so that particle size distribution analyses can be performed. Changes in the size of the settled particles as the experiments progress may be influencing measurements of the accumulated layer thicknesses.
- Alternative methods of measuring the amount of accumulated particles, including estimates of volume, mass, and surface area exposed to fluid flow, will be evaluated to provide further insight into the test results.
- Thicker accumulated layers will be intentionally formed via direct addition of particles to select areas of the system to better understand the ability to continue pouring and re-suspend particles. Testing may also utilize higher volume fractions (e.g., 0.5 to 1 vol %) of particles in the fluid, although this will have to be balanced with the ability to observe particle motion in a less transparent mixture.
- It may be possible to collect more quantitative information from the system in order to support crystal accumulation modeling efforts. Density-matched polymer spheres have been procured to serve as particle flow followers in the system. This will be coordinated with modeling efforts underway at Idaho National Laboratory. Modeling may help with understanding why incremental increases in the thicknesses of the settled layers are not occurring after each pouring and idling cycle.
- Results from the room temperature system will continue to be correlated with observations and data from the Research Scale Melter at Pacific Northwest National Laboratory.

6.0 References

1. Vienna, J. D., D. C. Skorski, D. S. Kim, and J. Matyáš, "Glass Property Models and Constraints for Estimating the Glass to be Produced at Hanford by Implementing Current Advanced Glass Formulation Efforts," *U.S. Department of Energy Report EWG-RPT-003, Revision 0,* Pacific Northwest National Laboratory, Richland, WA (2013).

2. Belsher, J. D. and F. L. Meinert, "High-Level Waste Glass Formulation Model Sensitivity Study 2009 Glass Formulation Model Versus 1996 Glass Formulation Model," *U.S. Department of Energy Report RPP-RPT-42649, Revision 0,* Washington River Protection Solutions, Richland, Washington (2009).

3. Jantzen, C. M. and K. G. Brown, "Predicting the Spinel-Nepheline Liquidus for Application to Nuclear Waste Glass Processing. Part I: Primary Phase Analysis, Liquidus Measurement, and Quasicrystalline Approach," *Journal of the American Ceramic Society,* **90** [6] 1866-1879 (2007).

4. Vienna, J. D. and D. S. Kim, "Preliminary IHLW Formulation Algorithm Description," *U.S. Department of Energy Report 24590-HLW-RPT-RT-05-001, Revision 0,* River Protection Project, Hanford Tank Waste Treatment and Immobilization Plant, Richland, Washington (2008).

5. Matyáš, J., J. D. Vienna, A. Kimura, M. Schaible, and R. M. Tate, "Development of Crystal-Tolerant Waste Glasses"; pp. 41-51 in Ceramic Transactions, Vol. 222, *Advances in Materials Science for Environmental and Nuclear Technology*. Edited by K. M. Fox, E. N. Hoffman, N. Manjooran and G. Pickrell. John Wiley & Sons, Inc., Hoboken, NJ, 2010.

6. Bickford, D. F. and C. M. Jantzen, "Devitrification of SRL Defense Waste Glass"; pp. 557-565 in Scientific Basis for Nuclear Waste Management VII, Edited by G. L. McVay. Elsevier, New York, 1984.

7. Vienna, J. D., D. S. Kim, M. J. Schweiger, J. S. McCloy, J. Matyáš, G. F. Piepel, and S. K. Cooley, "Test Plan: Enhanced Hanford Waste Glass Models," *U.S. Department of Energy Report TP-EWG-00001, Revision 0,* Pacific Northwest National Laboratory, Richland, WA (2013).

8. Muller, I. S., I. L. Pegg, and I. Joseph, "Test Plan: Enhanced LAW Glass Property-Composition Models, Phase 2," *U.S. Department of Energy Report VSL-13T3050-1, Revision 0,* Vitreous State Laboratory, Washington, DC (2013).

9. Matyáš, J., J. D. Vienna, A. A. Kruger, D. K. Peeler, K. M. Fox, and C. C. Herman, "Road Map for Development of Crystal-Tolerant High Level Waste Glasses," *U.S. Department of Energy Report SRNL-STI-2013-00734, Revision 0 / PNNL-23363,* Savannah River National Laboratory, Aiken, SC (2014).

10. Matyáš, J., A. R. Huckleberry, C. P. Rodriguez, J. B. Lang, A. T. Owen, and A. A. Kruger, "Empirical Model for Formulation of Crystal-Tolerant HLW Glasses"; pp. 121-128 in Ceramic Transactions, Vol. 236, *Advances in Materials Science for Environmental and Energy Technologies*. Edited by T. Ohji, M. Singh, E. Hoffman, M. Seabaugh and G. Yang. John Wiley & Sons, Inc., Hoboken, NJ, 2012.

11. Fox, K. M., M. D. Fowley, and D. H. Miller, "Crystal Accumulation in the Hanford Waste Treatment Plant High Level Waste Melter: Preliminary Settling and Resuspension Testing," *U.S. Department of Energy Report SRNL-STI-2016-00113, Revision 0,* Savannah River National Laboratory, Aiken, SC (2016).

12. Fox, K. M., M. D. Fowley, and D. H. Miller, "Crystal Accumulation in the Hanford Waste Treatment Plant High Level Waste Melter: Summary of FY2016 Experiments," *U.S. Department of Energy Report SRNL-STI-2016-00644, Revision 0,* Savannah River National Laboratory, Aiken, SC (2016).

13. Fox, K. M., "Task Technical and Quality Assurance Plan for Hanford Waste Glass Development and Characterization," *U.S. Department of Energy Report SRNL-RP-2013-00692, Revision 1,* Savannah River National Laboratory, Aiken, SC (2016).

14. Fox, K. M., M. D. Fowley, and D. H. Miller, "FY2017 Experimental Plan for the WTP High Level Waste Melter Riser Crystal Settling Test Platform," *U.S. Department of Energy Report SRNL-STI-2016-00673, Revision 0,* Savannah River National Laboratory, Aiken, SC (2016).

15. André, L., "RPP Pilot Melter Prototypic LAW Container and HLW Canister Glass Fill Test Results Report," *U.S. Department of Energy Report 24590-101-TSA-W000-0009-101-00007, Rev. 00A, TRR-PLT-080, Rev. 0,* Duratek, (2004).

16. Nelson, J., "IHLW Waste Form Qualification Report for the Hanford Tank Waste Treatment and Immobilization Plant - Introduction," *U.S. Department of Energy Report 24590-HLW-RPT-RT-08-001, Rev 0,* River Protection Project, Waste Treatment Plant, Richland, Washington (2010).

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