

Contract No:

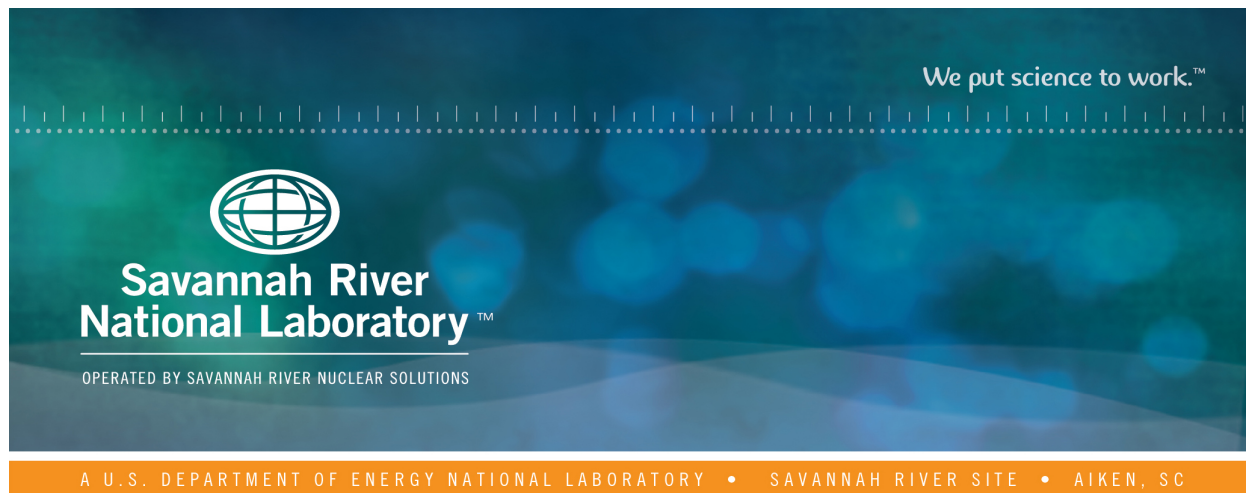
This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U. S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1) warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2) representation that such use or results of such use would not infringe privately owned rights; or
- 3) endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.



Crystal Accumulation in the Hanford Waste Treatment Plant High Level Waste Melter: Preliminary Settling and Resuspension Testing

K. M. Fox

M. D. Fowley

D. H. Miller

May 2016

SRNL-STI-2016-00113, Revision 0



DISCLAIMER

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
2. representation that such use or results of such use would not infringe privately owned rights; or
3. endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

Printed in the United States of America

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

Keywords: *Nuclear waste glass,
crystallization, melter operation*

Retention: *Permanent*

Crystal Accumulation in the Hanford Waste Treatment Plant High Level Waste Melter: Preliminary Settling and Resuspension Testing

K. M. Fox
M. D. Fowley
D. H. Miller

May 2016

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



REVIEWS AND APPROVALS

AUTHORS:

K. M. Fox, Hanford Mission Programs	Date
-------------------------------------	------

M. D. Fowley, Engineering Process Development	Date
---	------

D. H. Miller, Engineering Process Development	Date
---	------

TECHNICAL REVIEW:

E. K. Hansen, Hanford Mission Programs, Reviewed per E7 2.60	Date
--	------

APPROVAL:

C. C. Herman, Director, Hanford Mission Programs	Date
--	------

ACKNOWLEDGEMENTS

The authors thank Andy Foreman, Michael Restivo, Vernon Bush, Jon Duvall, Kenn Gibbs, and Monica Phillips for their skilled assistance with design, construction, and operation of the room temperature melter test system; Jake Amoroso for characterization of the particles used for testing; and Josef Matyáš at Pacific Northwest National Laboratory for several helpful suggestions and discussions.

Funding for this work provided by William F. Hamel, Jr., Federal Project Director of the U.S. Department of Energy Office of River Protection Waste Treatment & Immobilization Plant Project through Inter-Entity Work Order M0SRV00101 as managed by Albert A. Kruger is gratefully acknowledged.

EXECUTIVE SUMMARY

The full scale, room temperature Hanford Tank Waste Treatment and Immobilization Plant (WTP) High-Level Waste (HLW) melter riser test system was successfully operated with silicone oil and magnetite particles at a loading of 0.1 vol %. Design and construction of the system and instrumentation, and the selection and preparation of simulant materials, are briefly reviewed. Three experiments were completed. A prototypic pour rate was maintained, based on the volumetric flow rate. Settling and accumulation of magnetite particles were observed at the bottom of the riser and along the bottom of the throat after each experiment. The height of the accumulated layer at the bottom of the riser, after the first pouring experiment, approximated the expected level given the solids loading of 0.1 vol %.

More detailed observations of particle resuspension and settling were made during and after the third pouring experiment. The accumulated layer of particles at the bottom of the riser appeared to be unaffected after a pouring cycle of approximately 15 minutes at the prototypic flow rate. The accumulated layer of particles along the bottom of the throat was somewhat reduced after the same pouring cycle. Review of the time-lapse recording showed that some of the settling particles flow from the riser into the throat. This may result in a thicker than expected settled layer in the throat.

These experiments targeted a relatively low volume fraction of particles in the fluid to ensure that flow and settling could be visually observed. In actual operation, the amount of spinel crystals in the melter may approach 1 vol % or more. Therefore, the next stages of testing will utilize higher volume fractions of particles in the fluid. Thicker accumulated layers will be intentionally formed in order to better understand the ability of the system to continue pouring and resuspend particles.

Future work will evaluate the impact of maintaining a minimal air flow to the lance on particle settling and accumulation, since this is expected to be the mode of operation for the actual melter. The position of the air lance may be modified to determine whether it can effectively resuspend particles from the bottom of the riser. It may be possible to collect more quantitative information from the system in order to support crystal accumulation modeling efforts. In addition, other potential methods for recovering from an unacceptable accumulation of particles will be explored.

TABLE OF CONTENTS

LIST OF TABLES	viii
LIST OF FIGURES	viii
LIST OF ABBREVIATIONS	ix
1.0 Introduction	1
1.1 Quality Assurance	2
2.0 Experimental Procedure	2
2.1 Design and Construction of Test System	2
2.1.1 Mockup	2
2.1.2 Support Equipment	4
2.1.3 Instrumentation	5
2.1.4 Cameras	6
2.2 Materials Selection and Preparation	6
2.3 Operating Protocol	7
2.3.1 Preparation	7
2.3.2 Starting and Stopping the Pour	8
2.3.3 Post Test and Restarting the Pour	9
3.0 Preliminary Particle Settling Tests	9
3.1 Data and Observations for Experiments 1 through 3	9
3.2 Resuspension of Particles During Experiment 3	12
3.3 Particle Settling after Experiment 3	17
4.0 Summary and Future Work	21
5.0 References	22

LIST OF TABLES

Table 2-1. Instrumentation List.....	6
Table 2-2. Viscosity Measurements of 5000 cSt Silicone Oil	7
Table 2-3. Values Used in Calculating the Mass of Magnetite Needed for a Loading of 0.1 Volume Percent	8
Table 3-1. Slurry Temperature and Calculated Silicone Oil Viscosity.....	11

LIST OF FIGURES

Figure 2-1. Drawings of the SRNL feed tank, throat, and riser (a), the SRNL mockup trough (b), and the WTP HLW melter trough (c).....	3
Figure 2-2. Full-scale, room temperature WTP HLW melter riser test system	4
Figure 2-3. Piping and instrumentation diagram for full-scale, room temperature WTP HLW melter riser test system.....	5
Figure 2-4. View of silicone oil and magnetite particles exiting trough during pouring.....	9
Figure 3-1. Platform scale output and calculated pour rate during operation	10
Figure 3-2. Air flow and pressure to the air lance.....	11
Figure 3-3. First bubble at the start of the third experiment	13
Figure 3-4. Front of suspended particles (white arrows) advancing up the throat during pouring.....	13
Figure 3-5. Particles flowing through the throat and riser, leaving a dead zone at the bottom of the riser (white arrows).....	14
Figure 3-6. Dead zone is no longer visible after approximately 15 minutes of pouring.....	14
Figure 3-7. Comparison of settled layers at the start of pouring (left) and after 15 minutes of pouring (right).....	15
Figure 3-8. Comparison of settled layer along the bottom of the throat at the start of pouring (left) and after 15 minutes of pouring (right)	16
Figure 3-9. Particle settling in the riser and throat after the third pouring experiment.....	18
Figure 3-10. Accumulation of particles at the bottom of the riser and throat after the third pouring experiment	19
Figure 3-11. Accumulation of particles along the bottom of the throat after the third pouring experiment	20

LIST OF ABBREVIATIONS

DAS	Data Acquisition System
DOE	U.S. Department of Energy
HLW	High-Level Waste
LAW	Low-Activity Waste
ORP	Office of River Protection
PC	Personal Computer
SRNL	Savannah River National Laboratory
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 vol% (nominally at 950 °C).⁴ An alternative crystal-tolerant glass approach⁵ 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 form⁶ 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. 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

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

system uses transparent materials to allow for the observation of particle behavior under a variety of process 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.

This report describes the design and construction of the full-scale, room temperature WTP HLW melter riser system. Preliminary experiments using materials simulating molten glass and spinel crystals are described. 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.¹¹ Experimental plans were developed and issued for system design and material selection,¹² as well as preliminary particle settling experiments.¹³

1.1 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Savannah River Site Manual E7 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 L008-00162-01.

2.0 Experimental Procedure

The following sections describe the design of the test system, the materials used to simulate molten glass and spinel crystals, and operation of the system to evaluate particle settling and resuspension.

2.1 Design and Construction of Test System

2.1.1 *Mockup*

A mockup of the WTP HLW melter pour spout, consisting of the throat, riser, and trough, was assembled using clear acrylic tubing as shown in Figure 2-1(a). The inner diameters, lengths, and orientation of the riser and throat are prototypic. The trough is partially prototypic (Figure 2-1(b)); the body was simplified such that the flow path up to the weir is prototypic with that of the actual trough (Figure 2-1(c)) but the flow path after the weir is dissimilar. The flow path was constructed as such to simulate the hydraulic head and glass volume above the riser tube; the dynamic effects past the weir were disregarded since the glass is assumed to flow to the canister at that point. Drawings WTP-M-21106^a (throat and riser), WTP-M-21562^b (trough), and WTP-M-21953^c (final assembly drawing) were supplied by DOE-ORP and referenced for prototypic dimensions.

The throat is connected to a cylindrical feed tank (Figure 2-1(a)) that supplies the simulated glass and crystals (silicone oil and magnetite particles) for the pour. The feed tank represents the melt pool of the actual melter, although only the height of the throat relative to the bottom of the tank is prototypic. The feed tank was kept much smaller in volume than the actual melter in order to minimize the quantity of fluid required for testing. The height of the feed tank is prototypic to simulate the hydraulic head affecting the throat and riser. Pressure differentials resulting from melter off gas system operation were not considered in the design of this system. Figure 2-2 is a photograph of the assembled system.

^a "Refractory Details Monofrax E," WTP-M-21106 Rev. 1, March 7, 2002.

^b "Discharge Chamber Trough," WTP-M-21562 Rev. 0, September 27, 2002

^c "HLW Melter Final Assembly Sequence Section 2," WTP-M-21953 Rev. 3, February 14, 2005

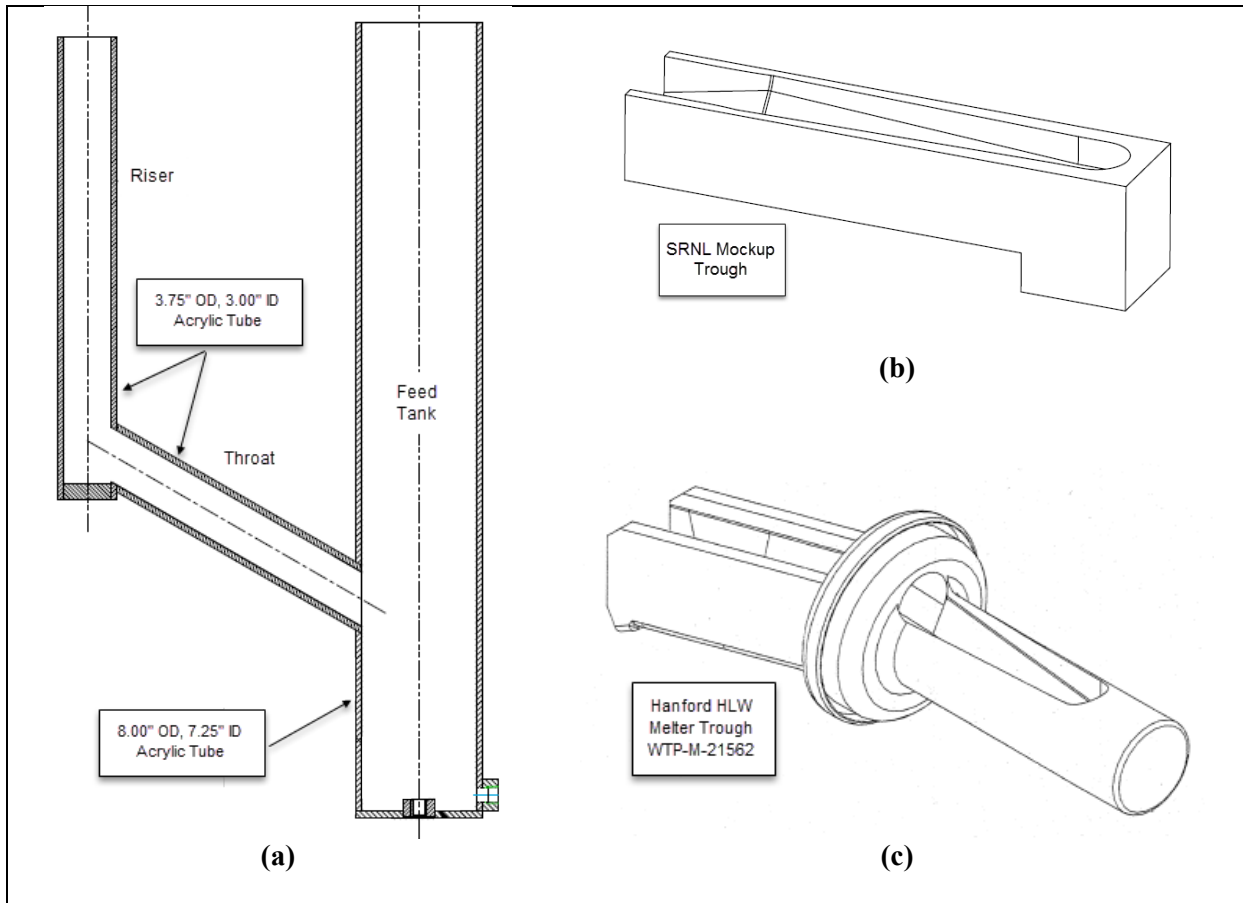


Figure 2-1. Drawings of the SRNL feed tank, throat, and riser (a), the SRNL mockup trough (b), and the WTP HLW melter trough (c)

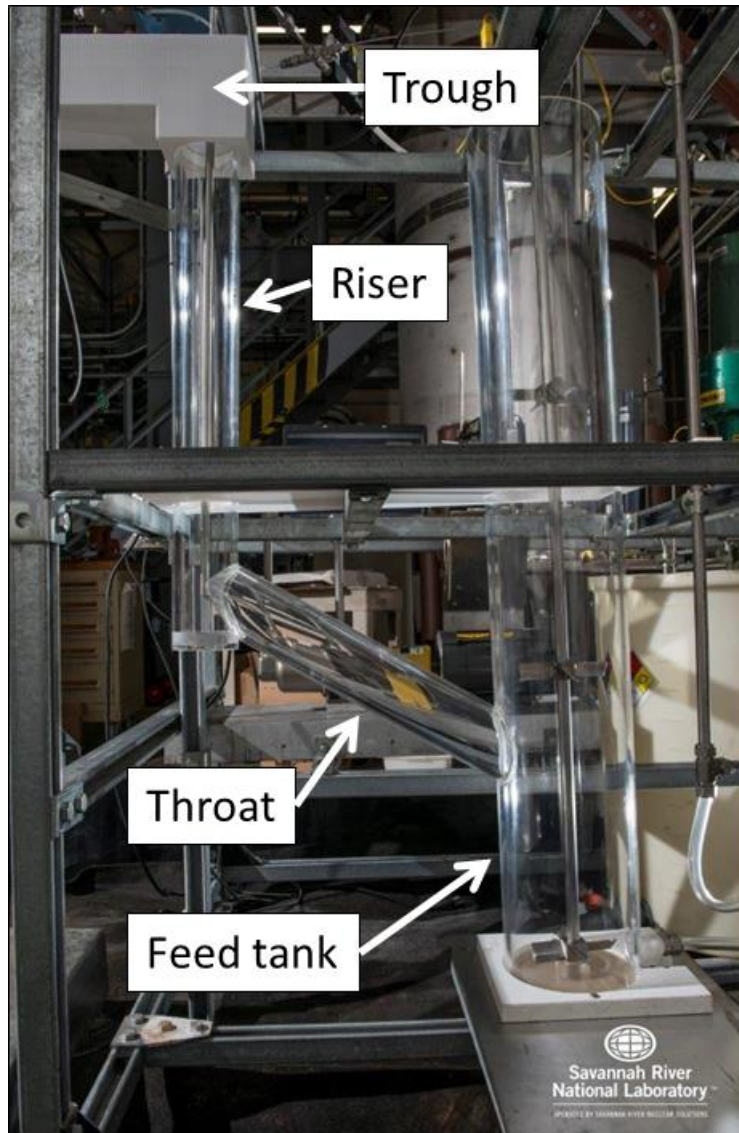


Figure 2-2. Full-scale, room temperature WTP HLW melter riser test system

2.1.2 Support Equipment

Figure 2-3 shows the mockup and support equipment for particle accumulation testing. An air lance, made of 0.375 inch inside diameter stainless steel tubing, was inserted into the riser to a prototypic height above the bottom of approximately 3.2 inches. The end of the tube is flat and deburred. The air lance dimensions, geometry, and location were provided via email from EnergySolutions, Inc. (see Appendix A). Air is supplied from building process air to a flow controller that regulates the air flow to the lance and thus controls the pour rate. The trough discharges material into a receipt tank placed on a platform scale. The output of the scale is used to determine the pour rate.

Material is transferred to the feed tank from a 30 gallon makeup tank with a progressive cavity pump (P-1 in Figure 2-3). A second peristaltic transfer pump (P-3) is available to assist P-1 as necessary. Normal duty for P-3 is transferring slurry back to the makeup tank from various tanks or containers. Mixing in

the makeup tank is provided by a double impeller configuration with a 4-bladed flat impeller at the bottom and a 3-bladed airfoil impeller above.

Mixing in the feed tank is provided by a triple impeller configuration using three 4-bladed, flat, angled (45°) impellers. A peristaltic circulation pump (P-2 in Figure 2-3) assists the feed tank mixing by transferring solids-laden material from the bottom to the top of the tank.

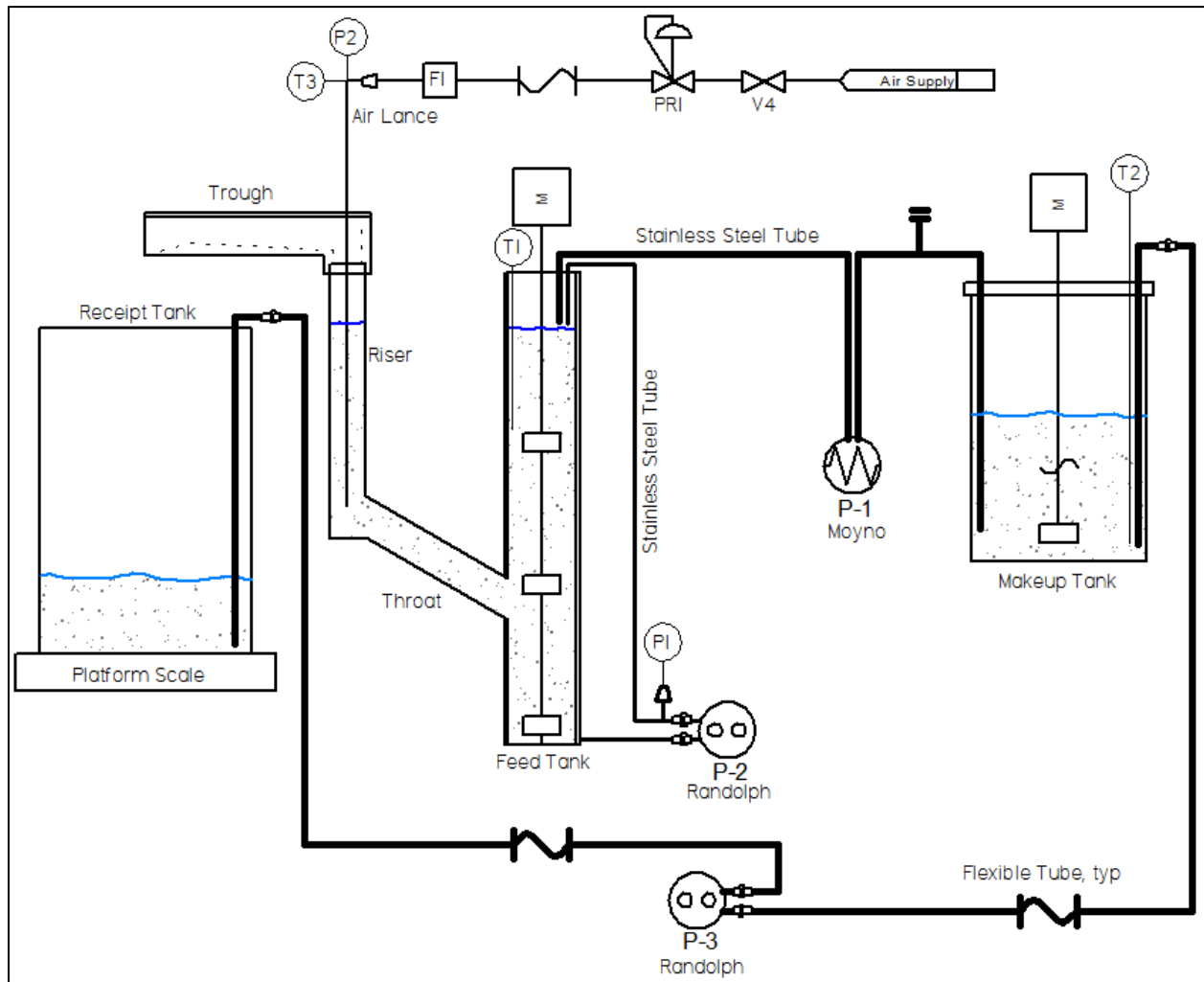


Figure 2-3. Piping and instrumentation diagram for full-scale, room temperature WTP HLW melter riser test system

2.1.3 Instrumentation

System instrumentation includes temperature measurements in the feed tank, makeup tank, and air supply to the lance. Pressure is measured in the lance and at the discharge of the peristaltic pump P-2 (which is necessary to verify that the pump tubing pressure limit is not exceeded). Air flow to the lance is measured and controlled. The platform scale measures mass accumulation during the pour, with the time-averaged data being converted to a pour rate. Detail of the instruments used in the system is provided in

Table 2-1. The output of these instruments is displayed and logged with a data acquisition system (DAS) consisting of a personal computer (PC) using LabVIEW software^a and signal processing hardware.

Table 2-1. Instrumentation List

Designation (see Figure 2-3)	Description	Instrumentation	Range	Tolerance
M1	Platform scale	Metler-Toledo PTHN	0 - 1000 lbs	0.275 lbs, plus 0.03% of reading
F1	Mass flow controller	Omega FMA5542A	0 - 100 lpm	4 lpm
P1	Gauge pressure transmitter	Rosemont 1144G0120A22M1	0 - 50 psig	0.5% of full scale
P2	Differential pressure transmitter	Rosemont 1151DP4E22	0 - 150 in. H ₂ O	0.5% of full scale
T1	Type K thermocouple 0.125 in. dia., 18 in. long	Omega KQXL-18U-18	0 - 100 °C	2.2°C, or 0.75% of reading
T2	Type K thermocouple 0.25 in. dia., 38 in. long	Omega ICSS-14U-38-NHX	0 - 100 °C	2.2°C, or 0.75% of reading
T3	Type K thermocouple 0.125 in. dia., 6 in. long	Omega KQXL-18U-6	0 - 100 °C	2.2°C, or 0.75% of reading

2.1.4 Cameras

A camera system consisting of three digital cameras is used to capture activity in the mockup during a pour and following a pour when entrained solids are settling. The first camera is positioned to capture the junction of the riser and throat. The second camera is focused on the underside of the throat. The third camera captures an overall view of the riser and throat. The digital outputs are recorded on a PC using LabVIEW software.^b The system can be configured to record either real time video or time lapse photographs. Typically, real time video is recorded during a pour and time-lapse photographs are captured during settling and later combined into a video.

2.2 Materials Selection and Preparation

Selection of a silicone oil to simulate molten glass and magnetite particles to simulate spinel crystals is described in the first experimental plan.¹² In summary, silicone oil with a vendor designated viscosity of 5000 cSt and a specific gravity at 25 °C of 0.975, was procured from Clearco Products Company, Inc. to support the first series of experiments.^c After receipt, duplicate (minimum) flow curve measurements were obtained using a Haake RS6000 rheometer and Z41 geometry (concentric geometry) for three different temperatures covering the expected operating temperatures during testing. The flow curve measurements covered the shear rate range of 0 to 100 1/sec. These data were fitted with a Newtonian fluid model. The average and one standard deviation results are provided in Table 2-2.

^a National Instruments™ LabView 2014, Service Pack 1, version 14.0.1

^b National Instruments™ LabView 2012, Professional Development System, Version 12.0

^c High Viscosity Polydimethylsiloxane Fluid / PDMS Silicone Fluid (CAS # 63148-62-9)

Table 2-2. Viscosity Measurements of 5000 cSt Silicone Oil

Temp (°C)	Viscosity (cP)	Std (cP)	Measurements
15.03	6848	0.6	2
20.08	6149	1.7	3
24.15	5553	0.5	2

The data in Table 2-2 were regressed using an exponential model to predict the viscosity over the range of expected temperatures in the laboratory:

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

where η is the viscosity of the fluid (cP) and T is the temperature of the fluid (Kelvin).

Magnetite particles were selected to simulate spinel crystals, as described in the first experimental plan.¹² Magnetite particles screened to a vendor designated size range of -120 to +140 mesh (125 to 105 μm) were procured from F. J. Brodmann and Company. Significant fines were discovered in the magnetite during earlier bench top testing, which implied that either the vendor's sieving was inadequate for the experiments, the particles were fracturing during mixing, or both. The fines remained suspended in the oil during the bench top experiments, hindering visual observation of settling. The particle size distribution of the magnetite particles was improved by further sieving and elutriation in water. To maximize the amount of "cleaned" material available, particles 75 μm and greater (i.e., a range of 75 μm to 125 μm , assuming the vendor's upper size range is correct) were retained and used for testing in the full scale system.

2.3 Operating Protocol

In general, an experiment was conducted by preparing the slurry, filling the system, starting a pour, stopping a pour and allowing particles to settle, and finally restarting a pour to determine the effects of settled solids in the riser and throat. These steps are described in further detail in the following sections.

2.3.1 Preparation

Initial testing of the riser system was completed using silicone oil without magnetite particles. This testing confirmed operation of each of the system components, and the results were summarized in a memorandum.¹⁴ At the completion of these tests, the silicone oil was drained from the system and stored in the makeup tank.

To prepare for the particle settling tests, a mass of magnetite particles sufficient to provide a loading of 0.1 vol % was combined and mixed with the silicone oil in the makeup tank. The volume of silicone oil in the system was determined geometrically using a calibrated scale on the side of the makeup tank. The values used in calculating the appropriate mass of magnetite to produce a loading of 0.1 vol % in the silicone oil are presented in Table 2-3.

Table 2-3. Values Used in Calculating the Mass of Magnetite Needed for a Loading of 0.1 Volume Percent

Silicone Oil Volume	102.91 liters
0.1 Volume %	102.9 ml
Magnetite Density	5.2 g/ml
Magnetite Mass	535.1 g

The blended material was then transferred to the feed tank using pump P-1, where the feed tank agitator and circulation pump P-2 were used to maintain mixing.

2.3.2 Starting and Stopping the Pour

Prior to starting a pour, the DAS log and the real time video cameras were started. The pour was initiated by starting air flow to the lance in the riser. The air flow was set using the DAS and the pour rate was monitored with the platform scale. As necessary, adjustments to the air flow were made to maintain the pour rate. Once the pour started, the transfer pump P-1 was used to maintain the feed tank level at 44 inches. The second transfer pump, P-3, was used as needed to assist pump P-1. The pour (change in mass measured with the platform scale) was maintained at a nominal rate of 3.18 liters per minute (Figure 2-4) until the makeup tank was nearly empty (approximately 15 minutes). This pouring rate was selected to be prototypic of the volume of glass 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/cm³.

Prior to stopping the pour, the real time video recording was stopped and recording of time-lapse photographs was started. The pour was stopped by turning off the air flow to the lance. The transfer to the feed tank and the tank agitators were also stopped. The system was left idle to allow the particles to settle in the riser and throat. During the idle time, the poured slurry was transferred from the receipt tank to the makeup tank using transfer pump P-3.

^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 rate production rate, and therefore higher pouring rate, was used in this study and considered to be an upper bound. Future experiments may utilize lower pouring rates for comparison.

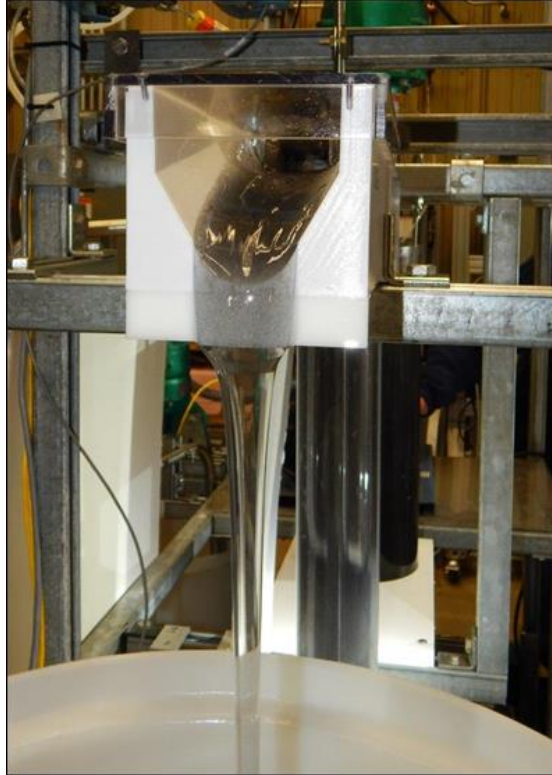


Figure 2-4. View of silicone oil and magnetite particles exiting trough during pouring

2.3.3 Post Test and Restarting the Pour

After a period of time where settling occurred, recording of time-lapse photographs was stopped. The series of images from each camera was converted into a movie file. The DAS log was analyzed. The mockup was then prepared for the pour restart. The agitators in the makeup tank and feed tank were first started to mix the slurry. The peristaltic pump, P-2, was operated to transfer solids-laden material from the bottom to the top of the feed tank. The DAS log and real time video recording were restarted. The pour was restarted by applying air to the lance using the DAS, and then starting the transfer pumps to maintain the level in the feed tank.

3.0 Preliminary Particle Settling Tests

Three particle settling experiments have been completed with the WTP HLW melter riser test system. The first test was used to verify visual observation as a viable method for identifying particle movement and settling in the full scale system. The second test was used to demonstrate a restart of pouring with particles in the system, to demonstrate the ability to maintain a consistent pour rate, and to increase the thickness of the settled layers of particles within the throat and riser. The third test monitored changes in the thickness of the settled layers during a pouring cycle. Detailed results from the third test are provided in the discussion that follows.

3.1 Data and Observations for Experiments 1 through 3

Figure 3-1 shows the platform scale output recorded during the three particle settling experiments, as well as the volumetric pour rate calculated from the scale output and slurry density. As described earlier, a pour rate of 3.18 lpm was targeted for each pour, corresponding to a Hanford HLW melter pouring rate of

12 metric tons of glass per day. The system was able to maintain the targeted pour rate reasonably well during each of the experiments. The scatter in the calculated rate is due to scale resolution and pour rate fluctuation (due to the pulsing nature of the air lift). The settling period between pours was several days each (approximately 100 hours after the initial pour and approximately 212 hours after the first restart).

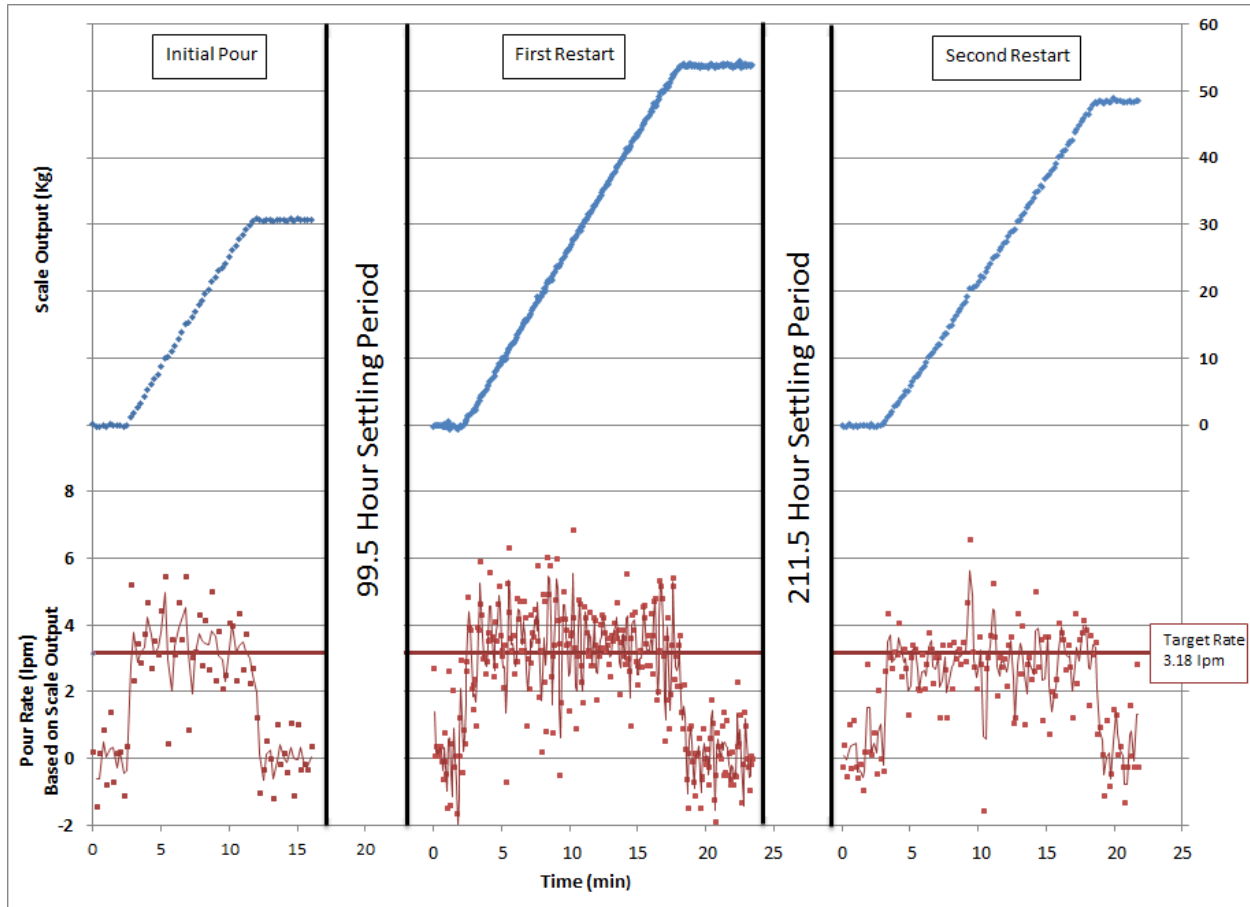


Figure 3-1. Platform scale output and calculated pour rate during operation

The air flow and air pressure to the lance during the three experiments are shown in Figure 3-2. The air flow rate required to achieve the targeted pour rate was approximately 37 standard liters per minute (slpm), which would produce a pressure in the lance of approximately 24 in. H₂O (6 kPa). The air flow rate was generally kept constant throughout a pouring cycle.

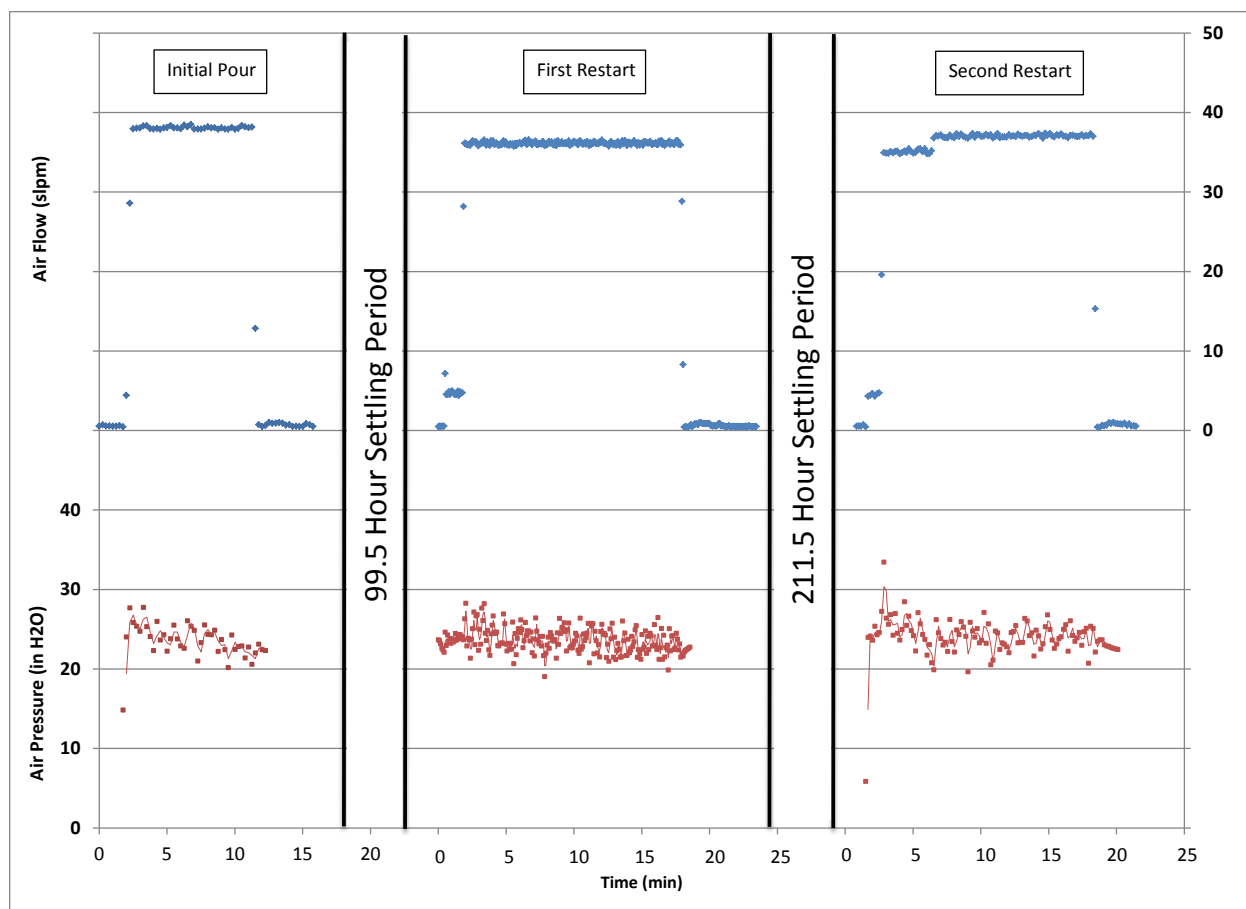


Figure 3-2. Air flow and pressure to the air lance

The temperature of the slurry during the three experiments, recorded at T1, and the calculated viscosity of the silicone oil at those temperatures (from Equation 1) are shown in Table 3-1. The ambient temperature in the laboratory varied from day to day, resulting in changes to the viscosity of the silicone oil among the three experiments. Qualitatively, this did not appear to impact particle settling behavior.

Table 3-1. Slurry Temperature and Calculated Silicone Oil Viscosity

Particle Settling Experiment	Slurry Temperature	Calculated Silicone Oil Viscosity
1	22.5 °C	5842 cP
2	24.7 °C	5585 cP
3	16.7 °C	6599 cP

A thin layer of solids developed at the bottom of the riser and in the throat after each settling period. After the first experiment, the solids layer at the bottom of the riser was approximately 0.05 inches (1.27 mm) thick (determined via visual estimation through the transparent tube). Assuming a solids packing factor of about 40%, the settled solids account for about 0.1% of the riser volume, indicating that the solids concentration in the mockup was consistent with the targeted value (with the assumption that

all of the solids suspended in the riser settled to the bottom). After the second experiment, the layer of solids in the riser increased in height.

3.2 Resuspension of Particles During Experiment 3

The third experiment was used to monitor particle movement during pouring and resuspension of the settled material accumulated during the first two pouring cycles. Figure 3-3 is a photograph of the junction between the throat and the riser at the start of the third experiment. The first bubble can be seen emerging from the air lance. An accumulated layer of magnetite particles is visible at the bottom of the riser and along the bottom of the throat. The silicone oil is relatively clear after approximately 212 hours of settling time since the previous experiment. After approximately one minute of pouring, a front of suspended particles from the feed tank can be seen advancing up the throat toward the riser in Figure 3-4.

As pouring continued, the suspended particles entered the riser and were lifted upwards and out. At approximately 1.5 minutes after the start of pouring, the particles were observed to flow from the throat to the riser, leaving a “dead zone” at the bottom of the riser (Figure 3-5). This indicates that the flow of silicone oil and suspended particles was not interacting with the settled layer of particles at the bottom of the riser. After approximately 15 minutes of pouring, the concentration of particles in the throat and riser had increased to the point where the dead zone was no longer visible. However, the settled layer at the bottom of the riser appeared unchanged (Figure 3-6). The air lift and flow from the throat did not mobilize this region of the riser.

Figure 3-7 provides a comparison of the thickness of the settled layers at the beginning and end of the third experiment. The white lines overlaid on each photograph are equally spaced. The thickness of the settled layer at the bottom of the riser, labeled A in Figure 3-7, appears to be unchanged after 15 minutes of pouring. The thickness of the settled layer along the bottom of the throat, labeled B in Figure 3-7, appears to be reduced after 15 minutes of pouring. Figure 3-8 provides alternate views of the distribution of particles along the bottom of the throat at the beginning and end of the third experiment. The white lines overlaid on each photograph are equally spaced. Again, the thickness of the settled layer along the bottom of the throat, labeled A in Figure 3-8, appears to be reduced after 15 minutes of pouring. These observations indicate that the flow of material through the throat during this experiment was sufficient to resuspend some of the accumulated particles. Note however that smaller particles are expected to have settled last, and may therefore have been easier to resuspend in the subsequent pouring cycle as compared to larger particles.



Figure 3-3. First bubble at the start of the third experiment



Figure 3-4. Front of suspended particles (white arrows) advancing up the throat during pouring

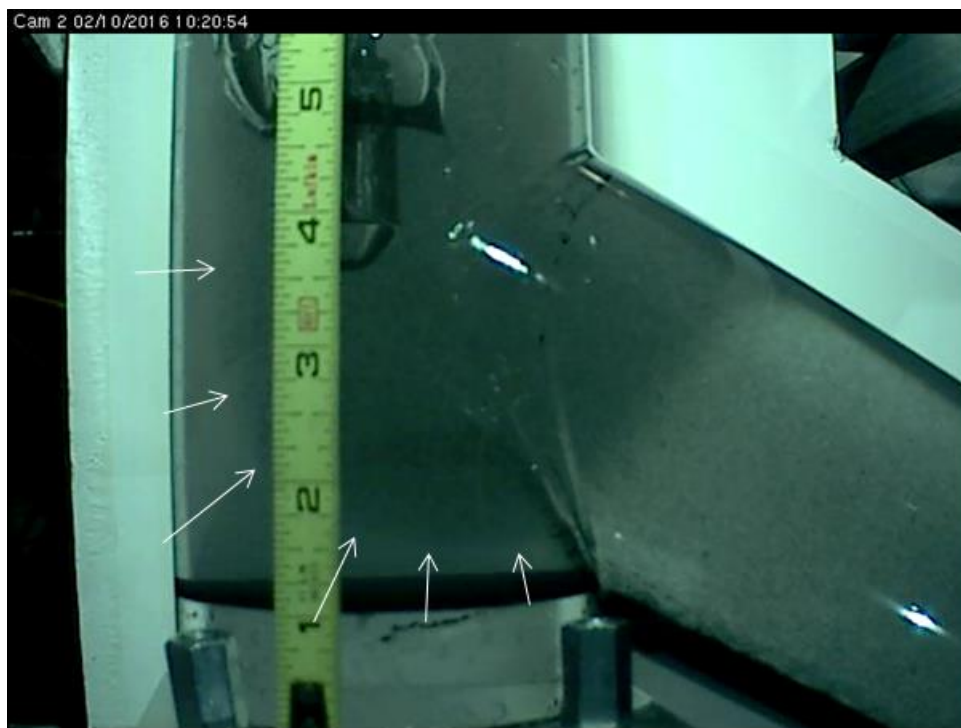


Figure 3-5. Particles flowing through the throat and riser, leaving a dead zone at the bottom of the riser (white arrows)



Figure 3-6. Dead zone is no longer visible after approximately 15 minutes of pouring

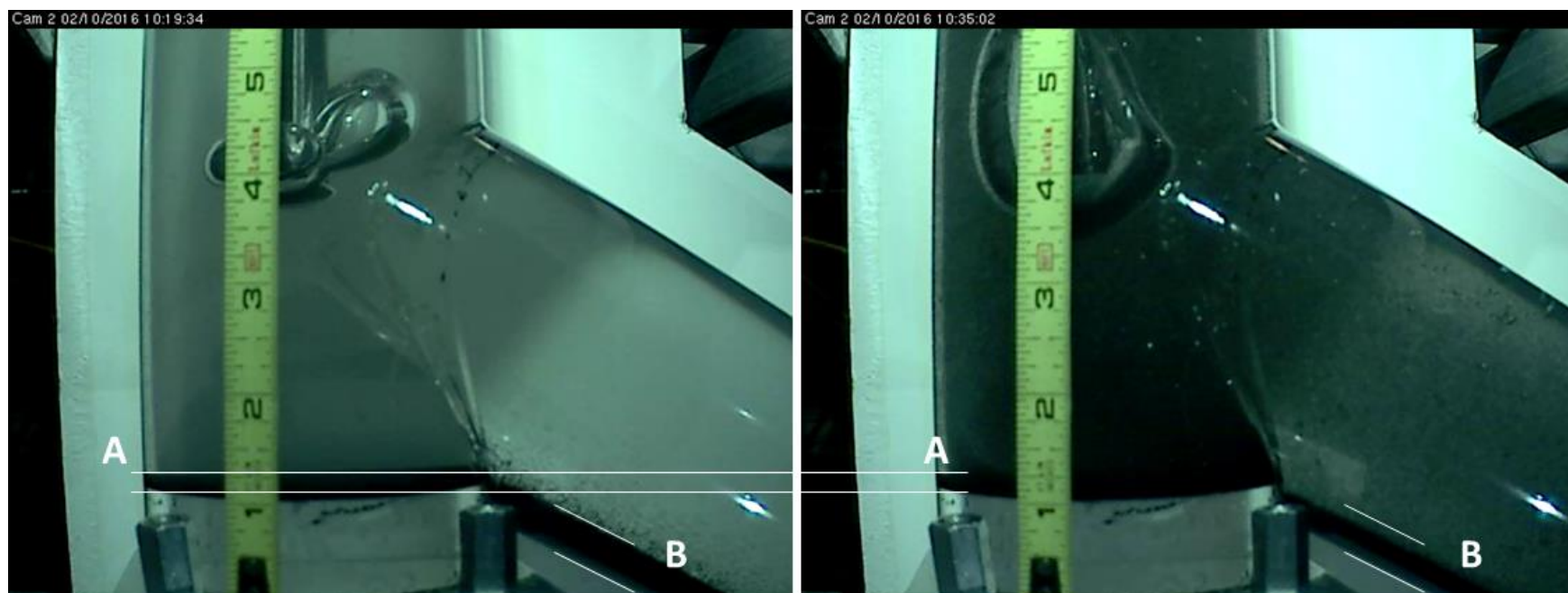


Figure 3-7. Comparison of settled layers at the start of pouring (left) and after 15 minutes of pouring (right)

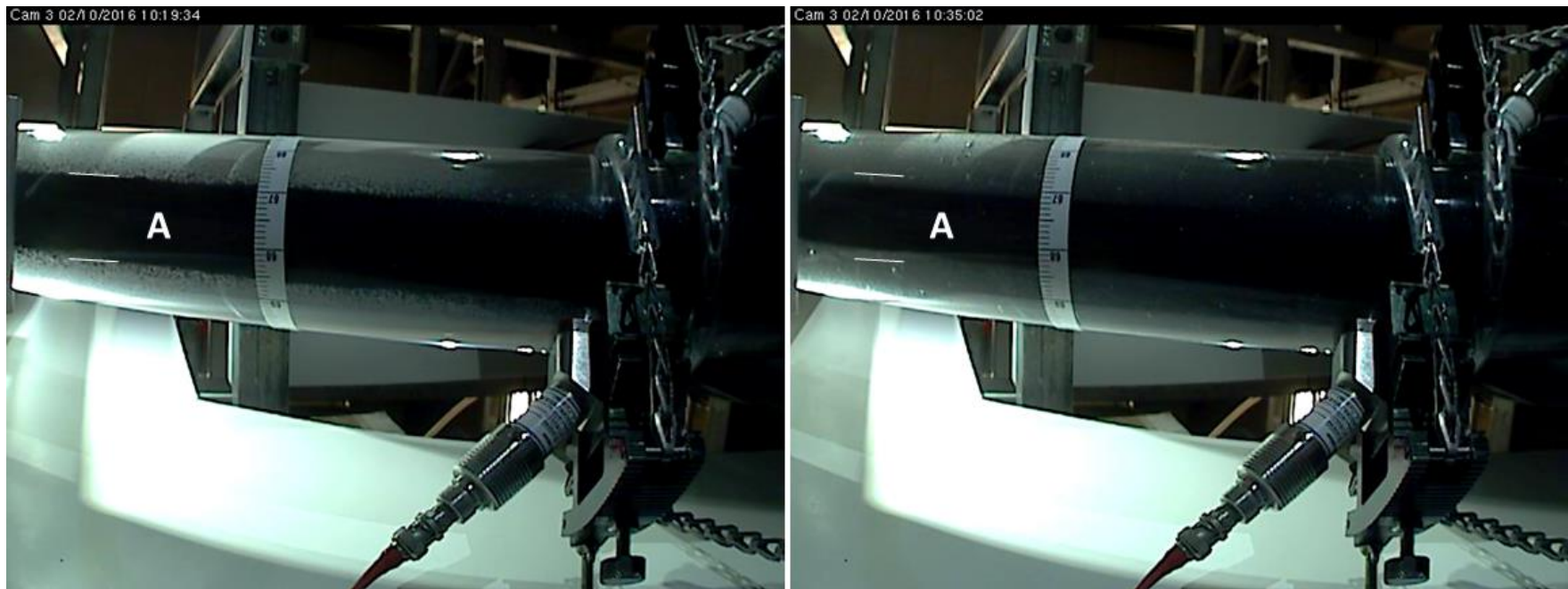


Figure 3-8. Comparison of settled layer along the bottom of the throat at the start of pouring (left) and after 15 minutes of pouring (right)

3.3 Particle Settling after Experiment 3

Particle settling in the riser and throat was recorded via time-lapse photography after the third pouring experiment. Figure 3-9 shows the state of settling in the riser and throat at two intervals (approximately 4.5 and 20.5 hours) after pouring was stopped. The silicone oil is seen to become more clear, and accumulated layers are visible at the bottom of the riser and along the bottom of the throat.

Figure 3-10 includes four photographs of the junction between the throat and riser as settling occurred after pouring was stopped. A review of the time-lapse video from this camera view showed that a considerable amount of particles flowed from the riser to the throat during settling. This likely increases the thickness of the settled layer within the throat relative to what would accumulate solely from settling of the material remaining in the throat.

Figure 3-11 includes four photographs of the bottom of the throat at the same time intervals shown in Figure 3-10. The accumulation of particles along the bottom of the throat over time is apparent. A review of the time-lapse video from this camera view showed sliding of the particles along the inner wall of the throat, which may not be representative of the actual melter should the spinel crystals adhere to the refractory.

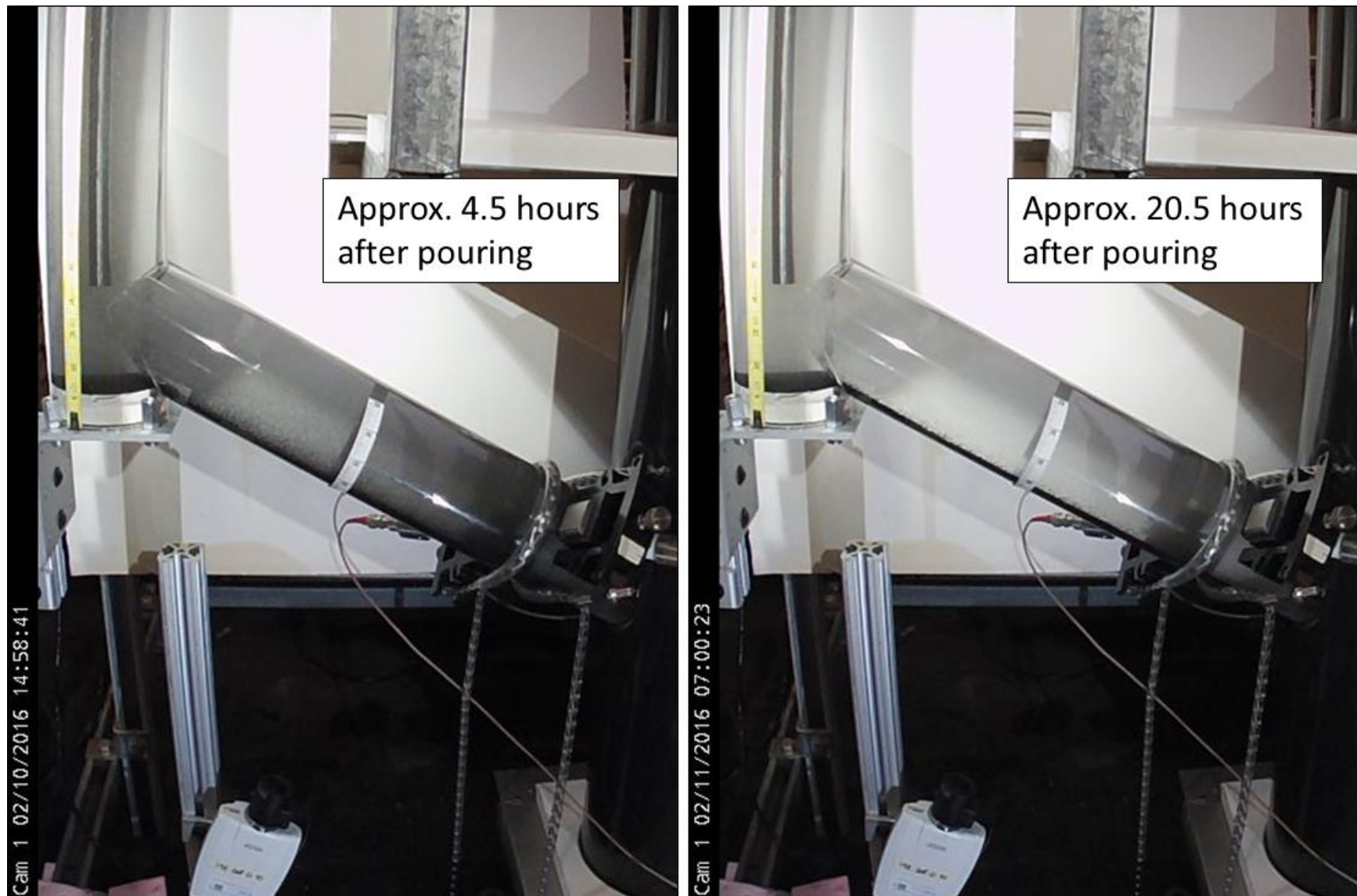


Figure 3-9. Particle settling in the riser and throat after the third pouring experiment

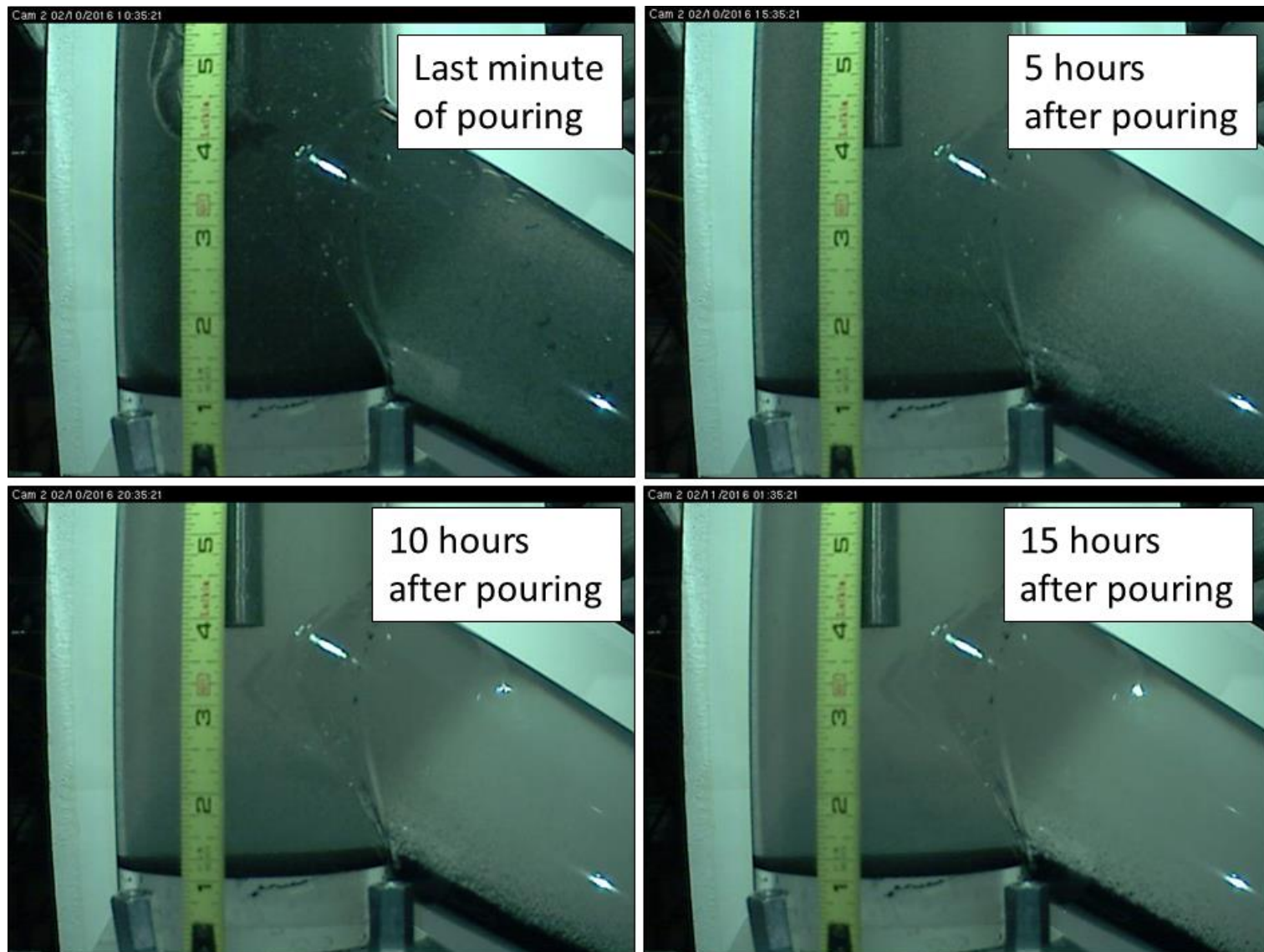


Figure 3-10. Accumulation of particles at the bottom of the riser and throat after the third pouring experiment

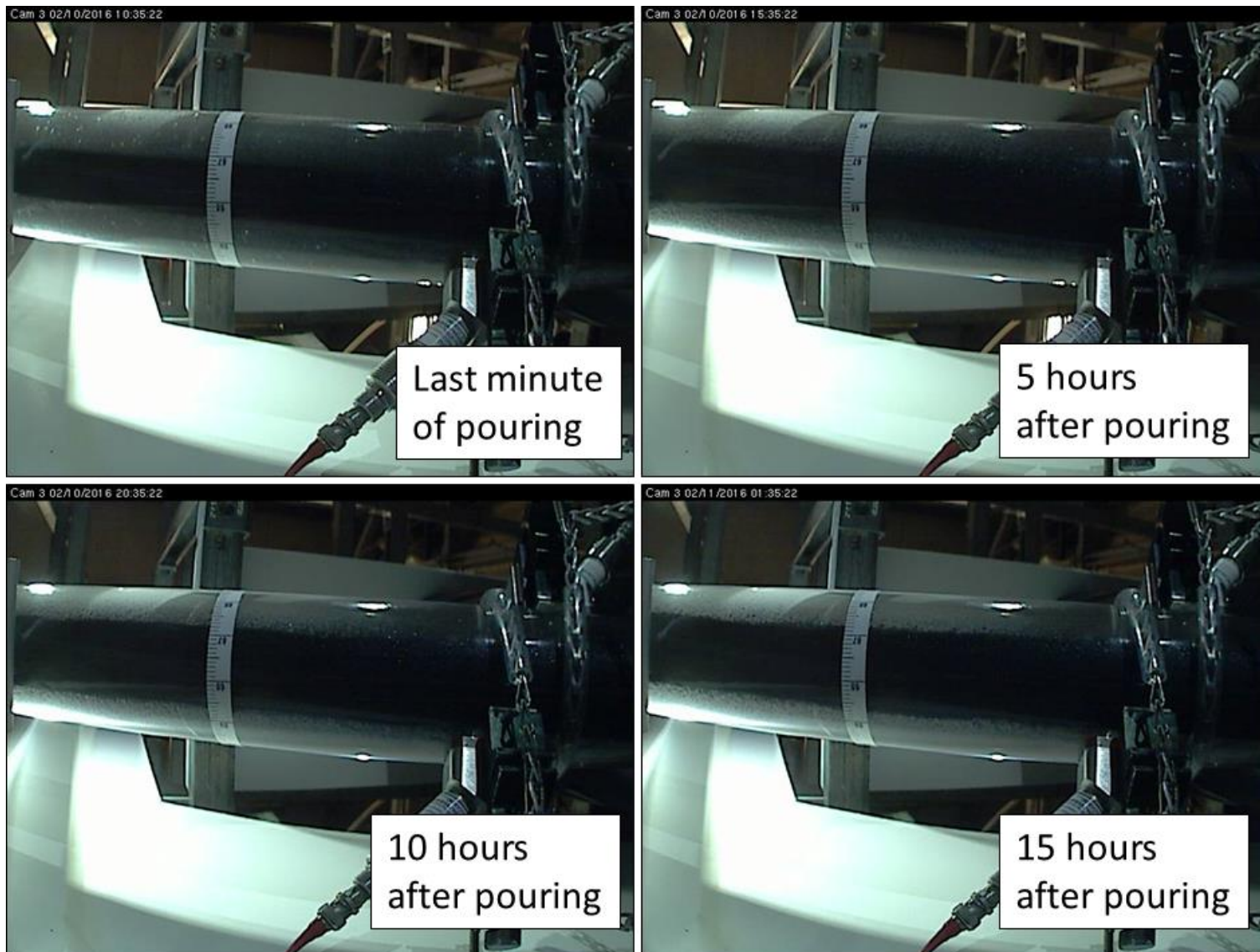


Figure 3-11. Accumulation of particles along the bottom of the throat after the third pouring experiment

4.0 Summary and Future Work

The full scale, room temperature WTP HLW melter riser test system was successfully operated with silicone oil and magnetite particles at a loading of 0.1 vol %. Three experiments were completed. A prototypic pour rate was maintained, based on the volumetric flow rate. Settling and accumulation of magnetite particles were observed at the bottom of the riser and along the bottom of the throat after each experiment. The height of the accumulated layer at the bottom of the riser, after the first pouring experiment, approximated the expected level given the solids loading of 0.1 vol %.

More detailed observations of particle resuspension and settling were made during and after the third pouring experiment. The accumulated layer of particles at the bottom of the riser appeared to be unaffected after a pouring cycle of approximately 15 minutes at the prototypic flow rate. The accumulated layer of particles along the bottom of the throat was somewhat reduced after the same pouring cycle. Settling of the particles was observed via time-lapse photography after pouring was completed. Review of the time-lapse recording showed that some of the settling particles flow from the riser into the throat. This may result in a thicker than expected settled layer in the throat.

The preliminary resuspension experiments may be conservative in that the system is constructed of smooth wall material rather than refractory. Therefore, resuspension of the particles may require less effort than in the actual melter. Smaller particles are expected to have settled last, and may therefore be easier to resuspend in a subsequent pouring cycle as compared to larger particles. Also, the particles are unlikely to agglomerate or adhere to the walls during testing, which may not be true of crystals in a molten glass. The particle size distribution in the experiments was intentionally kept narrow, with removal of fine particles to support visual observations. A broader particle size distribution may form a better packed settled layer that would be more difficult to resuspend.

Based on the results presented in this report, the next stages of work have been defined:

- The experiments described here targeted a relatively low volume fraction of particles in the fluid to ensure that flow and settling could be visually observed. In actual operation, the amount of spinel crystals in the melter may be 1 vol % or more. Therefore, the next stages of testing will utilize higher volume fractions of particles in the fluid. Thicker accumulated layers will be intentionally formed in order to better understand the ability of the system to continue pouring and resuspend particles.
- Air flow to the lance was stopped during settling periods for these experiments. In actual operation, a minimal flow through the air lance will be maintained during idle periods. The next stages of testing will evaluate the impact of maintaining a minimal air flow to the lance on particle settling and accumulation.
- Future experiments will continue to monitor pressures and air flow to determine whether pouring functions are being impacted by accumulated particles. The position of the air lance may be modified to determine whether it can effectively resuspend particles from the bottom of the riser. It may be possible to collect more quantitative information from the system in order to support crystal accumulation modeling efforts.
- Correlation of the results from the room temperature system described here with data from the Research Scale Melter (RSM) at Pacific Northwest National Laboratory is of interest. To assist with these correlations, the next stages of testing will attempt to quantify the particle settling rates for magnetite in silicone oil for comparison with the measured settling rates of spinel crystals in molten glass, and the changes in thickness of the accumulated layers will be measured.

5.0 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. and D. K. Peeler, “Task Technical and Quality Assurance Plan for Hanford HLW Glass Development and Characterization,” *U.S. Department of Energy Report SRNL-RP-2013-00692, Revision 0*, Savannah River National Laboratory, Aiken, SC (2013).

12. Miller, D. H. and M. D. Fowley, "Experimental Plan for Crystal Accumulation Studies in the WTP Melter Riser," *U.S. Department of Energy Report SRNL-STI-2015-00240, Revision 0*, Savannah River National Laboratory, Aiken, SC (2015).
13. Fox, K. M., D. H. Miller, M. D. Fowley, and M. L. Restivo, "Phase 2 Experimental Plan for the Hanford WTP High Level Waste Melter Riser Crystal Settling Test System," *U.S. Department of Energy Report SRNL-STI-2015-00451, Revision 0*, Savannah River National Laboratory, Aiken, SC (2015).
14. Fowley, M. D., D. H. Miller, and K. M. Fox, "Status of the WTP HLW Melter Riser Test System," *U.S. Department of Energy Memorandum SRNL-L3000-2015-00017*, Savannah River National Laboratory, Aiken, SC (2015).
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).

Appendix A Air Lance Dimensions

From: Innocent Joseph <IJOSEPH@energysolutions.com>
Date: March 20, 2015 at 11:28:55 AM PDT
To: "Kruger, Albert A (Albert_A_Kruger@orp.doe.gov)" <Albert_A_Kruger@orp.doe.gov>
Cc: Brad Bowan <BBOWAN@energysolutions.com>, Eric Smith <ESMITH@energysolutions.com>
Subject: Dimensions of the air lift lance

Albert,

Given below are the dimensions of the air lift lance.

- (1) Outer diameter of the tube is 0.500"
- (2) Inner diameter of the tube is 0.375"
- (3) The distance from the bottom of the rise to the lance mounting flange is 86.568"
- (4) The length of the lance below the flange is 83.34"

The air lift lance does not have a nozzle at the end.

Please let us know if you need any other information.

Thank you
Innocent

EnergySolutions , Inc.
6350 Stevens Forest Road, Suite 200
Columbia, MD 21045
Phone: (240) 565-6142

Distribution:

J. W. Amoroso, 999-W
H. P. Boyd, 707-7E
T. B. Brown, 773-A
H. H. Burns, 773-41A
A. S. Choi, 999-W
A. D. Cozzi, 999-W
C. L. Crawford, 773-42A
D. E. Dooley, 999-W
A. P. Fellingner, 773-42A
T. L. Fellingner, 766-H
S. D. Fink, 773-A
E. J. Freed, 704-S
M. D. Fowley, 786-A
K. M. Fox, 999-W
E. K. Hansen, 999-W
C. C. Herman, 773-A
E. N. Hoffman, 999-W
E. W. Holtzscheiter, 766-H
J. E. Hyatt, 773-A
J. F. Iaukea, 704-27S
D. C. Iverson, 704-30S
C. M. Jantzen, 773-A

F. C. Johnson, 999-W
D. S. Kim, PNNL
A. A. Kruger, DOE-ORP
J. Matyáš, PNNL
D. J. McCabe, 773-42A
D. L. McClane, 999-W
R. T. McNew, 766-H
D. H. Miller, 999-W
D. K. Peeler, PNNL
F. M. Pennebaker, 773-42A
M. R. Poirier, 773-42A
J. W. Ray, 704-27S
M. L. Restivo, 773-42A
A. Samadi-Dezfouli, 704-27S
M. J. Schweiger, PNNL
M. E. Stone, 999-W
C. L. Trivelpiece, 999-W
J. D. Vienna, PNNL
J. R. Vitali, 704-30S
B. J. Wiedenman, 773-42A
W. R. Wilmarth, 773-A
Records Administration (EDWS)