U.S. DEPARTMENT OF ENERGY

PNNL-26330, EWG-RPT-014 Rev. 0

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

Vitrification of HWI-Al-19 Glass in Research-Scale Melter

JS Hardy
GJ Sevigny
ML Kimura
DR Dixon
JJ Neeway
CD Lukins
PP Schonewill
WC Buchmiller
MR Zumhoff
RP Pires
MP Portch
CM Fischer
BD Williams
CD Johnson
JB Lang
CP Rodriguez
MJ Schweiger

September 2015

Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by Battelle *Since 1965*

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC05-76RL01830

Printed in the United States of America

**Available to DOE and DOE contractors from the
Office of Scientific and Technical Information,
P.O. Box 62, Oak Ridge, TN 37831-0062;
ph: (865) 576-8401
fax: (865) 576-5728
email: reports@adonis.osti.gov**

**Available to the public from the National Technical Information Service
5301 Shawnee Rd., Alexandria, VA 22312
ph: (800) 553-NTIS (6847)
email: orders@ntis.gov <http://www.ntis.gov/about/form.aspx>
Online ordering: http://www.ntis.gov**

This document was printed on recycled paper.

(8/2010)

PNNL-26330
EWG-RPT-014, Rev. 0

Vitrification of HWI-Al-19 Glass in Research-Scale Melter

JS Hardy
GJ Sevigny
ML Kimura
DR Dixon
JJ Neeway
CD Lukins
PP Schonewill
WC Buchmiller
MR Zumhoff
RP Pires
MP Portch
CM Fischer
BD Williams
CD Johnson
JB Lang
CP Rodriguez
MJ Schweiger

September 2015

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

Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

This test report describes the experimental results from small-scale tests using the research-scale melter (RSM) at Pacific Northwest National Laboratory to demonstrate processing of a simulated feed stream in 6- and 10-inch diameter melt chambers, referred to hereafter as RSM-6 and RSM-10, respectively, according to the PNNL test plan, *RSM Vitrification of Simulated WTP High Level Waste*, and PNNL operating procedure, *Research Scale Melter Safe Operating Procedure* (SOP-80). This test vitrified HWI-Al-19, a high alumina high level simulated waste and compared feed processing rates at standard bubbling rates to larger-scale melter tests conducted at the Vitreous State Laboratory (VSL) at The Catholic University of America (CUA). The bubbling rate was varied to monitor its effect on feed processing rate. The test also verified glass quality through microscopic examination and chemical analyses of glass products and characterization of glass properties of interest, including liquidus temperature, durability, electrical conductivity, and viscosity. In the cases of liquidus temperature, electrical conductivity, and viscosity, a direct comparison of the properties of glasses produced by the melters at the two research facilities could not be made because properties reported by VSL came from crucible melts rather than from glass produced by their DM melters. Thus, for these properties, the glasses produced by the RSM at PNNL were compared to the crucible melts reported by VSL to provide a general assessment of conformity.

The RSM is a small, joule-heated melter capable of processing melter feed on a continuous basis. The melter is equipped with Inconel® 693 electrodes, Monofrax® K-3 refractory, and an Inconel 690 pour spout. For the experiments described here, an electric kiln surrounded the melter body and minimized heat loss from the melter body during operation. The RSM was equipped with an off-gas treatment system that employed quenching, wet scrubbing, and high-efficiency mist elimination. The glass-discharge section was heated to facilitate pouring of the glass. The melter was fitted with melt cavities that were ~15 and ~25 cm (6 and 10 in.) in diameter with a nominal glass depth of 7.6 cm (3 in.). The melters were operated with a target glass temperature of 1150°C and plenum temperature between 350 and 650°C.

The RSM tests were broken into three segments to determine the maximum processing rates achievable at different bubbling rates and in two different melter cavities with different melt volumes. The test segments were

- RSM-6 melter, 1.4–1.5 L/min bubbling rate
- RSM-6 melter, 0.7 L/min bubbling rate
- RSM-10 melter, 4.1 L/min bubbling rate.

Overall, 64 kg and 101 kg of glass were produced during operation of the RSM-6 and RSM-10, respectively.

At the conclusion of the test, the melter and exhaust lines were visually inspected for particulate deposition and corrosion. Entrained material had adhered to the underside of the melter lid and to the exhaust piping. Enrichments in elements such as S, Cr, P, and Ni were discovered in these deposits through inductively coupled plasma–optical emission spectroscopy and X-ray fluorescence analysis. When the melter electrodes and bubbler tube were removed from the glass in the RSM, the electrodes appeared discolored, but no significant loss of metal was observed.

The processing of HWI-Al-19 in the RSM melter was similar to tests performed in larger joule-heated melters. The average glass production rate was from 0.71 to 0.82 kg/h in RSM-6 and 1.95 kg/h in RSM-10 resulting in a melter-surface-area normalized glass generation rate of 922 to 1077 kg/day/m^2 for all RSM tests, which falls within the range of 896 to 1557 kg/day/m^2 reported for the VSL melters. (Matlack et al. 2008) As expected, a higher bubbling rate caused an increase in the maximum glass production rate. The RSM product glass composition met the same criteria of conformity to the target composition as VSL reported for the DM glass. It also passed the toxic characteristic leaching procedure (TCLP) requirement and product consistency test (PCT) requirement for both quenched and canister centerline cooling (CCC)-treated glasses, with lower PCT performance for CCC-treated glasses likely caused by crystallization. The glasses at VSL also passed TCLP and PCT. The TCLP concentrations of the RSM glass were 0.03-1.27 mg/L higher than those of the VSL glass while the PCT normalized releases were 0.02-0.11 g/m^2 lower. Comparisons to the properties measured on crucible melts at VSL find that the liquidus temperature of the RSM glass was ~187°C higher, while the electrical conductivity and viscosity measurements as a function of temperature were very similar in both magnitude and slope.

Acknowledgments

This work was undertaken with funding authorized by the Federal Project Director William F. Hamel, Jr. for the Waste Treatment and Immobilization Plant. Albert A. Kruger, Glass Scientist, Office of River Protection, U.S. Department of Energy (DOE) provided approval and oversite. Authors would like to thank Dong Sang Kim and David K. Peeler for their insightful discussions and comments.

Pacific Northwest National Laboratory is operated by Battelle Memorial Institute for the U.S. Department of Energy under contract DE-AC05-76RL01830.

Acronyms and Abbreviations

ASTM	American Society for Testing and Materials
CCC	canister centerline cooling
DAC	data acquisition and control
DOE	U.S. Department of Energy
DWPF	Defense Waste Processing Facility
EDS	energy dispersive spectroscopy
EVS	ejector venturi scrubber
HASQARD	Hanford Analytical Services Quality Requirements Document
HDI	PNNL's standards-based management system—*How Do I*?
HEME	high-efficiency mist eliminator
HLW	high-level waste
IC	ion chromatography
ICP-OES	inductively coupled plasma–optical emission spectroscopy
JHM	joule-heated melter
LSM	laboratory-scale melter
PCT	product consistency test
PNNL	Pacific Northwest National Laboratory
RSM	research-scale melter
SEM	scanning electron microscope
SRNL	Savannah River National Laboratory
SwRI	Southwest Research Institute
TCLP	toxicity characteristic leaching procedure
VSL	Vitreous State Laboratory
WTP	Hanford Tank Waste Treatment and Immobilization Plant
XRD	X-ray diffraction
XRF	X-ray fluorescence

Contents

Figures

Tables

1.0 Introduction

Resolution of the nation's high-level nuclear waste legacy requires the design, construction, and operation of large and technically complex one-of-a-kind processing waste treatment and vitrification facilities. Vitrification technology was chosen to treat three types of waste: the high-level waste (HLW) fraction of tank waste at the U.S. Department of Energy's (DOE's) Hanford and Savannah River Sites, the low-activity waste fraction of tank waste at Hanford, and potentially other defense waste streams such as the sodium-bearing tank waste or calcine HLW at Idaho National Laboratory (INL). Joule-heated melters (JHMs) are being used at the Defense Waste Processing Facility (DWPF) and will be used at the Hanford Tank Waste Treatment and Immobilization Plant (WTP) to vitrify tank waste fractions.

This test report describes the experimental results from small-scale tests using the research-scale melter (RSM), a JHM at Pacific Northwest National Laboratory (PNNL). The RSM includes Inconel® 693 plate electrodes and Monofrax® K-3 as the glass contact refractory. The melt cavities are 15.2 cm (6 in.) and 25.4 cm (10 in.) in diameter and 8.9 cm (3.5 in.) deep. The results described herein were the product of the first two of five tests planned for fiscal year 2013. In these tests, processing rate combined with bubbling rates and overall control of melter operating parameters in the 6-inch and 10-inch cavities of the RSM were compared to scaled testing that was previously performed with the same nonradioactive simulant slurry (HWI-Al-19) by the Vitreous State Laboratory (VSL) at the Catholic University of America (CUA) in Washington, DC (Matlack et al. 2008; Joseph et al. 2010). Primary comparisons were production rate and glass quality. Glass quality was evaluated by composition, crystallinity, and product consistency test (PCT) and toxicity characteristic leaching procedure (TCLP) analyses of a limited number of melter glass samples. The purpose of this study and the comparisons with previous tests conducted in larger-scale melters at VSL was to demonstrate that a smaller-scale RSM system can provide data to support the WTP programmatic objectives for glass processing operations.

2.0 Objectives

The primary objective of the RSM-6 and RSM-10 tests with HWI-Al-19 glass feed was to evaluate processing rates versus operating conditions in the RSM system and develop data for comparison with previous tests conducted with larger-scale melters and thus demonstrate that a smaller-scale RSM system can provide data to support the WTP programmatic objectives for glass processing operations.

Data required for such an assessment included, but were not limited to, successful production of a durable, compliant glass with an acceptable waste loading at design production rates without causing excessive corrosion of JHM components or operating instability.

The HWI-Al-19 glass feed used in this study was previously processed in the DM-100 and DM-1200 melters at VSL (Matlack et al. 2008; Joseph et al. 2010), facilitating a comparison of melter operation and glass properties between these and the RSM.

The specific objectives of the study are listed below:

- Complete at least three melter volume turnovers under typical melter operating conditions.
- Obtain steady-state operations to determine processing rate and melter operating characteristics and evaluate differences from VSL results.
- Collect and analyze samples from feed, glass, and off-gas for comparison to corresponding VSL samples.
- Verify quality of glass produced through X-ray diffraction (XRD), PCT, TCLP, and microscopic analyses.

3.0 RSM System Description

The RSM processing system used to support the objectives of this study is described in this section. The Process Development Laboratory East building located at PNNL in Richland, Washington, housed the RSM system, for which a schematic is given in Figure 3.1, showing liquid lines in blue and gas lines in black.

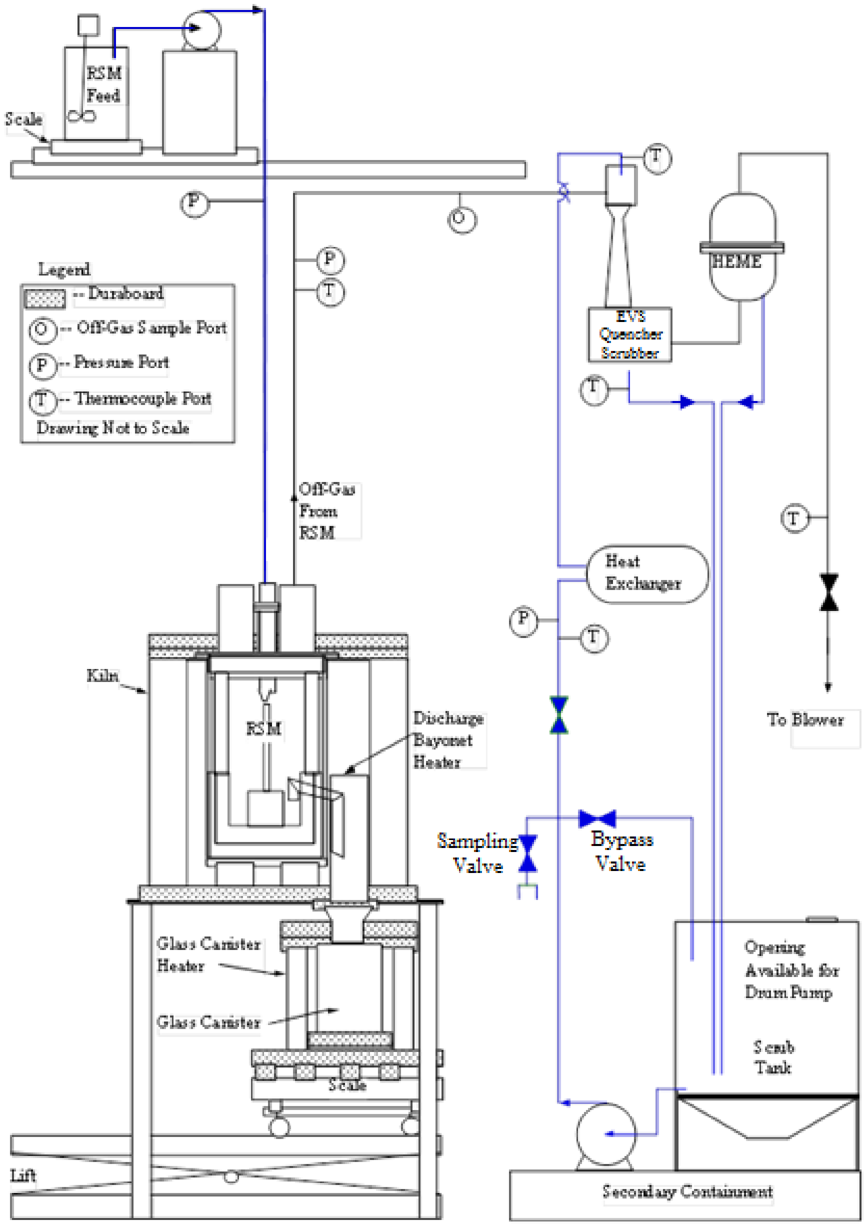


Figure 3.1. Research-Scale Melter Process Flow Diagram

The RSM is a small JHM capable of processing melter feed on a continuous basis, which is a key capability to be representative of a full-scale melter system. Testing in the RSM allows parametric studies to be conducted in a relatively short time. The RSM processing system provides unit off-gas treatment operations of quenching, wet scrubbing, and high-efficiency mist elimination. The off-gas port contains a simple film cooler fabricated from a sintered metal filter that allows injected air to pass from the outside of the filter through the sintered metal into the off-gas line while melter exhaust gas passes through the middle of the filter, combining with the injected air. The aqueous quench scrubber is an ejector venturi scrubber (EVS), previously shown to be functionally equivalent to the WTP submerged-bed scrubber (SBS) technology (Goles and Schmidt 1992). The exhaust of the RSM EVS is treated by a high-efficiency mist eliminator (HEME) that not only demists the influent stream but efficiently removes sub-micron aerosol matter penetrating the EVS. Table 3.1 provides RSM dimensions and other operational features.

Table 3.1. RSM Dimensions and Operational Features

Parameter	RSM-6	RSM-10
Melter cavity diameter	15.2 cm	25.4 cm
Melt surface area	172 cm^2	500 cm^2
Melter cavity height	17 cm	17 cm
Melter internal volume	3.1 L	8.6 L
Nominal glass melt depth	8.9 cm	8.9 cm
Nominal glass melt volume	1.6 L	4.5 L
Maximum operating temperature	1200°C	1200°C
Nominal operating temperature for borosilicate glass	1150°C	1150°C
Bubbler dimensions	¼" OD[(a)] Tubing	¼" OD Tubing
Bubbler material	Inconel 690	Inconel 690 & 625
Electrode dimensions (W × H × T)	7.6 × 7.6 × 0.9 cm	11 × 10 × 0.9 cm
Electrode material	Inconel 693	Inconel 693
Electrode distance from bottom	0 cm	0 cm
Electrode current (average)	75 A	90 A
Electrode voltage (average)	40 V	60 V
Electrode current density (average/maximum)	1.3/2.0 A/cm^2	0.5/2.0 A/cm^2

(a) OD = outer diameter

RSM Melter: The body of the RSM is an Inconel 625 closed-ended cylinder lined with Alfrax® refractory that contains a Monofrax K3 refractory melt cavity. A cross-sectional diagram of the RSM-6 is shown in Figure 3.2, the RSM-10 is similar except with a large melt cavity of 10 inches diameter. An Inconel pour spout tube discharges molten glass into a stainless steel canister. An electric kiln surrounds the melter body to heat the melter during startup and to minimize heat loss from the melter body during operation by decreasing the temperature gradient across the melt chamber walls. The discharge section is heated to facilitate pouring of the glass. The stainless steel canister sits inside a smaller kiln maintained between 700 and 900°C to promote uniform canister filling. Two Inconel 693 electrodes enter the melter through ports in the lid and are suspended in the glass to supply joule-heating power to the RSM. Accounting for the areas displaced by the electrodes and bubblers, the melt surface areas of the RSM-6 and RSM-10 are 172 and 500 cm^2, respectively, with a nominal glass depth of 8.9 cm, resulting in corresponding glass inventories of 4 and 11 kg, respectively, assuming a glass density of 2.5 g/cm^3 at 1150°C. The melter is controlled using the data acquisition and control (DAC) system, which allows temperature or power control. Temperature control was typically employed during these tests, with alarms set to alert the operators if temperature or power stray outside of their predefined ranges.

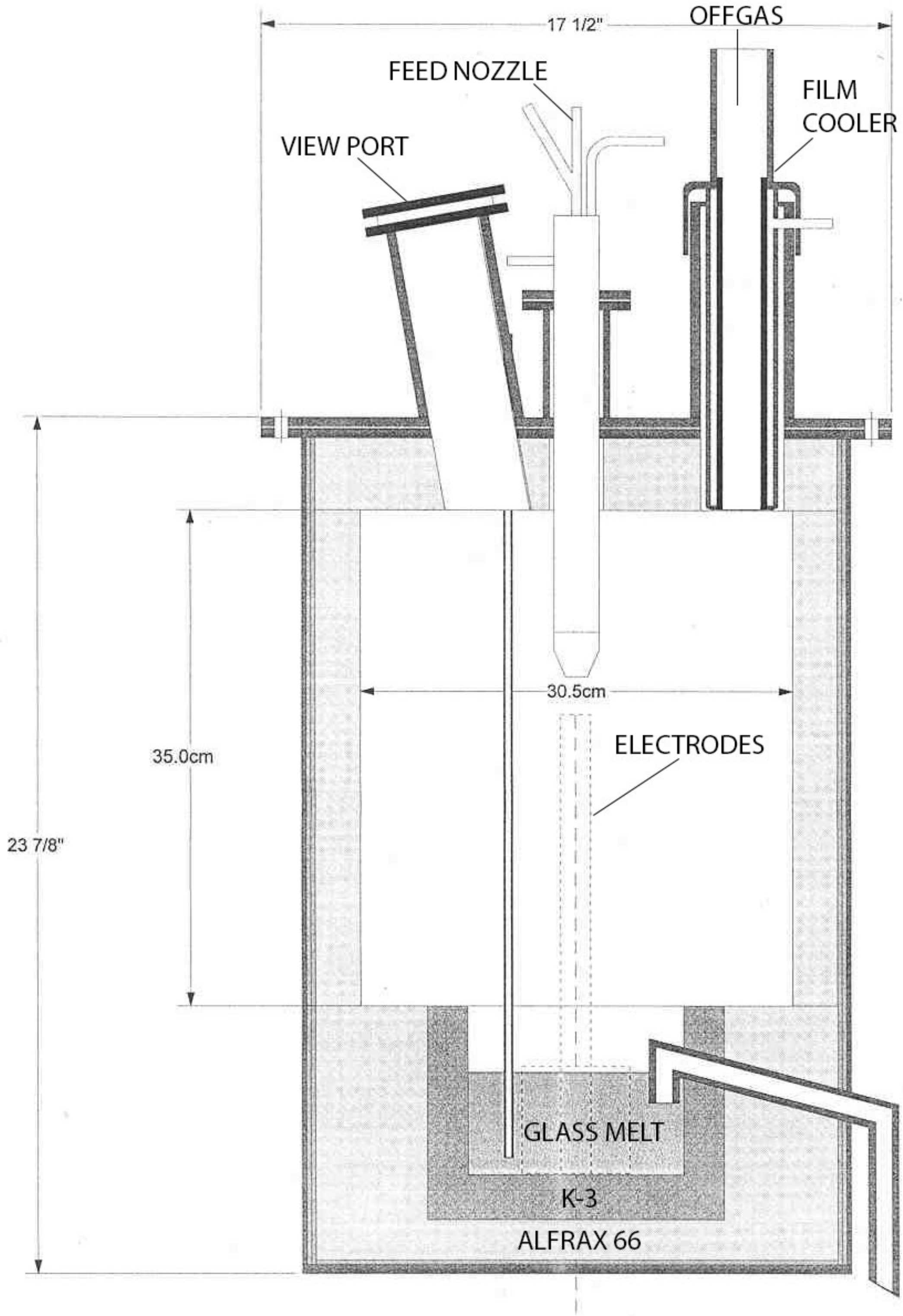


Figure 3.2. Cross-Sectional Diagram of the RSM-6

RSM Feed System: The HWI-Al-19 melter feed was shipped from the supplier to PNNL in 55-gallon drums and subsequently transferred into the feed tank. During the tests, feed is delivered from the conical bottom of the feed tank to the RSM feed nozzle by a set of peristaltic pumps. One pump recirculates the slurry to a point near the melter to keep solids in the feed line suspended and the second pump controls the flow of the slurry to the melter feed nozzle. An agitator in the feed tank keeps the slurry well mixed. The feed tank is mounted on a scale that is monitored by the computer DAC system. The speed of the second pump was used to control the feed rate to the melter.

RSM Off-gas System:

Ejector Venturi Scrubber **(EVS):** The EVS sprays scrubber solution through a nozzle for direct contact with the melter exhaust (Figure 3.1). At the beginning of a melter run, the scrubber solution consists only of water. As the melter operates, the EVS condenses water from the melter exhaust and removes particulates and some acid gases. The resulting two-phase stream travels through a separator chamber and the scrubber solution returns to the scrub tank under the force of gravity (Figure 3.1). The scrubber solution is recirculated from the scrub tank with a pump located adjacent to the RSM platform and through a heat exchanger to remove the heat transferred from the melter exhaust. From the scrubber, the exhaust passes through a HEME to remove condensed-phase aerosols. Quench-scrubber samples are collected periodically during the test.

4.0 Test Conditions

To satisfy the technical objectives of this process flowsheet, the goal was to produce at least three melt volume turnovers using the RSM. The process condition targets used during testing are described below and shown in Table 4.1.

Table 4.1. Target RSM Operating Conditions

Parameter	RSM-6	RSM-10
Glass inventory[a]	4 kg	11 kg
Glass melt temperature	1150°C	1150°C
Plenum temperature range	350–650°C	350–650°C
Plenum pressure	−0.5 to −1.5 in. water	−0.5 to −1.5 in. water
Post-film-cooler temperature range	200–350°C	200–350°C
Melt bubbling rate	1.4 L/min	4.1 L/min
Initial scrub solution volume	80 L	80 L
Melt condensate pH	<2	<2

(a) Assuming a glass density of 2.5 g/cm^3 at 1150°C.

4.1 Process Conditions

The major process conditions that were controlled were glass pool temperature, melter vacuum, melt pool bubbling rate, feed composition, processing rate, plenum temperature, off-gas temperature, and quench-scrubber condensate temperature. Strategies for maintaining baseline conditions are discussed below.

Glass Pool Temperature: The 1150°C target temperature was automatically controlled by the RSM DAC system. The electrode current density was constrained to ≤2 A/cm^2 (~115 A and 190 A for the RSM-6 and -10, respectively) to prevent excessive corrosion of electrodes. If the electrode current density were to become a constraint in maintaining the target glass temperature, the kiln temperature could be adjusted to mitigate heat loss from the melt. The thermocouples monitoring the melt temperature are located within the melter electrodes.

Melter Vacuum: The RSM blower is capable of providing up to 28 in. water gauge vacuum (at 200 cfm). The RSM melter pressure was automatically controlled at a set point, nominally between 0.5 and 2 in. water gauge below ambient conditions. Adjustments to the vacuum pressure were made to instigate and postpone glass pours.

Melt Pool Bubbling Rate: Glass pool agitation using subsurface air injection was employed to enhance melter feed processing rates. To accomplish this, a flow meter delivered air into the RSM-6 at 0.7 L/min to 1.5 L/min through one submerged end of a straight Inconel 690 tube which entered the melt from the top of the melter. The RSM-10 had air delivered at 4.1 L/min, divided between one Inconel 690 tube and one Inconel 625 tube which entered the melt from the top of the melter. The accuracy of the flow meter was such that the bubbling rates in the RSM-6 were within ± 0.075 L/min of the rates indicated by the flow meter, whereas in the RSM-10, they were within ± 0.050 L/min.

Feed Composition: A single feed composition, HWI-Al-19 (see Table 4.2 and Table 4.3), was used throughout the tests.

Processing Rate: Steady-state feed processing rates for the melter were controlled based on cold-cap conditions, which were visually observed through a view port in the lid of the melter at least once every hour and also obliquely monitored by tracking the plenum temperature. The target cold-cap coverage was 85 to 95 percent. To achieve glass processing rates within the expectation range of 0.7 to 1.1 MT/day/m^2, feed rates of 1.0 to 1.6 L/h and 2.9 to 4.6 L/h were necessary in the RSM-6 and RSM-10, respectively.

Plenum Temperature: The targeted plenum temperature range was 500 ± 150°C during periods when maximum feeding rates are sustained. While plenum temperature is not directly controlled, inleakage, melter kiln temperature, and bubbling rate all influence it under steady-state processing conditions (85 to 95 percent cold-cap coverage).

Off-Gas Temperature: The post-film-cooler off-gas temperature was constrained to <350°C to prevent the off-gas lines from becoming plugged by particulate deposition. The film-cooler air injection rate was used to control the temperature of the off-gas.

Quench-Scrubber Solution Temperature: The expected EVS solution temperature was ~30 to 45°C. If there was a need to increase or decrease this temperature, the cooling flow rate of the condensate heat exchanger was adjusted appropriately.

After changes in operating parameters, some operating time was needed to allow the glass melt to approach a new equilibrium. After bubbling rate changes, a significant amount of time was required to find the appropriate feed rate for reaching steady-state conditions. Stability can be difficult to assess because plenum temperatures have normal fluctuations and cold-cap observations are subjective. For the present tests, conditions were required to be stable for >5 h to declare the system at steady state. Many feed rates were tested in order to determine the maximum sustainable rate.

4.2 Simulated Waste and Melter Feed

Table 4.2 summarizes the Al-limited waste and HWI-Al-19 glass compositions. Table 4.2 also shows the relative proportions of the glass formers used and the resultant target glass composition to be prepared during melter testing. The simulated feed recipe to produce 100 kg of HWI-Al-19 glass with a target glass yield of 500 g/L is shown in Table 4.3. The composition of the feed mixture was the same as that tested in VSL melters (Matlack et al. 2008; Joseph et al. 2010) and the simulated feed was purchased from the same supplier with instruction to prepare it in the same way that it was prepared for VSL. Sampling of the melter feed stream during the test provided for post-test analytical validation of feed compositions.

Table 4.2. HWI-Al-19 Glass Composition (from Matlack et al. 2008)

Component	Al-Limited Waste[a]	Waste in Glass	Glass-Forming Additives	Target Glass HWI-Al-19
Al_2O_3	53.27	23.97	-	23.97
B_2O_3	0.42	0.19	19.00	19.19
BaO	0.12	0.05	-	0.05
Bi_2O_3	2.54	1.14	-	1.14
CaO	2.39	1.08	4.50	5.58
CdO	0.05	0.02	-	0.02
Cr_2O_3	1.16	0.52	-	0.52
F	1.48	0.67	-	0.67
Fe_2O_3	13.11	5.90	-	5.90
K_2O	0.31	0.14	-	0.14
Li_2O	0.38	0.17	3.40	3.57
MgO	0.26	0.12	-	0.12
Na_2O	7.96	3.58	6.00	9.58
NiO	0.89	0.40	-	0.40
P_2O_5	2.34	1.05	-	1.05
PbO	0.91	0.41	-	0.41
SO_3	0.44	0.20	-	0.20
SiO_2	10.88	4.90	22.10	27.00
TiO_2	0.02	0.01	-	0.01
ZnO	0.18	0.08	-	0.08
ZrO_2	0.88	0.39	-	0.39
Sum	100.0	45.00	55.00	100.0[b]

(a) Renormalized from (DOE 2006) after removal of radioactive components.
(b) The sum does not equal 100.00 because of rounded decimals.

Table 4.3. Simulant Recipe (from Matlack et al. 2008)

Al-Limited Waste Simulant		Glass-Forming Additives	
Starting Materials	**Target Weight (kg)** [(a)]	**Starting Materials**	**Target Weight (kg)**[(a)]
$Al(OH)_3$	37.047	-	-
H_3BO_3	0.341	H_3BO_3	34.089
$BaCO_3$	0.070	-	-
Bi_2O_3	1.156	-	-
CaO	1.099	$CaSiO_3$ (Wollastonite)	9.798
CdO	0.025	-	-
Cr_2O_3	0.532	-	-
NaF	1.483	-	-
$Fe(OH)_3$ (13% Slurry)	48.539	-	-
KNO_3	0.308	-	-
Li_2CO_3	0.432	Li_2CO_3	8.625
MgO	0.121	-	-
NaOH	2.190	Na_2CO_3	10.364
$Ni(OH)_2$	0.514	-	-
$FePO_4{\bullet}2H_2O$	2.795	-	-
PbO	0.413	-	-
Na_2SO_4	0.358	-	-
SiO_2	4.945	SiO_2	17.276
TiO_2	0.010	-	-
ZnO	0.084	-	-
$Zr(OH)_4{\bullet}8H_2O$	1.020	-	-
H_2O	91.903	-	-
Na_2CO_3	0.314	-	-
$NaNO_2$	0.346	-	-
$NaNO_3$	0.984	-	-
$H_2C_2O_4{\bullet}2H_2O$	0.119	-	-
Simulant Total	197.148	Additives Total	80.152
-	-	FEED TOTAL	277.300
(a) Target weights adjusted for assay information of starting materials.			

5.0 Run Description

Prior to melting in the RSM, a preliminary melt of the HWI-Al-19 feed was performed in a laboratory-scale melter (LSM) at 1150°C to evaluate the melting characteristics and resulting glass product. The LSM consists of a 10 cm diameter quartz melting chamber that is top-loaded into a laboratory furnace (Kim et al. 2011b). Unlike the RSM, there is no bubbler to agitate the melt in the LSM. Before placing the melt chamber into the furnace, the bottom of the chamber was covered with a layer of A0 glass (Schweiger et al. 2010). This was used as a starter glass because it has a composition similar to that of HWI-Al-19. After the chamber was lowered into the furnace and the starter glass had melted, the HWI-Al-19 feed was introduced into the quartz chamber at rates between 3 and 6.5 ml/min using a peristaltic pump. The feed rates of the peristaltic pump were calibrated by operating the pump over a wide range of rotation speed settings and measuring the average feed rate at each setting. The melt was completed with no difficulties, and examination of the resulting glass under an optical microscope found little to no crystallization.

5.1 Test Segments

The RSM tests were broken into three segments to determine the maximum processing rates achievable at different bubbling rates and in two different melter cavities with different melt volumes. The test segments were

- RSM-6 melter, 1.4–1.5 L/min bubbling rate
- RSM-6 melter, 0.7 L/min bubbling rate
- RSM-10 melter, 4.1 L/min bubbling rate.

The glass in all test segments was melted under similar operating conditions, as discussed in Section 4.0 and shown graphically in Appendix A. A summary of the main operating parameters during each segment (“Seg.”) is given in Table 5.1.

Table 5.1. Summary of RSM Steady-State (SS) Operations

Melter	Seg. #	Seg. Start Date (2013)	Seg. Start Time	Total Hours of Normal Operation	Avg. SS Plenum Temp (°C)	Avg. SS Melt Temp (°C)	Bubbling rate (L/min)	Avg. SS Feed Rate (L/h)	Avg. Seg. Glass Pour Rate (kg/h)	Max. Sustained Production (kg/day/m^2)[(a)]
RSM-6	1	4/15	0948	52.2	641	1150	1.4–1.5	1.57	0.82	1159
RSM-6	2	4/17	1626	26.5	653	1155	0.7	1.49	0.71	1008
RSM-10	1	5/20	0709	33.3	588	1117	4.1	3.62	1.95	968

(a) Based on the surface area of the glass melt

The RSM operating segments were targeted to have at least three melter turnovers of glass and a prolonged period of operation at a consistent feed rate. The operation of the melter was sufficient to satisfy the melter-related objectives and to obtain basic operational data. The plenum temperatures ranged between 537 and 732°C during steady-state operation; these were higher than the 350 to 650°C target range given in the test plan. Plenum temperatures were only above 700°C for three minutes while electrode power ramped back up after having been shut off for three minutes to knock excess feed material off of the feed nozzle during the second segment in the RSM-6. Less than one-third of the plenum temperature measurements were above the 650°C maximum. VSL also reported plenum temperatures outside their target range (Matlack et al. 2008). They reported plenum temperatures ranging from 382 to 782°C during steady-state operation, which was a slightly wider range than was measured in the RSM and overstepped the upper and lower bounds of their 450 to 650°C targets. Thus, the high plenum temperatures measured in the RSM do not cause any issue in the comparisons made with VSL melters. Because plenum temperature is not directly controlled, but is instead dependent upon melter design and operational events, it is more likely than other parameters to stray outside intended limits. The maximum steady-state feed rate was attempted in each segment, but additional time was not taken to painstakingly optimize it. Meanwhile, the north and south electrode temperatures were relatively stable during each segment, with no electrode temperature data having a standard deviation greater than 15°C during any segment. Likewise, the standard deviation of the supplied power data was less than 0.9 kW for each segment, meaning good temperature control was maintained without large swings in electrode power. Feed-line and feed-nozzle plugging problems were the primary cause of disruption during processing and the main obstacle that had to be overcome in obtaining a steady equilibrium melt rate. Interruptions in feeding to clear these plugs in the feed nozzle totaled less than two hours for each test segment and did not prevent achieving steady-state conditions. There was no foaming in the glass discharge or on the melt pool surface. The cold-cap coverage was continuously changing throughout the tests. However, during the majority of testing it remained within the target range. The feed did not seem to settle in the feed tank, as evidenced by a lack of material in the bottom of the tanks when the tanks were emptied after test completion.

RSM-6. Before the actual feed processing was initiated, the melter was loaded with start-up glass A0 (Schweiger et al. 2010) and heated with the melter kiln heater to bake out the castable refractory installed behind the Monofrax K-3. The A0 glass was prepared at PNNL and supplemented with an additional 240 g H_3BO_3 and 10 g Na_2CO_3 to closely match the composition of HWI-Al-19. The kiln heater was operating at 95 percent power, and the glass temperature was 923°C before joule heating was started at 0758 hours on Thursday, April 4, 2013. The melter reached the targeted glass temperature of 1150°C at 0856 hours on Friday, April 5, 2013. The melter was then set to 1075°C and allowed to idle at this temperature for several days until the scheduled start date for the first test. Shakedown testing with feed began on Wednesday, April 10, 2013, at 1309 hours and continued until 1710 hours. The melter was then restarted on Monday, April 15, 2013 and testing operations were initiated at 0931 hours, although refinements to the processing continued during testing, which produced a total of 63.8 kg of glass. During testing, erroneously fluctuating temperature readings were observed on several of the thermocouples including the pour spout, discharge canister, kiln, off-gas, heat exchanger, and process water thermocouples. This was corrected by unplugging the pour spout thermocouple from the DAC system and instead reading it on a handheld device while continuing to collect the other temperature measurements using the data acquisition system. Apparently the pour spout thermocouple had been a source of interference causing the fluctuating temperature readings in the entire group of affected thermocouples. Plots of melter power, melter feed rate, plenum temperature, and glass temperature are shown in Figure 5.1 and Figure 5.2.

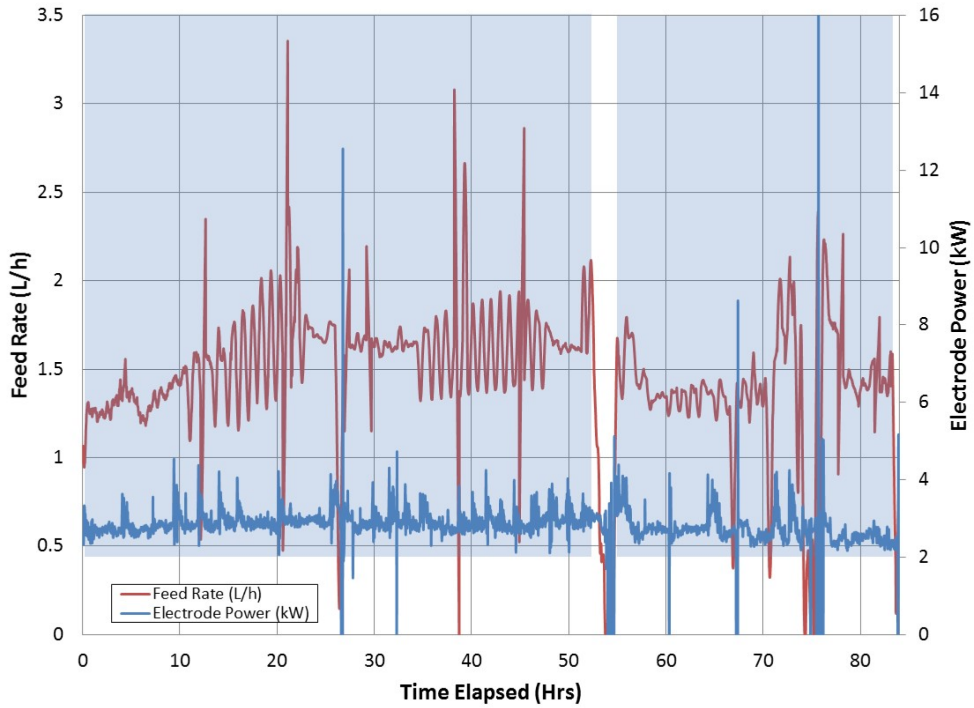


Figure 5.1. Feed Rate (30 min average) and Electrode Power in the RSM-6. Test segments are blue shaded regions.

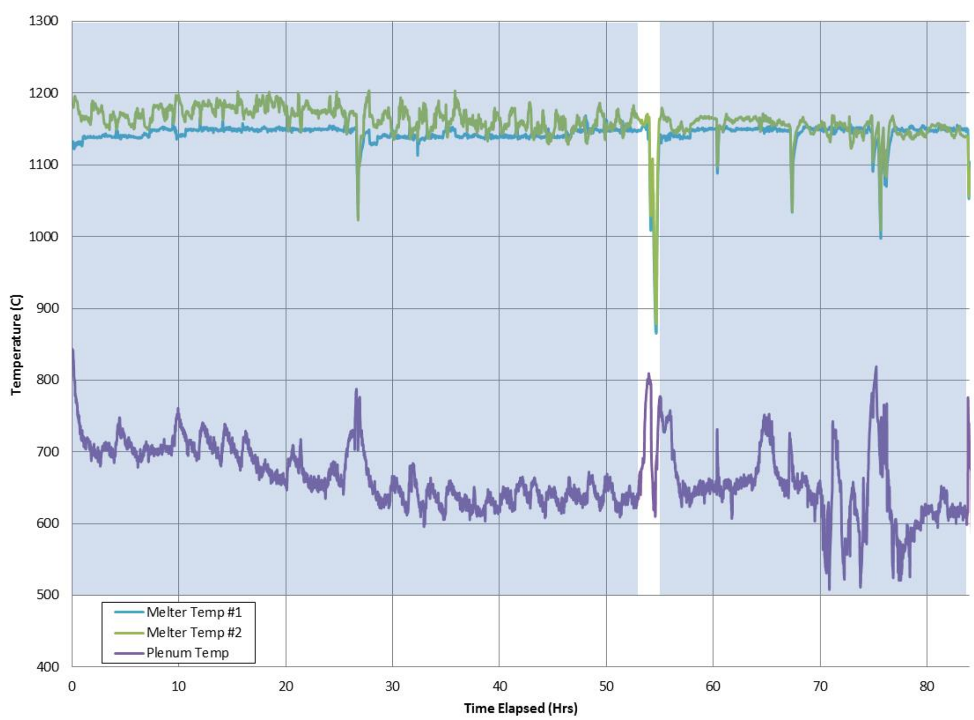


Figure 5.2. Melter Glass and Plenum Temperatures in the RSM-6. Test segments are shaded in blue.

RSM-6 Segment 1. Segment 1 (1.4–1.5 L/min bubbling rate) of the RSM-6 test commenced on April 15, 2013, at 0948 hours with an initial feed rate of 1.3 L/h. During this segment, the initial bubbling rate was 1.5 L/min and was decreased to 1.4 L/min after 33–34 hours. The feed rate was gradually increased during testing until a maximum rate of slightly above 1.7 L/h was achieved. About midway through this segment, on April 16, 2013 at 1159 hours, feeding was interrupted for 26 minutes to remove a blockage in the feed nozzle. The average glass temperature was 1150°C and the average melt-surface-area–adjusted power supplied to the electrodes was 160 kW/m^2. The average plenum temperature during steady-state operation was 641°C, which was at the high end of the target range of 350–650°C. The cold cap grew to cover the entire surface of the molten glass pool with the exception of two holes through which the bubbler gas exited the melt (Figure 5.3). After several of the periods during which glass pours took place, it was observed that the cold cap had disappeared even though feeding was uninterrupted. The cold cap would then begin to regenerate and expand in coverage until only the two bubble vents remained uncovered by cold cap. It is believed that the decrease in glass depth associated with pours caused the cold cap to sink below the surface of the melt. Over 117 kg of feed was processed during segment 1 and the average feed rate during stable processing was 1.57 L/h. The overall average feed rate, including feed outages, was 1.56 L/h with a glass pour rate of 0.82 kg/h (1077 $kg/day{\cdot}m^2$). Calculations of the glass processing rates that were determined based on the weight of poured glass collected were complicated by the fact that the pour events were cyclic in response to melter level, plenum vacuum and cold-cap behavior. For this reason, supplemental calculations of production rates were performed based on measured feed rates for comparison.

Figure 5.3. Photograph of the Cold Cap in RSM-6 Test Segment 1

RSM-6 Segment 2. For segment 2 (0.7 L/min bubbling rate) of the RSM-6 test, the bubbling rate was reduced on April 17, 2013, at 1626 hours to determine the effect that decreased bubbling would have on the feed rates that could be accommodated. This segment ran for about 27 h and the decrease in bubbling rate resulted in a slight decrease in the feed rate to 1.49 L/h. The overall feed rate including feed outages was 1.37 L/h with a glass pour rate of 0.71 kg/h (939 $kg/day/m^2$). During this segment there were two feed interruptions. The first began a little more than 12 h into the segment at 0438 hours on April 18, 2013, and continued for 11 minutes to clear a blockage in the feed line. The second began a little more than 7 h after the first at 1147 hours on April 18, 2103, and continued for 79 minutes to clear repeated feed blockages. Over 56 kg of feed was processed during this test segment. The average melt temperature was 1150°C and the average melt-surface-area–adjusted electrode power was 149 kW/m^2. The average plenum temperature was 653°C during steady-state feeding, which was slightly above the 650°C maximum. Meanwhile, the same cold-cap behavior that was described for segment 1 was also observed at the lower bubbling rate employed in segment 2. After glass pours, the cold cap essentially disappeared and would then regenerate between pours. This is shown in Figure 5.4 with three frames that were captured from a video at 2.5-minute intervals.

Figure 5.4. Frames Captured from a Video Taken during RSM-6 Test Segment 2 at 2.5-Minute Intervals (a) Before and (b & c) After a Glass Pour

RSM-10. Before the actual feed processing was initiated, the melter was loaded with 6.1 kg of HWI-Al-19 glass that had been poured during the preceding RSM-6 test. The melter was heated with the melter kiln heater to bake out the castable refractory installed behind the Monofrax K-3. The kiln heater was operating at 26 A, and the glass temperature was 840°C before joule heating was started at 0756 hours on Friday, May 17, 2013. Shakedown testing with feed began on Monday, May 20, 2013, at 0605 hours and continued until testing operations were initiated at 0709 hours, although refinements to the processing continued during testing, producing a total of 101 kg of glass. At 0700 hours on May 22, 2013, it was discovered that there was no discernible motion of the glass surface near one of the bubblers, suggesting that the bubbler had failed. Consequently, the bubbler tube in question was manually pushed deeper into the melter. Bubbling activity was not observed until the bubbler had been lowered an additional ~12 inches. It was found that 8–12 inches of the bubbler tube had indeed broken off and was recovered from the melter during a brief power down. Subsequent inspection of the tube revealed that the corrosion and failure had occurred above the melt line. The precise time at which the bubbler failed is unknown. Chemical analyses by X-ray fluorescence (XRF) conducted after testing indicated that the failed bubbler was fabricated from Inconel 625 instead of Inconel 690, based on the Ni and Mo contents. Plots of melter power, melter feed rate, plenum temperature, and glass temperature are shown in Figure 5.5 and Figure 5.6. At 2105 hours on Wednesday, May 22, 2013, a blown fuse brought an abrupt stop to the current applied to the melter and resulted in faster cooling of the melter and its contents than occurs under normal shutdown procedures.

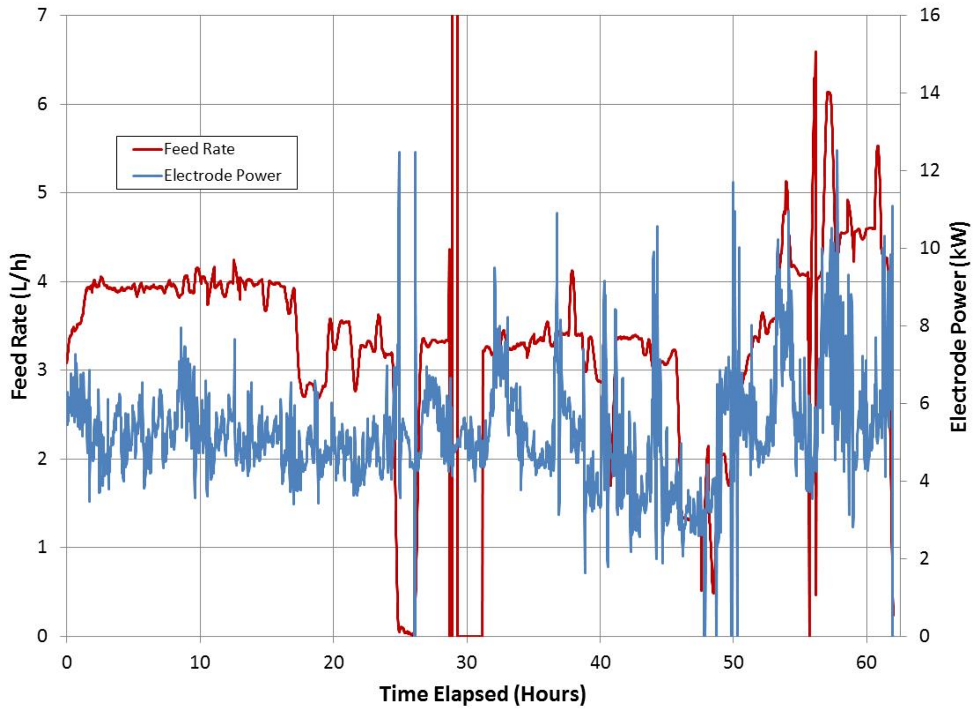


Figure 5.5. Feed Rate (30 min average) and Electrode Power in the RSM-10

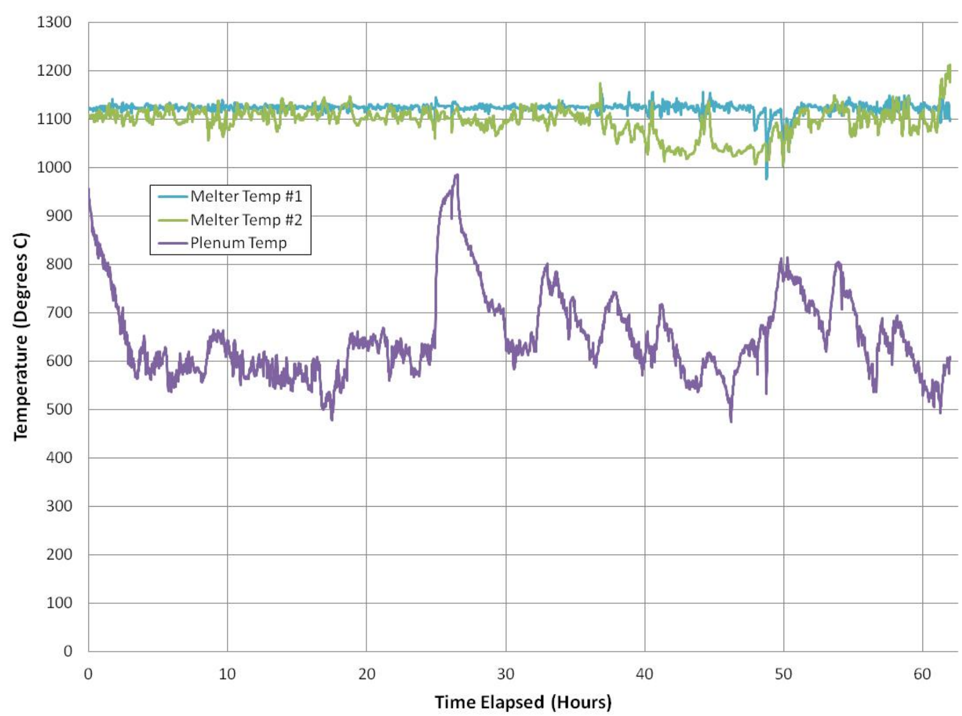


Figure 5.6. Melter Glass and Plenum Temperatures in the RSM-10

RSM-10 Segment 1. Because the melter stopped shortly after the bubbler failure was discovered and the precise time at which it failed is not known, the endpoint of the stable operation period designated for evaluation of the RSM-10 was selected to be a time almost 11 h before the upset in bubbler activity was recognized. This improves the likelihood of avoiding the potential effects of the failing bubbler on the melter assessment. Segment 1 (the only segment) of the RSM-10 test commenced on May 20, 2013, at 0709 hours with an average feed rate of 3.4 L/h, which was thereafter varied to optimize glass production and melter operation. Almost 25 h into the segment, at 0803 hours on May 21, 2013, feeding was interrupted for 99 minutes to make needed adjustments. During the feed stoppage, the temperature set point of the scrub tank chiller was increased, the feed line was flushed, the cold cap was allowed to completely burn off, the thermocouple monitoring the discharge canister was replaced, and the melter viewport was cleaned. The average glass temperature during the segment was 1117°C, the average electrode power was 103 kW/m^2, and the average plenum temperature was 588°C. The average glass temperature in the RSM-10 test was below the target temperature because the temperature set point was deliberately decreased after a thermocouple that was manually inserted into the melt indicated that the actual melt temperature was higher than the melt thermocouples were reporting. It was later recognized that the manually inserted thermocouple was the one providing faulty measurements. Very rapid decreases in the temperature measured by this manually inserted thermocouple were indicated when electrode power was turned off; thus it became clear that electrical noise was causing it to read artificially high temperatures. The cold cap was often observed to almost completely cover the surface of the molten glass pool, at times forming a thick, rigid crust. A few instances of bridging occurred, which were successfully managed by decreasing the feed rate. The disappearance of the cold cap during pours was not observed in the RSM-10. Almost 172 kg of feed was processed during the stable period of the RSM-10 test and the average feed rate was 3.62 L/h with a glass pour rate of 1.95 kg/h (922 $kg/day \cdot m^2$).

As shown in Figure 5.7 and Figure 5.8, with the exception of brief interruptions, the bubbling rates for both RSM-6 and RSM-10 were quite steady, oscillating around the target value without trending higher or lower.

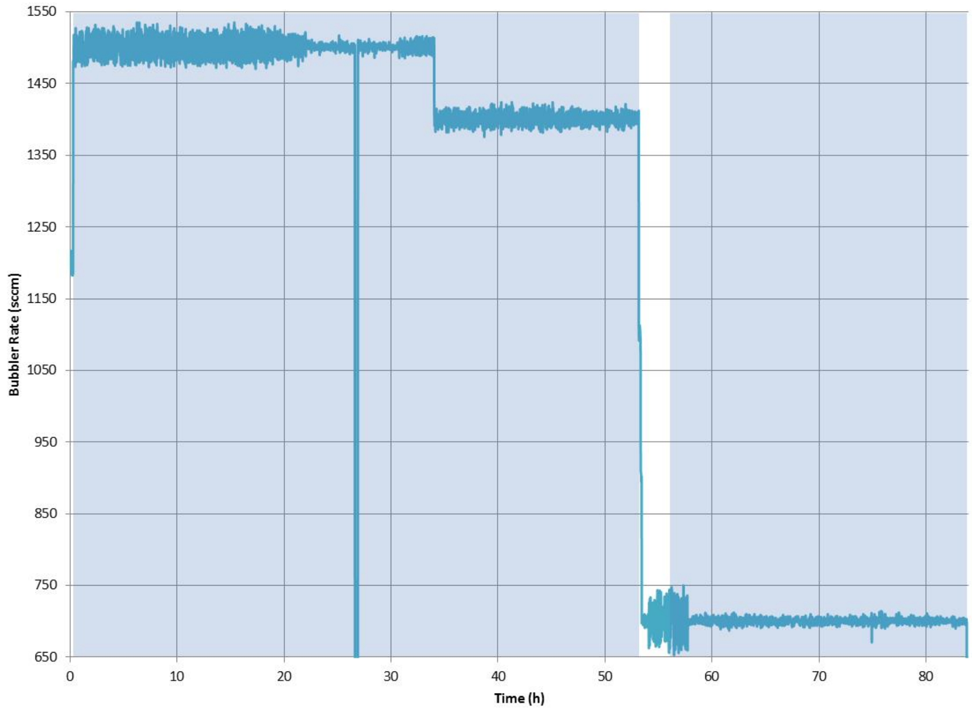


Figure 5.7. Bubbler Flow Rates throughout the RSM-6 Test. Shaded regions designate test segments.

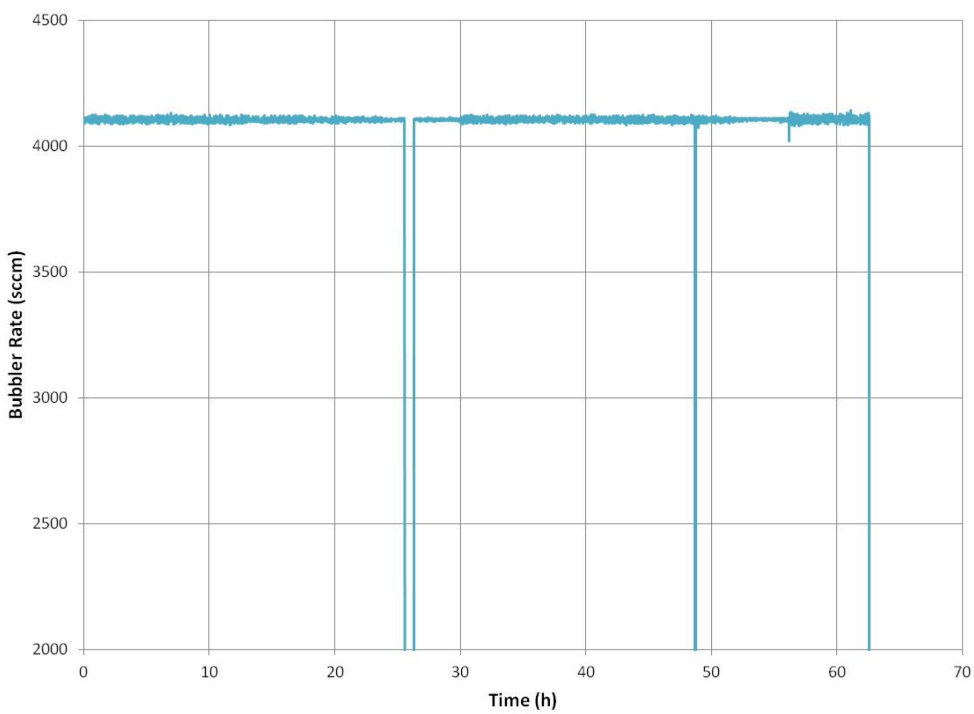


Figure 5.8. Bubbler Flow Rates throughout the RSM-10 Test

5.2 Melter System Inspection

At the conclusion of the tests, the melter and exhaust lines were inspected. The melter electrodes were removed from the melt while the glass was hot. After the glass cooled, the melter lid and the first section of the exhaust line were disassembled. The bubbler tube was then removed by chipping out the glass surrounding it.

As expected, there was no significant corrosion damage observed. The electrodes looked discolored, but it did not appear that a significant amount of metal had been removed. The edges of the electrodes were sharp, as shown in the pictures of the RSM-10 electrodes in Figure 5.9, although there was evidence of heavy oxidation and very small pits on the surface. The bubbler tube from the RSM-6, which was made from Inconel 690 tubing with a 0.05 in. wall thickness, was heavily oxidized, but was not structurally damaged. The bubbler flow in the RSM-10 was split into two bubbler tubes, one of which had essentially the same appearance after testing as the tube from the RSM-6. The only component found to have experienced excessive corrosion was the Inconel 625 bubbler tube, which did not exhibit the corrosion resistance of the Inconel 690 bubbler tube and consequently broke off into the melt as described previously.

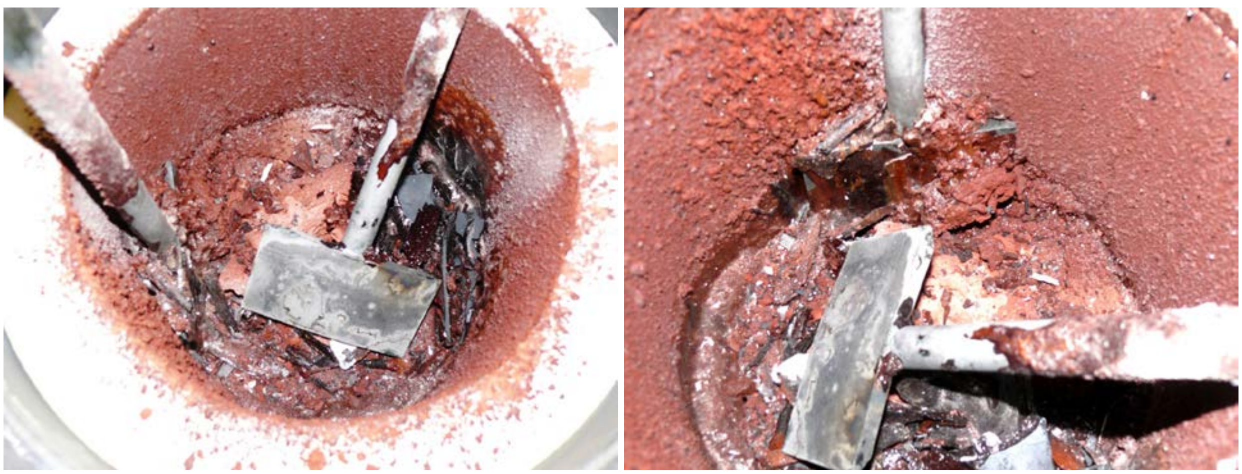


Figure 5.9. RSM-10 Melter Electrodes Pulled From the Glass Melt

Inspection of the melter lid showed that some entrained material adhered to the underside and to the exhaust piping. The deposits appeared to be entrained feed or glass.

The composition of the deposits on the off-gas line from the RSM-6 test and deposits on the bubbler tube inside the RSM-10, assuming oxide forms, is provided in Table 5.2. These measurements were performed using ICP-OES at PNNL and ICP-OES and ion chromatography (IC) analysis at Southwest Research Institute (SwRI). The totals in Table 5.2 do not add up to 100% because anions and water content are not included, and some elements, such as boron, were not analyzed. Boron was not included in the analysis because the method for digestion of the sample to prepare it for ICP-OES analysis used hydrofluoric acid and the samples had to be treated with boric acid before conducting the measurement. The boric acid treatment triggers the complexation of fluoride to protect the ICP torch. Additional discussion of the analysis is provided in Section 7.2. The off-gas line deposits measured at PNNL were enriched by a factor of 2 to 5 in Cr and P and by a factor of over 50 in S, as compared to the target glass composition. The concentrations measured at SwRI were all more dilute than those at PNNL, but the same elements, including Al, Fe, Na, S, and Si, were found to predominate in both measurements. Meanwhile, the off-gas line deposits were depleted by almost 50% in Al, Ca, Si, and Zn. The increased Cr in the off-gas line deposits is likely chromium oxide scale from the underlying metal surface, while the increased P and S were probably condensed from phosphorous and sulfur oxide volatiles in the off-gas. The bubbler tube deposits were enriched in S by a factor of 5, in Cr by a factor of 20, and in Ni by a factor of 50, as compared to the target glass composition, and were depleted in Al, Bi, Ca, Na, P, and Si by around 50%. The highly concentrated Cr and Ni are major components of the oxide scale that would be expected to form on a Ni-based superalloy such as Inconel 625 or Inconel 690. Thus, the bubbler tube deposits were likely cold-cap deposits and entrained particles that did not have time to melt, since the power was stopped abruptly due to a blown fuse, together with oxidation scale from the surface of the underlying bubbler tube.

Table 5.2. Plenum and Exhaust Line Deposit Composition (wt%). (Data is for information only.)

Compound	RSM-6 Off-Gas Line Deposits[a]	RSM-10 Deposits on Bubbler Tube[b]
Al_2O_3	9.32	13.00
Bi_2O_3	1.25	0.53
C_2O_4	ND[c]	NM
CaO	2.66	2.91
Cl	0.001[c]	NM
Cr_2O_3	1.26	10.54
Fe_2O_3	6.81	4.74
F	1.16[c]	NM
Li_2O	2.12	NM
MgO	0.08	0.16
NO_2	ND[c]	NM
NO_3	0.001[c]	NM
Na_2O	9.45	5.62
NiO	0.37	26.87
P_2O_5	4.93	0.58
PbO	0.30	0.26
SO_3	12.46	1.16
SiO_2	9.05	15.10
ZnO	0.34	0.05
ZrO_2	0.31	0.24

(a) Analyzed by ICP-OES and converted to oxide basis unless otherwise noted.
(b) Analyzed by XRF and converted to oxide basis.
(c) Analyzed by IC.
NM – Not measureable with this technique.
The concentrations sum to less than 100% because

some elements, such as boron, were not analyzed.

5.3 Operational Summary and Comparison to VSL Melters

The RSM test segments were successful in providing glass samples and basic operational data for comparison to those reported from studies performed in larger melters at VSL using the same HWI-Al-19 feed composition at the same target melt temperature. Where possible, the other RSM operating parameters were nominally aligned with those of the VSL melter studies on a melt-surface-area–adjusted basis. The most pertinent of the operational data is compared in Table 5.3. To facilitate comparison, data (other than times and dates) from one laboratory that is higher or lower than all corresponding data from the other laboratory is shaded in green or red, respectively.

Table 5.3. Melter Data at PNNL and VSL (Matlack et al. 2008; Joseph et al. 2010) Data (other than times and dates) from one laboratory that is higher or lower than all corresponding data from the other laboratory is shaded in green or red, respectively.

	PNNL Melters			**VSL Melters**		
	RSM-6 Segment 1	RSM-6 Segment 2	RSM-10 Segment 1	DM-100 Test 8	DM-1200 Test 1	DM-1200 Test 2
Segment Start Date	4/15/13	4/17/13	5/20/13	6/25/08	8/6/08	8/11/08
Segment Start Time	9:48	16:26	07:09:51	12:15	09:21	14:45
Total Hours of Melter Operation	84		65	433	157	157
Segment Hours of Slurry Feeding	52.2	26.5	33.3	59.3	48.0	48.3
Avg. Bubbling Rate (Lpm/m^2)	80	38	81	83	105	54
Steady-State Bubbling Rate (Lpm/m^2)	82	38	81	N/A	105	60
Avg. Glass Temperature (C)	1156	1150	1116	1138	1149	1150
Avg. Plenum Temperature (C)	668	641	643	430	653	571
Avg. Feed Rate (kg/h/m^2)	123.2	116.5	101.9	108.3	174.8	121.3
Avg. Glass Pour Rate (kg/day/m^2)	1077	939	922	896	1557	996
Avg. Production Rate* (kg/day/m^2)	1067	1009	883	938	1495	1038
Steady-State Production Rate* (kg/day/m^2)	1115	904	959	950	1500	1050
Avg. Power Use (kWh/kg)*	3.6	4.1	3.0	4.6	2.9	3.4

* Rate calculated from feed rate data.

All test segments conducted at both labs were carried out for between 26 and 60 hours and constituted some portion of a longer melter run, as indicated by the longer times recorded in the “Total Hours of Melter Operation” row in Table 5.3. Besides segment 2 of the RSM-6 melter run, in which the bubbling rate was intentionally decreased to determine the effect of this change on production rates, the average RSM bubbling rates were in line with those of the VSL melter runs, especially with the DM-100 test (Matlack et al. 2008; Joseph et al. 2010). The bubbling rates during periods of steady-state operation did not significantly differ from the corresponding overall average rates. No steady-state bubbling rate was reported for the DM-100 test (Matlack et al. 2008; Joseph et al. 2010). Periods of steady-state operation are defined as 5-hour time spans during which there is minimal variability in feed rate.

The average glass temperature in segment 1 of the RSM-6 tests was only slightly higher than the range of temperatures reported for VSL melter segments, while that for segment 2 fell within that range and was particularly close to the average DM-1200 test temperatures. The average glass temperature measured using the electrode thermocouples in the RSM-10 test was lower than that in the VSL tests. This is because the target electrode temperature was deliberately decreased in this test to compensate for higher temperatures that were measured when a thermocouple was intermittently inserted manually into the melt. It was later discovered that the manually inserted thermocouple produced inaccurate temperature readings due to electrical noise. When electrode power was turned off, this thermocouple alone registered very rapid decreases over periods of only seconds. The average plenum temperatures calculated for the RSM melter segments were on the high end of those reported for the VSL melter tests (Matlack et al. 2008; Joseph et al. 2010). Plenum temperatures are not actively controlled and are largely dependent on melter design as well as cold-cap coverage.

The average feed rates in the RSM melters were within the range of those reported by VSL with the exception of the RSM-10 test, which only fell below this range by less than 7 $kg/h/m^2$. The average glass pour rates of all RSM test segments fell within the range of those reported by VSL, while the average production rates calculated from feed rates obviously followed the same trend as was observed for those average feed rates. However, during periods of steady-state operation in the RSM, the production rates calculated from feed rates fell within the range of rates calculated by VSL for steady-state operation in their melters, except in the case of segment 2 of the RSM-6 test, in which the calculated steady-state production rate was lower. The average power use for glass production in the RSM melters also fell within the range reported by VSL (Matlack et al. 2008; Joseph et al. 2010).

Table 5.4 provides additional operational details of the test segments performed at the two laboratories. This table provides the average, minimum, and maximum values of the reported measurements. In cases for which measurements of temperature were made simultaneously at multiple points in the melter, these statistics are provided for each of the thermocouples. From this table, it is obvious that the RSM-10 exhibited low glass temperatures relative to the VSL melter as discussed above. Meanwhile, the VSL melters had several minimum glass temperatures that were higher than those measured in the RSM melters, meaning that their melters generally did not experience dips in the melt temperature that were as deep as those in the RSM. This is likely dependent on the control system as well as melter size and design and is probably not important to overall rates. Many of the reported VSL plenum temperature statistics were lower than those at PNNL, implying that the plenum temperature ranges were lower in the melters at VSL. The maximum bubbling rates in the RSM melters were lower than those in the VSL melters, while many of the bubbling rate statistics for VSL melters were higher than those for the RSM melters. It should be noted that the differences were small except in cases when the rates were intentionally modified and that higher surface-area-averaged bubbling rates increase the unit feed processing rates. The maximum electrode power in VSL melters was lower than that in RSM melters (Matlack et al. 2008; Joseph et al. 2010).

Figure 5.10 shows a fairly consistent correlation between steady-state glass production and average bubbling rate for PNNL's RSM-6 and VSL's DM-1200. The data points for the RSM-10 and the DM-100 deviated from this trend. The lower production rate for the RSM-10 (1116°C compared to 1150 and 1156°C) is likely attributable to a lower average melt temperature and/or an apparent difference in feed composition as discussed in Section 7.2. However, the cause of the lower production rate for the DM-100 is not clear, because the average melting temperature for the DM-100 was only slightly lower than for the DM-1200 (1138°C compared to 1149 and 1150°C).

Table 5.4. Melter Data at PNNL and VSL (Matlack et al. 2008; Joseph et al. 2010). Data (other than times and dates) from one laboratory that is higher or lower than all corresponding data from the other laboratory is shaded in green or red, respectively.

			RSM-6 Segment 1			RSM-6 Segment 2			RSM-10			DM-100 Test 8			DM-1200 Test 1			DM-1200 Test 2		
		T/C #	AVG	MIN	MAX	AVG	MIN	MAX	AVG	MIN	MAX	AVG	MIN	MAX	AVG	MIN	MAX	AVG	MIN	MAX
Temperature (C)	Glass	1	1144	1105	1169	1147	1070	1166	1125	1104	1152	1120	1075	1176	1149	1113	1177	1156	1131	1174
		2	1167	1098	1204	1153	1078	1180	1108	1057	1174	1133	1096	1183	1146	1110	1173	1153	1127	1170
		3										1146	1122	1200	1147	1113	1171	1153	1126	1171
		4										1153	1132	1210	1117	965	1158	1141	1090	1165
		5													1145	1102	1166	1144	1103	1165
		6													1148	1106	1175	1147	1106	1167
		7													1149	1101	1179	1146	1104	1170
		8													1127	738	1187	1137	1035	1169
	Plenum	1	668	596	781	641	508	777	643	479	970	446	345	626	559	452	763	572	482	664
		2										413	343	596	647	518	782	556	382	778
		3													714	516	986	560	404	726
Bubbling Rate (Lpm/m^2)			80.3	75.4	84.2	38.4	35.9	41.1	81.0	80.6	81.6	83.3	82.4	88.0	104.7	78.4	155.1	59.9	35.6	95.5
Electrode Power (kW/m^2)			160	82	253	149	0	271	103	62	215	174	93	241	190	173	203	148	118	170
Glass Resistance* (ohms)			0.579	0.449	0.886	0.486	0.392	0.805	0.726	0.590	0.998	0.092	0.078	0.101	0.099	0.089	0.110	0.093	0.087	0.102
Melter Pressure (in H_2O)			-1.7	-6.5	1.4	-2.7	-8.8	2.8	-2.0	-5.1	-0.2	-1.0	-2.9	0.4	-3.1	-7.1	0.6	-3.3	-7.9	-0.2

* Does not account for electrical pathway dimensions; therefore comparisons between melters are not meaningful.

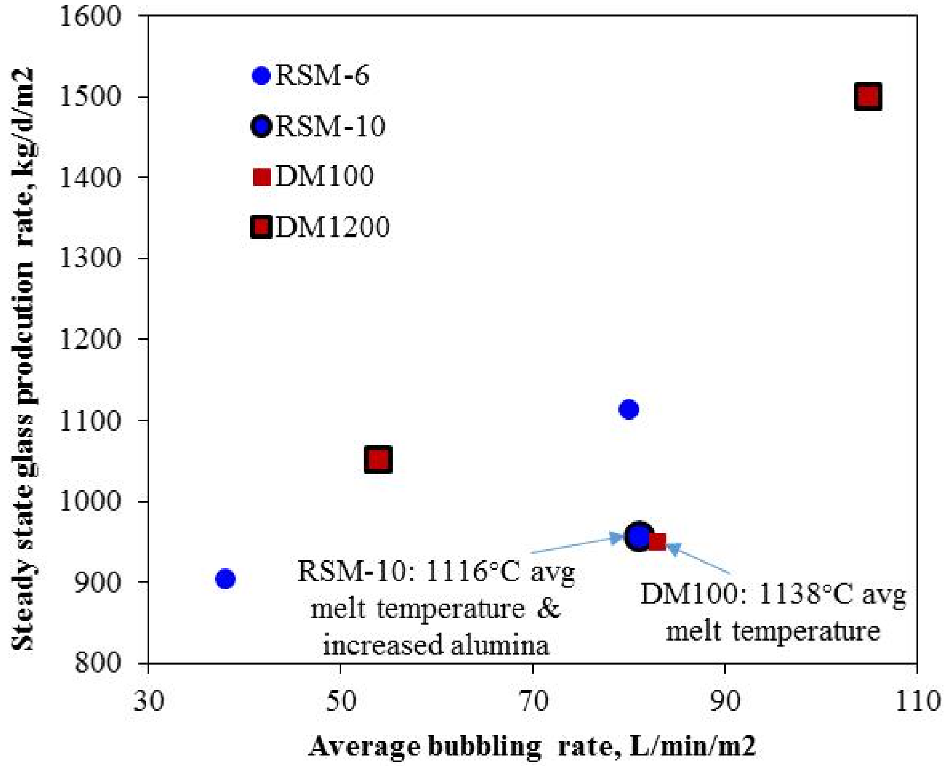


Figure 5.10. Steady-State Glass Production Rates as a Function of Average Bubbling Rate

6.0 Data Collection and Sample Collection/Analysis

Data collection and characterization activities supporting experimental test objectives included the collection of operational, monitoring, and control data as well as process stream compositional information.

6.1 Data Collection and Process Controls

The collection of process operational and control data was performed primarily by the RSM DAC system, which monitors, controls, and electronically logs key system variables at 1 s intervals. Process data not electronically logged by this system and selected parameters of greatest interest were recorded manually on operator datasheets every hour.

Table 6.1 identifies the process information that was electronically logged by the RSM DAC system and/or manually logged on RSM operation datasheets. The data documented important operational conditions associated with the melter, off-gas system, feed, glass, and secondary waste streams. Routine datasheets from the operating procedure were completed on an hourly basis throughout the duration of testing.

Table 6.1. RSM Process Data Logged Electronically (A; 1 s intervals) or Manually (M; 1 h intervals)

Parameter	Units	Electronic Log	Manual Log
Melt temperature (T1, T2)	°C	X	X
Plenum temperature	°C	X	X
Feed pump setting	%	X	X
Feed pump control mode	Int or Ext	---	X
Recirculation feed pump setting	N/A	---	X
Cold-cap coverage	%	---	X
Cold-cap flexibility	Y or N	---	X
Glass pouring	Y or N	---	X
Electrode potential	V	X	X
Electrode current	A	X	X
Electrode power	kW & %	X	X
Electrode power control mode	A or M	X	X
Melt resistance	Ω	---	X
Melt (electrode) set-point temperature	°C	X	X
Kiln power	kW & %	X	X
Kiln set-point temperature	°C	X	X
Kiln actual (middle) temperature	°C	X	X
Kiln top/bottom temperatures	°C/°C	X	X
Kiln control mode	A or M	X	X
Discharge can power	kW	X	X
Discharge can set-point temperature	°C	X	X

Table 6.1. (contd)

Parameter	Units	Electronic Log	Manual Log
Discharge can actual temperature	°C	X	X
Discharge can power output	%	X	X
Pour spout heater set-point temperature	°C	X	X
Pour spout heater temperature	°C	X	X
Pour spout heater power output	kW & %	X	X
Feed nozzle temperature	°C	X	X
Feed nozzle cooling flow	gph	---	X
Feed loop cooling flow	gph	---	X
Film cooler air flow rate (indicated)	scfm	---	X
Film cooler back pressure	psi	---	X
Sight glass air sweep flow	Lpm	---	X
Melter vacuum gauge	in. H_2O	---	X
EVS heat exchanger cooling flow	gpm	---	X
EVS scrub tank level	in. H_2O	X	X
EVS nozzle pressure	psi	---	X
EVS flow rate	gpm	---	X
Blower inlet vacuum	in. H_2O	---	X
Off-gas temperature	°C	X	X
Off-gas control valve	in.	---	X
Post-EVS off-gas temperature	°C	X	X
Scrub liquid EVS inlet temperature	°C	X	X
Heat exchanger temperature	°C	X	X
Post HEME Temperature	°C	X	X
HEME pressure	in. H_2O	X	X
System pressure	in. H_2O	X	X
Chiller temperature (#1, #2)	°C	---	X
Plenum vacuum	in. H_2O	X	X
Plenum vacuum control valve position	%	X	X
Bubbling rate	L/min	X	X
Alarm condition	On or Off	X	X
Feed line pressure	psi	X	X
Feed tank agitator setting	%	---	X
Feed tank weight	kg	X	X
Glass scale	kg	---	X

Other items noted in the RSM logbook included visual observations of the operating behavior of the feed system, melter, and off-gas system; processing anomalies involving the cold cap, glass conditions, off-gas behavior, or corrosion; and all feed rate adjustments, operational problems, and optimization activities.

6.2 Process Sample Collection and Analysis

Routine sampling of the feed, glass, and off-gas streams was conducted based on instructions in the test procedure. The melter feed recirculation loop allowed for direct sampling of the feed stream just before it entered the melter. Glass samples were collected from the melter pour spout stream with a rectangular graphite boat. Because the newly formed glass bar could shatter and create a sharp projectile hazard, glass samples were shielded while cooling. These samples were used as the rapidly cooled "quenched" samples for the PCT and TCLP tests. The EVS condensate samples were directly extracted from a valve on the condensate recirculation line. The HEME runoff was manually recycled back to the EVS recirculation tank. Collection of accumulated undissolved solids in the quench scrubber's condensate tank was conducted at the conclusion of testing to support mass balance evaluations.

Process samples collected for analysis included the feed slurry, glass product, and EVS scrubbing liquid. In general, process samples were collected at least once per day and for every identified "stable" operating condition (except for off-gas line deposits and the EVS undissolved solids (UDS), which were collected only at the conclusion of testing). Sample analyses were conducted to characterize the quantities, compositions, and properties of these process streams following the protocol called out in the Hanford Analytical Services Quality Requirements Document (HASQARD).

Glass sample analyses included inductively coupled plasma–optical emission spectroscopy (ICP-OES) to determine the elemental composition, XRD to determine crystallinity, and PCT and TCLP tests to determine durability.

Although several feed, glass, off-gas waste stream, and pipe-accretion samples were collected as indicated in the table, only those samples considered to be most representative of selected test conditions were analyzed. Furthermore, not all analytical samples of a particular type were subjected to identical analytical schedules. For example, some glass samples analyzed for elemental composition were not analyzed for durability or other properties. However at least one feed, glass, and EVS sample was analyzed for each test condition.

Sample Identification: Process stream samples were collected as detailed in the test instruction (Sevigny 2013). For example, feed slurry samples were taken at the beginning of the test and every ~24 hours thereafter, off-gas pipe accretions were collected at the end of testing, EVS solution samples were acquired in intervals of approximately 8 hours, and glass samples were procured according to the schedule in Table 6.2. All samples were identified according to the following unique sequential labeling scheme (starting with RSM-EWG1-001) and logged with the descriptive information listed below:

- date and military time
- sample description (e.g., feed, condensate, glass sample)
- initials of operations staff obtaining sample.

This information was recorded on sample log sheets and all sample containers were similarly labeled.

Table 6.2. Glass Sampling Schedule

Sample	Sampling Frequency	Size
Sample 1	Immediately after overflow glass stream begins	Standard sample boat
Samples 2–5	Sample every hour for 4 hours	Standard sample boat
Samples 6–9	After 4 hours, sample every 2 hours for 8 hours	Standard sample boat
Samples 10–13	After 12 hours, sample every 4 hours for 16 hours	Standard sample boat
Remaining Samples	After 28 hours, sample every 8 hours	Standard sample boat
At end of test	Take one large sample and cool rapidly	>300 grams (~120 cc) in wide stainless steel container

Process and off-gas samples were analyzed, as applicable, for elemental composition, durability, and density. Some analyses were contemporaneous with test operations. Other analyses required preparations of several hours or days, depending on the analysis performed, the sample preparation required prior to analysis, and the location of the analytical equipment. Table 6.3 briefly describes the different analyses included in this test program. Table 6.4 lists the glass samples analyzed in this study together with the analyses that were performed. Each of the analyses enumerated in this table was performed on an as-cooled (RSM-EWG1-034 only, which was cooled in a canister) or quenched glass, while those followed by the letter “c” were additionally performed on a canister centerline cooling (CCC)-treated specimen. Table 6.5 and Table 6.6 list the analyses that were performed on EVS solution and feed samples, respectively.

Table 6.3. Sample Analysis Methods for Process and Off-Gas Samples

Analysis	Sample Matrix	Analysis Method	Analysis Description
Cations	Solid or liquid	ICP-OES for metals, XRF	Analysis of total amount of element, regardless of speciation
Anions	Liquid	IC	Br^-, Cl^-, F^-, NO_3^-, NO_2^-, PO_4^{3-}, SO_4^{2-}
	Solid	XRF	F^-
Durability	Glass	PCT and TCLP	ASTM and EPA Procedures[(a)] (ASTM 2008, EPA 1992)
Density	Solid	Archimedes	Liquid displacement
	Liquid	Mass/Volume	Mass/Volume

(a) ASTM = American Society for Testing and Materials; EPA = U.S. Environmental Protection Agency.

Table 6.4. List of Glass Samples Analyzed

Seg. #	ID	Date	Time	Analyses	
RSM-6 #1	RSM-EWG1-013	4/16/2013	0122	ICP-OES	
RSM-6 #2	RSM-EWG1-028	4/17/2013	1916	ICP-OES	
RSM-6 #2	RSM-EWG1-034[(a)]	4/18/2013	1000	ICP-OES	TCLP c PCT c Visc Elec Cond SEM c XRD c Liq T IC SEM
RSM-10 #1	RSM-EWG2-027	5/22/2013	0738	ICP-OES	

c = canister centerline cooling (CCC) treated, Visc = viscosity, IC = ion chromatography
Elec Cond: electrical conductivity, Liq T: liquidus temperature, SEM = scanning electron microscopy
(a) large amount of glass poured in a discharge canister

Table 6.5. List of EVS Solution Samples Analyzed

Seg. #	ID	Date	Time	Analyses
Pre-RSM-6	RSM-EWG1-003	4/15/2013	0650	IC
RSM-6 #1	RSM-EWG1-020	4/16/2013	1630	IC
RSM-6 #1	RSM-EWG1-026	4/17/2013	1030	IC
RSM-6 #2	RSM-EWG1-033	4/18/2013	0950	IC
Post-RSM-6	RSM-EWG1-040	4/19/2013	1030	IC
Pre-RSM-10	RSM-EWG2-002	5/20/2013	0633	IC
RSM-10 #1	RSM-EWG2-016	5/21/2013	0715	IC
Post-RSM-10	RSM-EWG2-022	5/22/2013	0425	IC
Post-RSM-10	RSM-EWG2-030	5/22/2013	2330	IC

Table 6.6. List of Feed Samples Analyzed

Seg. #	ID	Date	Time	Analyses	
Pre-RSM-6	RSM-EWG1-002	4/15/2013	0650	ICP-OES	Density
RSM-6 #1	RSM-EWG1-017	4/16/2013	0751		Density
RSM-6 #1	RSM-EWG1-025	4/17/2013	0857		Density
RSM-6 #2	RSM-EWG1-032	4/18/2013	0926	ICP-OES	Density
Pre-RSM-10	RSM-EWG2-001	5/20/2013	0633		Density
Post-RSM-10	RSM-EWG2-023	5/22/2013	0720		Density

7.0 Glass Characterization

In the RSM-6 and RSM-10 tests, over 60 and 100 kg of glass were produced, respectively. With the exception of the quenched glass samples collected as described above, the glass was discharged from the RSM pour spout into stainless steel beakers and allowed to cool.

7.1 Heat Treatment of Glass Following Canister Centerline Cooling Profile

Selected glass samples were heat treated following the HLW CCC profile.[1] Glass samples were placed in a Pt-10%Rh crucible, covered with a lid, and placed in a high-temperature furnace at 1050°C for 1 h. They were then cooled using a programmable furnace controller following the CCC schedule shown in Table 7.1 and Figure 7.1. Glass sample RSM-EWG1-034, heat treated following the CCC profile, was characterized by scanning electron microscopy (SEM), XRD, TCLP, and PCT.

Table 7.1. Temperature Schedule during CCC Treatment

Segment	Time (min)	Start Temp. (°C)	Rate (°C/min)
1	0–45	1050	−1.556
2	45–107	980	−0.806
3	107–200	930	−0.591
4	200–329	875	−0.388
5	329–527	825	−0.253
6	527–707	775	−0.278
7	707–1776	725	−0.304

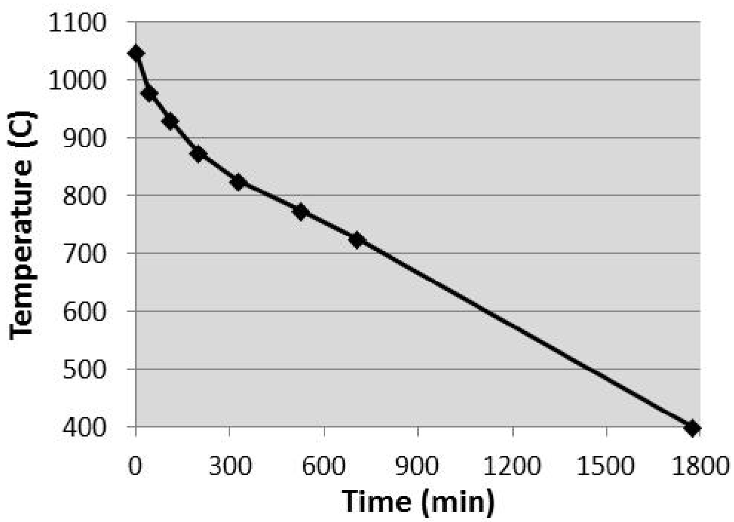


Figure 7.1. Graph of CCC Heat Treatments of HLW Glass

[1] Memorandum, Canister Centerline Cooling Data, Revision 1, CCN: 074851, RPP-WTP, October 29, 2003.

7.2 Analyses of Glass for Chemical Composition

To confirm that the fabricated glass corresponded to the target composition, representative quenched glass from sample RSM-EWG1-034 was chemically analyzed at Savannah River National Laboratory (SRNL) (Peeler and Edwards 2013; Fox and Edwards 2014). Duplicate samples were prepared by two techniques, sodium peroxide fusion and acid dilution. The resulting dissolutions were analyzed using ICP-OES twice for each of the elements of interest, which included Al, B, Bi, Ca, Cd, Cr, Fe, Pb, Li, Mg, Mn, Ni, P, K, Ru, Na, S, Si, Sr, and Zr concentrations. Two components of the study glasses, F and Ag, were not measured, since each of these species would have required the use of an additional preparation method and their measured values were likely to be near or below analytical detection limits. Several samples of the low-activity waste reference material (LRM) (Ebert and Wolfe 1999) were intermittently measured in duplicate to assess the performance of the ICP-OES instrument over the course of these analyses.

Specimens from the same glass sample were also analyzed using ICP-OES at SwRI. Here, the samples were readied for analysis using three techniques. The first technique was a closed vessel digestion using concentrated nitric, hydrochloric and hydrofluoric acids. Boron and silicon were reported from this digestion. The second technique was a lithium metaborate/tetraborate fusion. Aluminum, calcium, chromium, iron, and sodium were reported from this fusion. The third technique was performed using concentrated nitric, perchloric, hydrofluoric, and hydrochloric acids in an open vessel. The remaining metals were reported from this digestion. Blanks were run to ensure that no analytes were falsely detected above SwRI's reporting limits. Two solid laboratory control samples (NIST SRM[1] 278 Obsidian Rock and NIST SRM 688 Basalt Rock) were used during all sample preparation techniques. Aqueous laboratory control samples were also digested for calibration. Additionally, SwRI performed IC analysis on the sample. Approximately 0.200 g of the sample was fused using sodium carbonate. The fusion was diluted to 50 mL using deionized water and analyzed by IC using SW-846 Method 9056 (EPA 1994) and in accordance with SwRI procedure TAP 01-0406-042 Rev. 6.[2] Due to the high sodium carbonate concentration, they were diluted 10× prior to analysis.

The ICP-OES results from the two labs are summarized in Table 7.2, which provides a summary of the measured composition of the glass processed at PNNL together with the targeted composition and the relative differences between the measured and targeted values for major components (those with target concentrations >1 wt%). The results from SRNL were clearly closer to the target composition than those from SwRI, while SwRI provided measurements for several of the minor constituents not measured by SRNL. All of the major glass components measured at SRNL were within ±5% of the target concentration, indicating that the glass was very close to the target composition. In general, larger percentage differences were observed for the results from SwRI, especially for P_2O_5 and SiO_2. The ICP-OES measurements performed at PNNL on the RSM-EWG2-004 and RSM-EWG2-008 samples were not reported due to issues with obtaining reliable concentrations for Si.

VSL reports that in their tests of HWI-AL-19 glass melted in the DM100 and DM1200 melters, the oxides with a target concentration greater than 1 wt% showed around, or below, 10% deviation from the target values with the exception of phosphorous oxide which had an absolute deviation of no more than

[1] NIST SRM = National Institute of Standards and Technology Standard Reference Material
[2] SWRI. 2012. Test/Analytical Procedure (TAP) 01-0406-042, *Inorganic Anions and Disinfection Byproducts using Ion Chromatography*. July 2012, Revision 6. Southwest Research Institute, San Antonio, TX.

0.15 wt%. The ICP-OES measurements performed at SRNL indicate that sample RSM-EWG-1-034 from the glass melted at PNNL also meets this criteria.

Table 7.2. Composition of HWI-Al-19 Glass (RSM-EWG-1-034) Melted at PNNL and Analyzed by ICP-OES at SRNL (Fox 2014) and SwRI

		SRNL		SwRI	
Oxide	**Targeted (wt%)**	**Measured (wt%)**	**% Difference of Measured versus Targeted**	**Measured (wt%)**	**% Difference of Measured versus Targeted**
Al_2O_3	23.97	23.9022	-0.3%	21.999	−8.2%
BaO	0.05	NM	NM	0.0412	NA
B_2O_3	19.19	18.4017	-4.1%	18.003	−6.2%
Bi_2O_3	1.14	1.09	-4.4%	1.039	−8.9%
CaO	5.58	5.6773	1.7%	5.176	−7.2%
CdO	0.02	0.0166	NA	0.0228	NA
Cr_2O_3	0.52	0.3172	NA	0.34	NA
F	0.67	NM	NM	0.212[(a)]	NA
Fe_2O_3	5.9	5.9904	1.5%	5.112	−13.4%
K_2O	0.14	0.0952	NA	0.122	NA
La_2O_3	0	NM	NM	0.0003	NA
Li_2O	3.57	3.6061	1.0%	3.583	0.4%
MgO	0.12	0.1658	NA	0.0477	NA
MnO	0	0.1291	NA	0.021	NA
Na_2O	9.58	9.6449	0.7%	9.024	−5.8%
NiO	0.4	0.1482	NA	0.353	NA
P_2O_5	1.05	1.0008	-4.7%	0.759	−27.7%
PbO	0.41	0.363	NA	0.343	NA
RuO_2	0	0.0132	NA	NM	NM
SiO_2	27	28.0783	4.0%	32.247	19.4%
SO_3	0.2	0.1298	NA	0.082	NA
SnO_2	0	NM	NM	0.0007	NA
SrO	0	0.0118	NA	0.0019	NA
TiO_2	0.01	NM	NM	NM	NM
WO_3	0	NM	NM	0.0093	NA
ZnO	0.08	NM	NM	0.0767	NA
ZrO_2	0.39	0.3725	NA	0.382	NA
Sum	100	99.1541	NA	98.7856	NA

(a)Analyzed by IC at SwRI.
NM = not measured; NA = not applicable.
La_2O_3, MnO, RuO_2, SnO_2, WO_3, and SrO are listed as impurities.

Table 7.3 lists additional ICP-OES measurements performed at SwRI on samples taken from segment 1 (RSM-EWG1-013) and segment 2 (RSM-EWG1-028) of the RSM-6 test and from the RSM-10 test (RSM-EWG2-027). The compositions of the three samples were very consistent with one another and the deviations from target concentrations, which are negative for most constituents, may be in part explained by the 4–5 wt% loss on ignition (LOI). Overall, the measured compositions of glass product from RSM-6 and RSM-10 runs conformed reasonably well to the target composition. Here again, the glass melted in the RSM melters met the same compositional criteria as the glass melted in the DM melters except that the phosphorous concentrations deviated by up to 0.19 wt% as opposed to only 0.15 wt% at VSL.

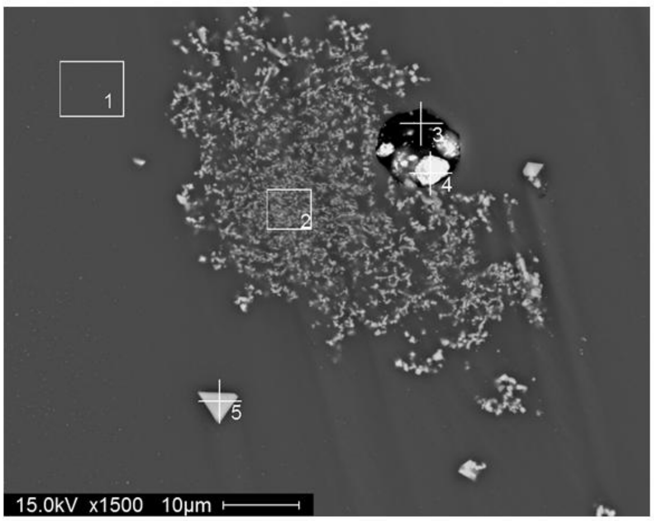
Table 7.3. Composition of HWI-Al-19 Glass Melted at PNNL and Analyzed by ICP-OES at SwRI

		RSM-EWG1-013		RSM-EWG1-028		RSM-EWG2-027	
Oxide	**Targeted (wt%)**	**Measured (wt%)**	**% Difference of Measured versus Targeted**	**Measured (wt%)**	**% Difference of Measured versus Targeted**	**Measured (wt%)**	**% Difference of Measured versus Targeted**
Al_2O_3	23.97	24.281	1.3%	24.564	2.5%	23.997	0.1%
BaO	0.05	0.065	NA	0.051	NA	0.050	NA
B_2O_3	19.19	17.808	−7.2%	18.065	−5.9%	17.969	−6.4%
Bi_2O_3	1.14	1.100	−3.5%	1.118	−1.9%	1.110	−2.6%
CaO	5.58	5.219	−6.5%	5.247	−6.0%	5.219	−6.5%
CdO	0.02	0.023	NA	0.023	NA	0.052	NA
Cr_2O_3	0.52	0.350	NA	0.362	NA	0.367	NA
F	0.67	NM	NM	NM	NM	NM	NM
Fe_2O_3	5.9	5.762	−2.3%	5.819	−1.4%	5.883	−1.1%
K_2O	0.14	0.107	NA	0.103	NA	0.096	NA
La_2O_3	0	ND	ND	ND	ND	ND	ND
Li_2O	3.57	3.272	−8.3%	3.294	−7.7%	3.272	−8.3%
MgO	0.12	0.210	NA	0.196	NA	0.199	NA
MnO	0	0.026	NA	0.023	NA	0.022	NA
Na_2O	9.58	9.241	−3.5%	9.315	−2.8%	9.301	−2.9%
NiO	0.4	0.372	NA	0.372	NA	0.368	NA
P_2O_5	1.05	0.887	−15.5%	0.871	−17.0%	0.864	−17.7%
PbO	0.41	0.359	NA	0.369	NA	0.362	NA
RuO_2	0	NM	NM	NM	NM	NM	NM
SiO_2	27	27.596	2.2%	28.024	3.8%	27.382	1.4%
SO_3	0.2	0.106	NA	0.137	NA	0.128	NA
SnO_2	0	0.0007	NA	0.0007	NA	0.0008	NA
SrO	0	0.0019	NA	0.0018	NA	0.0017	NA
TiO_2	0.01	0.041	NA	0.042	NA	0.040	NA
WO_3	0	0.0007	NA	0.0005	NA	0.0004	NA
ZnO	0.08	0.078	NA	0.081	NA	0.078	NA
ZrO_2	0.39	0.3725	NA	0.382	NA	0.377	NA
LOI		4.86		4.09		4.33	
Sum	100	102.0		102.5		101.3	

NM=Not measured; NA=not applicable; ND=not detected
La_2O_3, MnO, RuO_2, SnO_2, WO_3, and SrO are listed as impurities.

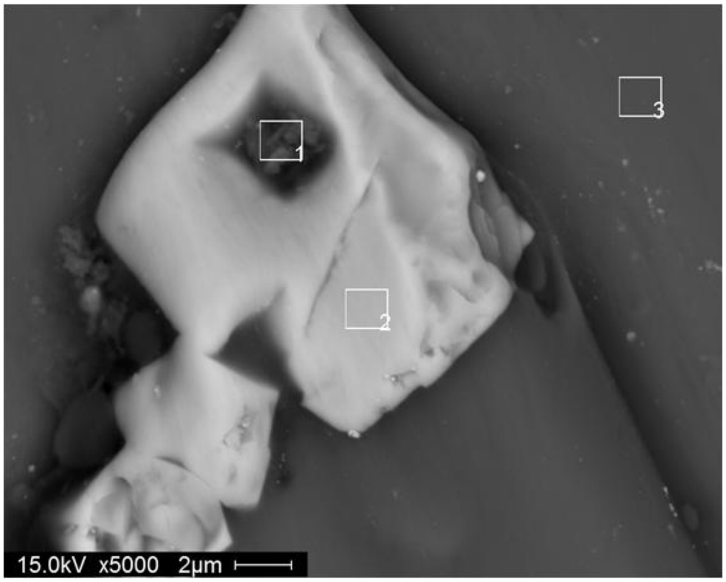
7.3 Crystallinity by Scanning Electron Microscopy and X-ray Diffraction

Figure 7.2 is an SEM micrograph of a cross-section of the middle of canister 3 from the RSM-6 melt (glass sample ID: RSM-EWG1-034) together with the results of the energy dispersive spectroscopy (EDS) analysis of designated regions. The micrograph revealed an area where small crystals had agglomerated (EDS region 2) in the surrounding glass matrix (EDS region 1). As compared to the glass matrix, the small crystals in region 2 contained increased Fe, Ni, and Cr, which are the major constituents of spinel phases typically detected in waste glass (Barnes and Larson 1981; Matlack et al. 2009; Rose et al. 2011). In addition, region 2 was also enriched by Al and Mg, which are found in spinel crystals from certain high-Al glasses (Smith et al. 2014). The larger crystal associated with EDS spot 5 was also found to contain higher levels of the five elements indicative of a spinel. The EDS analysis of spots 3 and 4 indicated that the dark region was a bubble containing polishing debris, which was lighter in color. The micrograph in Figure 7.3 is centered around another large crystal (region 2) that EDS indicated was also spinel. The dark area (region 1) atop the crystal is believed to include polishing debris, based on the EDS results. Careful microscopic examination of several different areas within the canister and within the sample described above found no undissolved feed material, only crystals from spinel formation in the melt.



	Wt%				
Element	Area3-1	Area3-2	Area3-3	Area3-4	Area3-5
OK	49.39	51.67	34.55	21.81	57.31
FK	0.00	0.00	0.00	28.52	0.00
NaK	10.50	7.64	3.29	0.87	2.04
MgK	0.28	0.41	0.57	0.00	1.26
AlK	17.00	20.70	10.75	1.23	10.74
SiK	17.88	11.23	12.08	2.49	1.92
WM	0.00	0.00	0.00	6.66	0.00
PK	0.71	0.66	0.32	0.63	0.13
SK	0.15	0.00	3.35	0.00	0.00
BiM	1.03	0.72	1.01	0.00	0.15
ClK	0.00	0.00	5.57	0.00	0.00
CaK	1.77	1.46	9.31	10.35	0.23
LaL	0.00	0.00	2.82	3.69	0.00
CeL	0.00	0.00	14.95	23.17	0.00
PrL	0.00	0.00	0.00	0.58	0.00
CrK	0.10	0.27	0.22	0.00	9.08
FeK	1.19	3.46	1.21	0.00	11.57
NiK	0.00	1.78	0.00	0.00	5.57
Total	100.00	100.00	100.00	100.00	100.00

Figure 7.2. Micrograph of a Polished Cross-Section from the RSM-6 Melt with EDS Analysis



	Wt%		
Element	Area3-1	Area3-2	Area3-3
OK	44.63	55.42	51.06
NaK	11.52	1.63	9.32
MgK	0.29	1.56	0.27
AlK	11.62	10.14	16.85
SiK	11.42	0.28	17.80
PK	0.44	0.07	0.78
SK	0.84	0.00	0.00
BiM	0.51	0.13	1.01
ClK	0.61	0.00	0.00
KK	0.60	0.00	0.00
CaK	5.47	0.16	1.70
CrK	5.15	12.38	0.06
FeK	4.94	12.14	1.15
NiK	1.52	5.30	0.00
ZnK	0.44	0.79	0.00
Total	100.00	100.00	100.00

Figure 7.3. Micrograph and EDS of a Large Crystal in the RSM-6 Melt

The XRD data were collected using a Bruker D8 X-ray diffractometer by scanning over the range of 5–70°2θ with a step size of 0.009° and a 0.3 s dwell at each step. Data were analyzed with EVA v.14 and TOPAS v.4.2 software (Bruker AXS, Karlsruhe, Germany) for phase identification. For the purposes of semiquantitative analysis, 5 wt% CaF_2 was added to each sample as an internal standard. The crystalline content was determined by Rietveld refinement of the XRD patterns using TOPAS software.

Figure 7.4 is an XRD pattern of a sample of glass taken from the same portion of canister 3 as the microscope sample. The XRD pattern confirmed the microscopy results, identifying spinel phases. Based on the SEM-EDS results discussed above, the spinel crystals are likely a solid solution of magnetite (Fe_3O_4), chromite ($FeCr_2O_4$), nichromite ($NiCr_2O_4$), trevorite ($NiFe_2O_4$), and spinel ($MgAl_2O_4$). VSL reported that visual observations of dip samples of the glass processed in their melters to detect the presence of any secondary phases on the melt pool surface revealed no evidence of secondary phases, however no SEM or XRD was reported. It is evident from the discussion of liquidus temperature determination hereafter that VSL was aware of the tendency of this glass composition to form secondary crystalline phases such as those detected in their crucible melts.

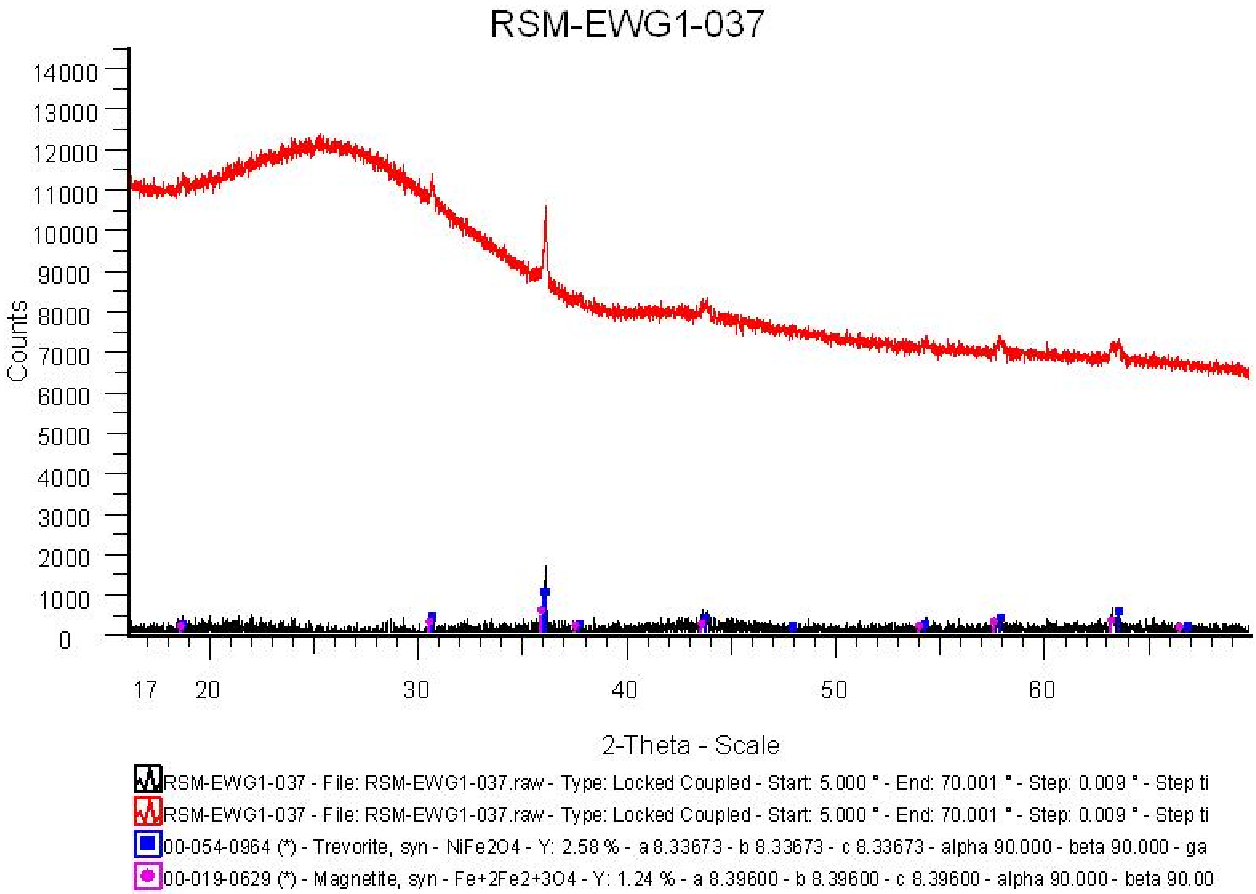


Figure 7.4. XRD Pattern of the RSM-6 Melt Evaluated Microscopically in Figure 7.2 and Figure 7.3

Figure 7.5 is an XRD pattern of a specimen of the RSM-6 glass (RSM-EWG1-034) after it was subjected to CCC heat treatment. A spinel phase was detected at a concentration of 6.7 wt%, corresponding to roughly 3.7 vol%, which is higher than the 1.9 vol% reported by VSL after CCC heat treatment of HWI-Al-19 glass that was melted in crucibles.

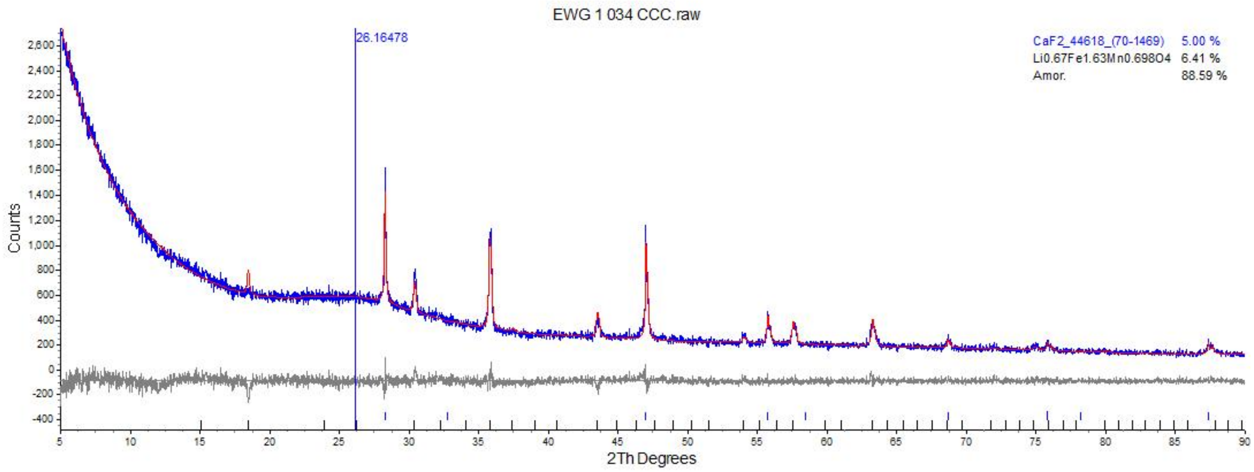


Figure 7.5. XRD Pattern of the RSM-6 Glass after CCC Heat Treatment

XRD analysis of glass from sample RSM-EWG1-034 was also utilized for the purpose of determining the liquidus temperature on the basis of crystalline content as a function of temperature. XRD patterns were collected after the glass had been isothermally heat treated at 950, 1000, 1050, and 1100°C for 24 h. The resulting data is shown in blue in Figure 7.6 together with data (in red) from the same glass composition melted in a crucible at VSL. From extrapolation of the linear fit line of temperature versus crystal vol% for the PNNL glass, the estimated liquidus temperature is 1218°C, as compared to 1031°C for the VSL glass. The temperature at 1 vol% spinel is 1003°C for the PNNL glass and 969°C for the VSL glass. The volume fraction of spinel at 950°C is 1.3 vol% for the glasses processed at both labs.

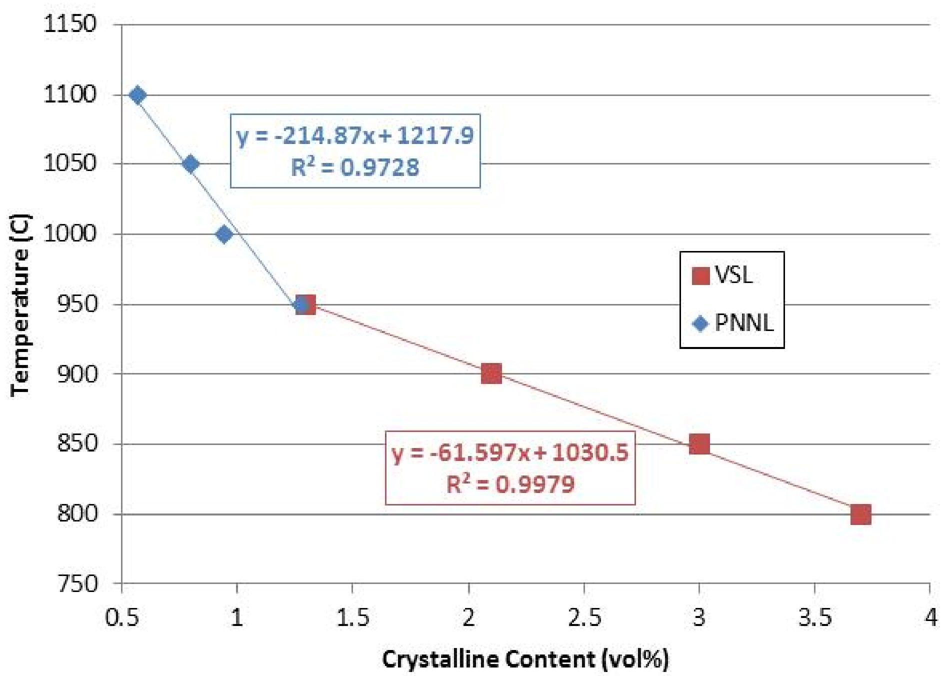


Figure 7.6. Temperature versus Spinel Content for HWI-Al-19 Glass Melted at PNNL and VSL

7.4 Toxicity Characteristic Leaching Procedure

As-cooled and CCC-treated samples of RSM-6 glass, RSM-EWG1-034, were sent to SwRI for TCLP analysis, following U.S. Environmental Protection Agency (EPA) Test Method 1311 (EPA 1992). Results from the glass melted in the DM-100 at VSL are included for comparison. It was not clear from the VSL report whether the samples were as-cooled or CCC treated, but the lack of designation suggests that they were as-cooled samples. The regulatory TCLP limit for WTP is that the TCLP Cd concentration for both quenched (or as-cooled) and CCC samples should be less than 0.48 mg/L (see Kim et al. 2011a). Table 7.4 shows that the RSM-6 sample meets this requirement. For the elements that were reported, TCLP concentrations for the VSL glass were lower than those for the PNNL glass. Table 7.4 also gives the results of other toxic elements for information only.

Table 7.4. TCLP Concentrations for HWI-Al-19 Glass from RSM-6 (RSM-EWG1-034)

CAS No.	Analyte	PNNL As-cooled Concentration (μg/L)	PNNL CCC Concentration (μg/L)	VSL Concentration (μg/L)
7440-38-2	As	20.0	20.0	NR
7440-39-3	Ba	236	232	200
7440-43-9	Cd	68.7	98.1	30
7440-47-3	Cr	188	22.6	80
7439-92-1	Pb	1660	1620	390
7439-97-6	Hg	0.20	0.20	NR
7782-49-2	Se	10.0	10.0	NR
7440-22-4	Ag	10.0	10.0	NR

7.5 Product Consistency Test

The PCT was conducted at both SRNL (Fox and Edwards 2014) and PNNL to determine the durability of the glass in deionized water according to ASTM C1285-02, Method B (ASTM 2008). Unsensitized Type 304L stainless steel vessels were cleaned and loaded with two samples and a glass standard, each in triplicate. The PCT was performed in the sealed vessels at 90°C for seven days. Blanks were also included in each test. Sample material was designated RSM-EWG1-034 or RSM-EWG1-034-CCC. The "CCC" denotes further sample treatment by CCC treatment prior to the PCT. The test reference material, DWPF Environmental Assessment (EA) Standard Reference Material, was supplied by SRNL.

The PCT normalized releases are summarized in Table 7.5. The results obtained from measurements performed at PNNL and SRNL agreed reasonably well with one another. The quenched and CCC-treated RSM specimens subjected to PCT analysis at both labs passed the Hanford HLW glass requirement (DOE 1996) for normalized B, Na, and Li releases, as shown in Table 7.5. Additionally, the PCT results reported for the HWI-Al-19 glass melted in VSL's DM-100 melter (sample ID# BLZ-G-37A in Matlack et al. 2008) are included in the table for comparison. The VSL melter glasses had normalized releases 20–35% higher than the RSM glasses.

Table 7.5. Summary of Average PCT Results

Normalized Release (g/m^2)						
Element	**PNNL Melter**				**VSL Melter**	**EA Glass**
	PNNL Q[(a)]	**SRNL Q**	**PNNL CCC**[(b)]	**SRNL CCC**	**VSL**	**Requirements**
B	0.204	0.206	0.229	0.227	0.31	<8.35
Li	0.264	0.242	0.266	0.232	0.32	<4.79
Na	0.184	0.181	0.198	0.206	0.25	<6.68
Si	0.081	0.071	0.090	0.087	0.11	
pH	9.30	9.46	9.28	9.28	9.37	

(a) Q = quenched
(b) CCC = also CCC treated

7.6 Electrical Conductivity versus Temperature

The electrical conductivity of HWI-Al-19 glass was measured at temperatures ranging from 900°C to 1300°C using a specimen from sample RSM-EWG1-034. Impedance spectroscopy was performed with a Solartron 1470E potentiostat/galvanostat coupled with a Solartron 1400 frequency analyzer (Solartron Analytical, Oak Ridge, TN) on the glass while cooling through the temperature range from 1300°C to 900°C. (Crum 2012a) Testing was done in a Pt-10% Rh crucible inside a Deltech DT-31-RS furnace (Denver, Colorado). The impedance probe used for these measurements consisted of two paddles made of Pt-10% Rh (7 mm wide × 12.7 mm long) spaced 9.32 mm apart. Duplicate impedance measurements were performed at each temperature. Prior to the measurements, the impedance analyzer had been calibrated with 0.1 M and 1 M KCl reference solutions at room temperature to determine the cell constant. The glass conductivity results are presented in Figure 7.7 together with conductivities measured by VSL in HWI-Al-19 glass from a crucible melt, since conductivity data was not reported for glass from their melters (Matlack et al. 2008). The electrical conductivities of the glass melted in the RSM-6 at PNNL are consistent with the conductivities measured by VSL, with PNNL measuring only slightly lower values.

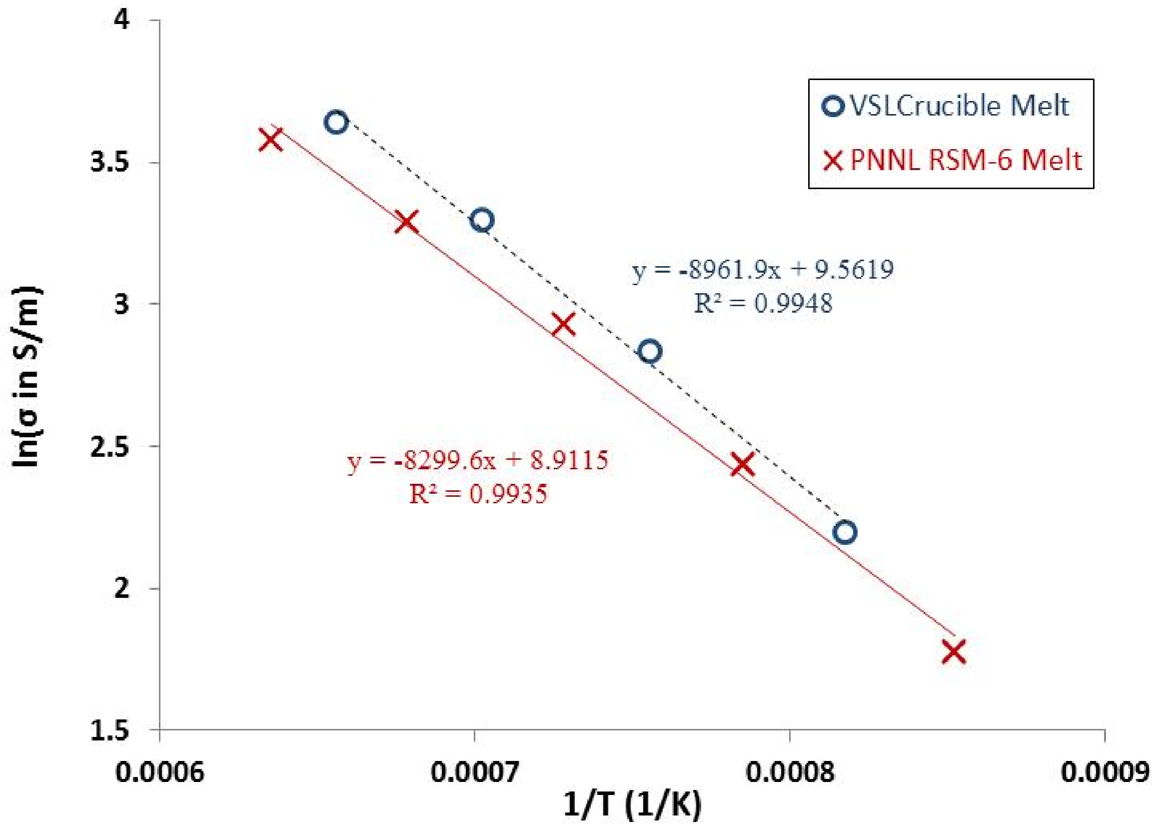


Figure 7.7. Electrical Conductivity of HWI-Al-19 Glass

7.7 Viscosity

Viscosity (η) of the glass was measured as a function of temperature using a rotating-spindle digital viscometer according to PNNL procedure GDL-Visc-Test-01 (Crum 2012b). Prior to viscosity measurements the instrument was calibrated using DWPF start-up frit (Crum et al. 2012) and PNNL procedure GDL-Visc-Test-01 (Crum 2012b). GDL-Visc-Test-01 complies with ASTM C965 Method A such that the spindle was rotated and the crucible was fixed (ASTM C-965-94 1994). Fifty mL of glass from the sample labeled RSM-EWG1-034 was measured and heated to ~1150°C in a Pt-10% Rh crucible and maintained until thermal equilibrium was reached. An initial torque reading at a constant spindle speed was taken at ~1370°C, with subsequent measurements taken as the glass was cooled to ~1050°C. The measured viscosities are plotted as a function of temperature in Figure 7.8 together with viscosity measurements performed by VSL on glass melted in a crucible at their facility, since no viscosity data was reported on the glass processed in their melters (Matlack et al. 2008). There is very close agreement between the viscosities measured at the two laboratories.

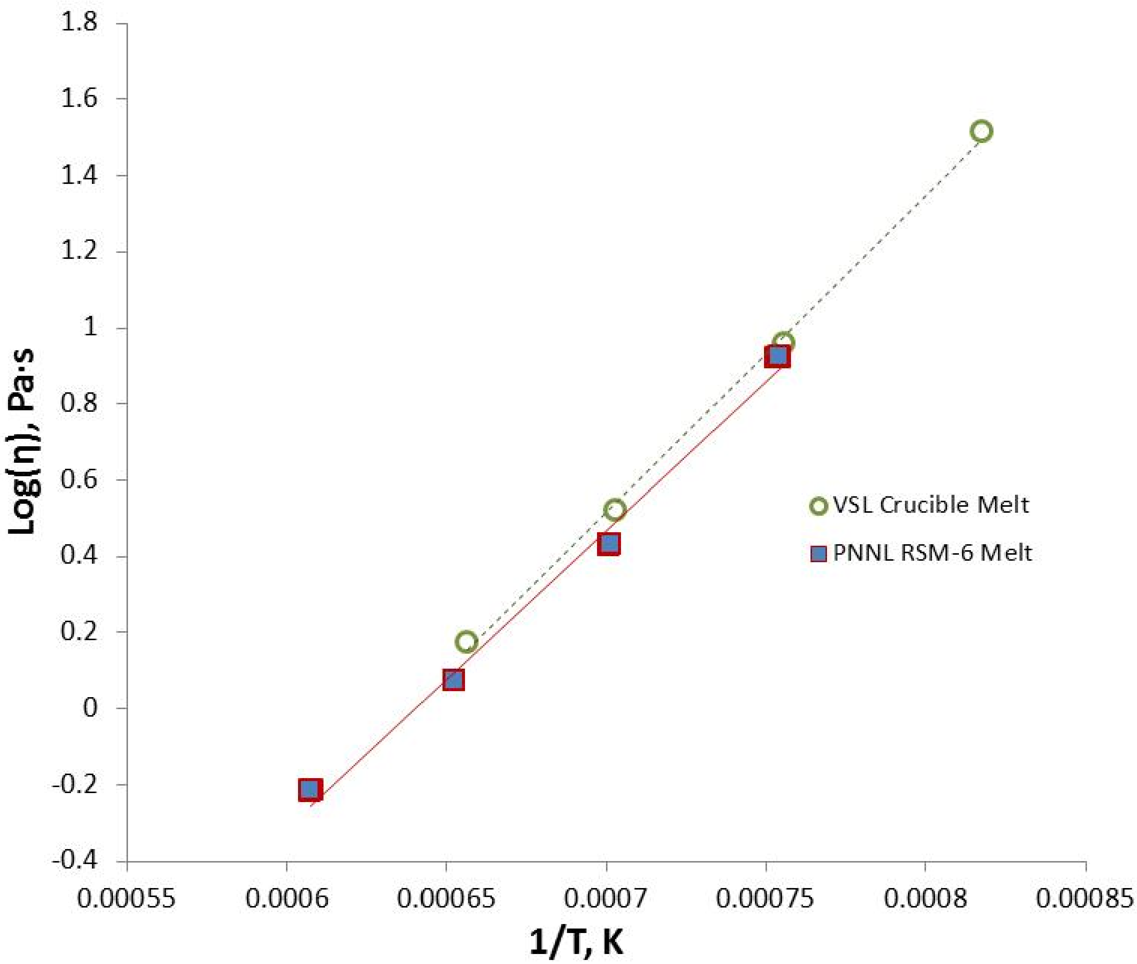


Figure 7.8. Viscosity of HWI-Al-19 Glass as a Function of Temperature

8.0 Feed and Scrubber Solution Analysis

Two feed samples were analyzed by ICP-OES at SwRI to verify the feed prepared by the supplier. As shown in Table 8.1, the analyzed compositions agreed well with the target composition. In Table 8.2, the densities of feed samples from RSM-10 tests were slightly higher than those from RSM-6, likely due to water evaporation. The results of EVS solution sample analyses by IC are given in Table 8.3, which show that the concentrations of these volatile and semi-volatile components are higher in the samples from the RSM-10 than from the RSM-6, which is as expected.

Table 8.1. Feed Sample Compositions (wt%) Measured by ICP-OES

Segment:		**Pre-RSM-6**	**RSM-6 #2**
Sample #:	**Target**	**RSM-EWG1-002**	**RSM-EWG1-032**
Al_2O_3	**8.73**	9.88	9.88
B_2O_3	**6.99**	8.08	7.47
BaO	**0.02**	0.02	0.02
Bi_2O_3	**0.42**	0.46	0.44
CaO	**2.10**	2.32	2.27
CdO	**0.01**	0.01	0.01
Cr_2O_3	**0.19**	0.15	0.15
Fe_2O_3	**2.13**	2.56	2.49
K_2O	**0.001**	0.055	0.056
Li_2O	**1.32**	1.26	1.27
MgO	**0.04**	0.09	0.09
MnO	**---**	0.01	0.01
Na_2O	**3.50**	3.52	3.49
NiO	**0.15**	0.17	0.17
P_2O_5	**0.38**	0.48	0.43
PbO	**0.15**	0.16	0.16
SO_3	**0.07**	0.09	0.10
SiO_2	**9.84**	10.27	10.46
TiO_2	**0.004**	0.020	0.019
ZnO	**0.03**	0.04	0.04
ZrO_2	**0.15**	0.18	0.18

Table 8.2. Feed Densities

Segment:	**Pre-RSM-6**	**RSM-6 #1**	**RSM-6 #1**	**RSM-6 #2**	**Pre-RSM-10**	**Post-RSM-10**
Sample #:	**RSM-EWG1-002**	**RSM-EWG1-017**	**RSM-EWG1-025**	**RSM-EWG1-032**	**RSM-EWG2-001**	**RSM-EWG2-023**
Density	1.40	1.41	1.41	1.41	1.49	1.43

Table 8.3. EVS Solution Sample Compositions (wt%) Measured by IC

Segment:	**Pre-RSM-6**	**RSM-6 #1**	**RSM-6 #1**	**RSM-6 #2**	**Post-RSM-6**	**Pre-RSM-10**	**RSM-10 #1**	**Post-RSM-10**	**Post-RSM-10**
Sample #:	**RSM-EWG1-003**	**RSM-EWG1-020**	**RSM-EWG1-026**	**RSM-EWG1-033**	**RSM-EWG1-040**	**RSM-EWG2-002**	**RSM-EWG2-016**	**RSM-EWG2-022**	**RSM-EWG2-030**
F	0.002	0.018	0.024	0.033	0.037	0.027	0.056	0.070	0.082
NO_3	ND[(a)]	0.009	0.012	0.014	0.016	0.024	0.020	0.021	0.022
NO_2	ND	ND	ND	ND	ND	0.007	0.008	0.007	0.006
SO_4	0.013	0.024	0.029	0.034	0.038	0.038	0.042	0.048	0.055

(a) ND = not detected; Br and PO_4 were not detected in any sample.

9.0 Quality Assurance

The PNNL quality assurance program description implements both DOE Order 414.1D (DOE 2011) and 10 CFR 830, Subpart A (10 CFR 830). PNNL has also adopted NQA-1-2000, *Quality Assurance Program for Nuclear Facilities* (ASME 2000), as its single consensus standard for implementation of quality assurance requirements, and graded in accordance with NQA-1-2000, Subpart 4.2, “Guidance on Graded Application of Quality Assurance (QA) for Nuclear-Related Research and Development.” PNNL’s standards-based management system—*How Do I*? (HDI)—is a web-based system for communicating the quality assurance program requirements through Laboratory-wide procedures or subject areas. All work at PNNL is subject to the applicable HDI requirements. In the facilities where work in support of this project was conducted, PNNL’s “Integrated Operations System” was used to implement HDI and safety procedures at the benchtop. As part of the graded approach to quality assurance, this project has a formal Quality Assurance Plan (QAP) that specifies project-specific quality procedures covering technical work.

All analytical project work was performed following the latest HASQARD. PNNL subcontracted to SwRI for analytical services that will require HASQARD compliance. PNNL has audited and accepted SwRI services as compliant with the HASQARD requirements and has placed SwRI on the PNNL Evaluated Suppliers List as an acceptable supplier for analytical services in accordance with HASQARD.

10.0 Conclusions

The RSM tests in this study provided glass samples and basic operational data for comparison to those reported from studies performed in the larger DM-100 and DM-1200 melters at VSL using the same HWI-Al-19 HLW glass simulant feed composition at the same target melt temperature. It was demonstrated that when the RSM was operated under parameters that were generally aligned with those of the larger melters at VSL, similar processing rates and characteristics could be achieved, on a melt-surface-area–adjusted basis. The tests demonstrated the process flowsheet through a small-scale integrated process by obtaining steady feeding operations for a prolonged period.

The RSM melter was operated with a target glass temperature of 1150°C, corresponding to the target glass temperature VSL used to process the HWI-Al-19 feed in their melters, and target plenum temperature between 350 and 650°C, corresponding to the approximate minimum actual plenum temperature measured by VSL and the maximum of their targeted range. The averages of the glass melt temperatures measured during the test segments in the RSM and VSL melters differed from one another and from the target temperature by less than 3%. Measured plenum temperatures ranged from 479 to 970°C at PNNL and from 343 to 986°C at VSL. The average surface-area-specific bubbling rates during test segments at PNNL fell within the range of those at VSL, except during the second RSM-6 test segment, in which the target bubbling rate was intentionally decreased to determine the effect on glass production rate. A direct relationship between bubbling rate and feed/production rate was observed at both labs.

The average glass production rates were 0.71 and 0.82 kg/h in the two RSM-6 test segments and 1.95 kg/h in the RSM-10, resulting in a melter surface-area-normalized glass generation rate of 922 to 1077 kg/day/m^2 for all RSM tests, which fell within the range of 896 to 1557 kg/day/m^2 reported for the VSL melters. However, during periods of steady-state operation in the RSM, the production rates calculated from feed rates fell within the range of rates calculated by VSL for steady-state operation in VSL melters, except in the case of segment 2 of the RSM-6 test, in which the calculated steady-state production rate was lower due to the decreased bubbling rate employed during this segment. The average feed rates in the RSM melters were within the range of those reported by VSL with the exception of the RSM-10 test, which only fell below the minimum of this range by less than 7 kg/h/m^2(< 1%). The average power use for glass production in the RSM melters also was within the range reported by VSL. Overall, 64 kg and 101 kg of glass were produced during operation of the RSM-6 and RSM-10, respectively.

The glasses processed at both facilities had PCT and TCLP responses well below the regulatory limits. The electrical conductivity and viscosity of the glasses processed at PNNL and VSL were very similar. Overall, this work demonstrates that the smaller-scale RSM can be used to support and supplement waste vitrification research while requiring less material and shorter turnaround times than the larger melters, yet providing comparable experimental results.

11.0 References

10 CFR 830. 2011. Code of Federal Regulations, Title 10, *Energy*, Part 830, "Nuclear Safety Management," Subpart A – Quality Assurance Requirements. U.S. Department of Energy. Accessed September 14, 2015 at http://www.gpo.gov/fdsys/search/pagedetails.action?collectionCode=CFR&searchPath=Title+10%2FChapter+III%2FPart+830%2FSubpart+A&granuleId=CFR-2011-title10-vol4-part830&packageId=CFR-2011-title10-vol4&oldPath=Title+10%2FChapter+III%2FPart+830%2FSubpart+A&fromPageDetails=true&collapse=false&ycord=1136.

ASME–American Society of Mechanical Engineers. 2000. *Quality Assurance Requirements for Nuclear Facility Applications*. ASME-NQA-1-2000. New York, NY.

ASTM— American Society of Testing and Materials. 1994. *Standard Practice for Measuring Viscosity of Glass Above the Softening Point.* ASTM C965-94(1994), American Society of Testing and Materials.

ASTM—American Society of Testing and Materials. 2008. *Standard Test Methods for Determining Chemical Durability of Nuclear, Hazardous, and Mixed Waste Glasses and Multiphase Glass Ceramics: The Product Consistency Test (PCT)*. ASTM C 1285-02(2008), American Society of Testing and Materials.

Barnes SM and DE Larson. 1981. *Materials Design Experience in a Slurry-Fed Electric Glass Melter*. PNL-3959, Pacific Northwest Laboratory, Richland, WA.

Crum JV. 2012a. *PNWD Procedure: High-Temperature Electrical Conductivity*, GDL-Elec-Test-01, Rev. 0. Pacific Northwest National Laboratory, Richland, WA.

Crum JV. 2012b. *PNWD Procedure: High-Temperature Viscosity Measurement*, GDL-Visc-Test-01, Rev. 0, Pacific Northwest National Laboratory, Richland, WA.

Crum JV, TB Edwards, RL Russell, PJ Workman, MJ Schweiger, RF Schumacher, DE Smith, DK Peeler, and JD Vienna. 2012. "DWPF Startup Frit Viscosity Measurement Round Robin Results." *J. Am. Ceram. Soc*. 95(7):2196-2205.

DOE—U.S. Department of Energy, Office of Environmental Management. 1996. *Waste Acceptance Product Specifications for Vitrified High-Level Waste Forms*. EM-WAPS Rev. 2.

DOE—U.S. Department of Energy, Office of River Protection, Richland, WA. 2006. *Test and Evaluate High Level Waste (HLW) Vitrification System Improvements*. Contract Number DE-AC27-06RV14790.

DOE—U.S. Department of Energy. 2011. Order 414.1D, Quality Assurance. U.S. Department of Energy, Washington, D.C. Accessed September 14, 2015, at https://www.directives.doe.gov/directives-documents/400-series/0414.1-BOrder-d.

Ebert WL and SF Wolfe. 1999. *Round-robin Testing of a Reference Glass for Low-Activity Waste Forms*. U.S. Department of Energy Report ANL-99/22, Argonne National Laboratory, Argonne, IL.

EPA—U.S. Environmental Protection Agency. 1992. *Method 1311: Toxicity Characteristic Leaching Procedure*. Washington, D.C.

EPA—U.S. Environmental Protection Agency. 1994. *Method 9056: Determination of Inorganic Anions by Ion Chromatography*. Washington, D.C.

Fox KM and TB Edwards. 2014. *Chemical Composition and PCT Data for the Initial Set of Hanford Enhanced Waste Loading Glasses*. SRNL-STI-2014-00063, Savannah River National Laboratory, Aiken, SC.

Goles RW and AJ Schmidt. 1992. *Evaluation of Liquid-Fed Ceramic Melter Off-Gas System Technologies for the Hanford Waste Vitrification Plant.* PNL-8109, Pacific Northwest Laboratory, Richland, WA.

Joseph I, BW Bowan, H Gan, WK Kot, KS Matlack, IL Pegg, and AA Kruger. 2010. *High Aluminum HLW Glasses for Hanford's WTP*. Energy*Solutions*, Inc., Columbia, MD.

Kim DS, MJ Schweiger, CP Rodriguez, WC Lepry, JB Lang, JV Crum, JD Vienna, FC Johnson, JC Marra, and DK Peeler. 2011a. *Formulation and Characterization of Waste Glasses with Varying Processing Temperature.* PNNL-20774, Pacific Northwest National Laboratory, Richland, Washington.

Kim D, MJ Schweiger, WC Buchmiller, and J Matyas. 2011b. *Laboratory-Scale Melter for Determination of Melting Rate of Waste Glass Feeds*. PNNL-21005 (EMSP-RPT-012), Pacific Northwest National Laboratory, Richland, WA.

Matlack KS, H Gan, M Chaudhuri, W Kot, W Gong, T Bardacki, IL Pegg, and I Joseph. 2008. *Melt Rate Enhancement for High Aluminum HLW Glass Formulations.* VSL-08R1360-1, Vitreous State Laboratory, The Catholic University of America, Washington, DC.

Matlack KS, WK Kot, W Gong, W Lutze, IL Pegg, and I Joseph. 2009. *Effects of High Spinel and Chromium Oxide Crystal Contents on Simulated HLW Vitrification in DM100 Melter Tests.* VSL-09R1520-1, Vitreous State Laboratory, The Catholic University of America, Washington, DC.

Peeler DK and TB Edwards. 2013. *An Analytical Plan for Measuring the Chemical Compositions of an Initial Set of Glasses Supporting Hanford HLW Glass Development and Characterization*. U.S. Department of Energy Memorandum SRNL-L3100-2013-00205, Savannah River National Laboratory, Aiken, SC.

Rose PB, DI Woodward, MI Ojovan, NC Hyatt, and WE Lee. 2011. "Crystallisation of a simulated borosilicate high-level waste glass produced on a full-scale vitrification line." *J. Non-Cryst. Solids*, 357(15):2989-3001.

Schweiger MJ, P Hrma, CJ Humrickhouse, J Marcial, BJ Riley, and NE TeGrotenhuis. 2010. "Cluster Formation of Silica Particles in Glass Batches during Melting," *J. Non-Cryst. Solids* 356:359–1367.

Smith GL, D Kim, MJ Schweiger, JC Marra, JB Lang, JV Crum, CL Crawford, and JD Vienna. 2014. *Silicate Based Glass Formulations for Immobilization of U.S. Defense Wastes Using Cold Crucible*

Induction Melters. PNNL-23288 (EMSP-RPT-021), Pacific Northwest National Laboratory, Richland, WA.

Appendix A

Operating Parameter Data Plots from RSM Tests

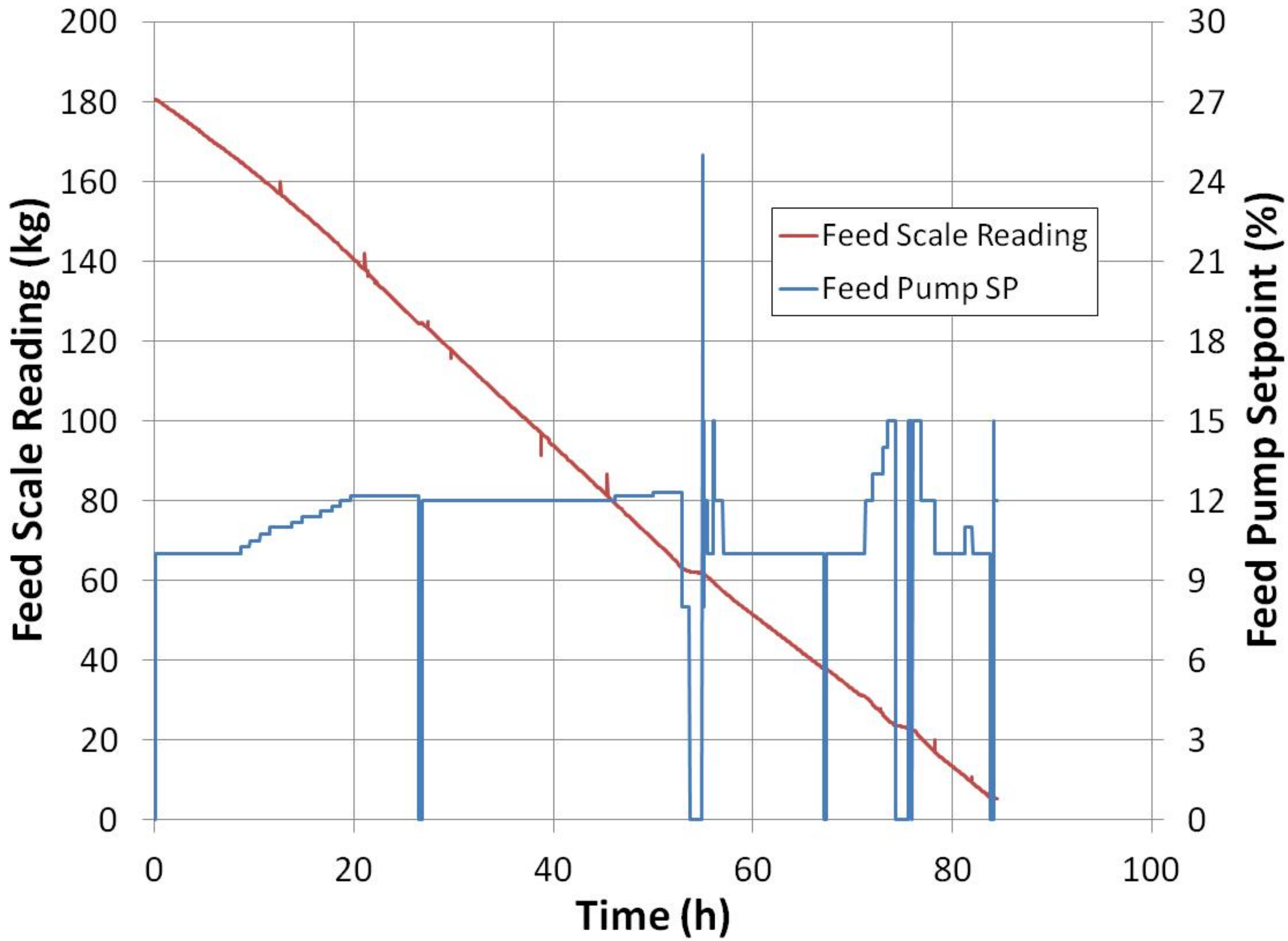


Figure A.1. Simulant Feed Pump Set Point and Balance Reading for RSM-6

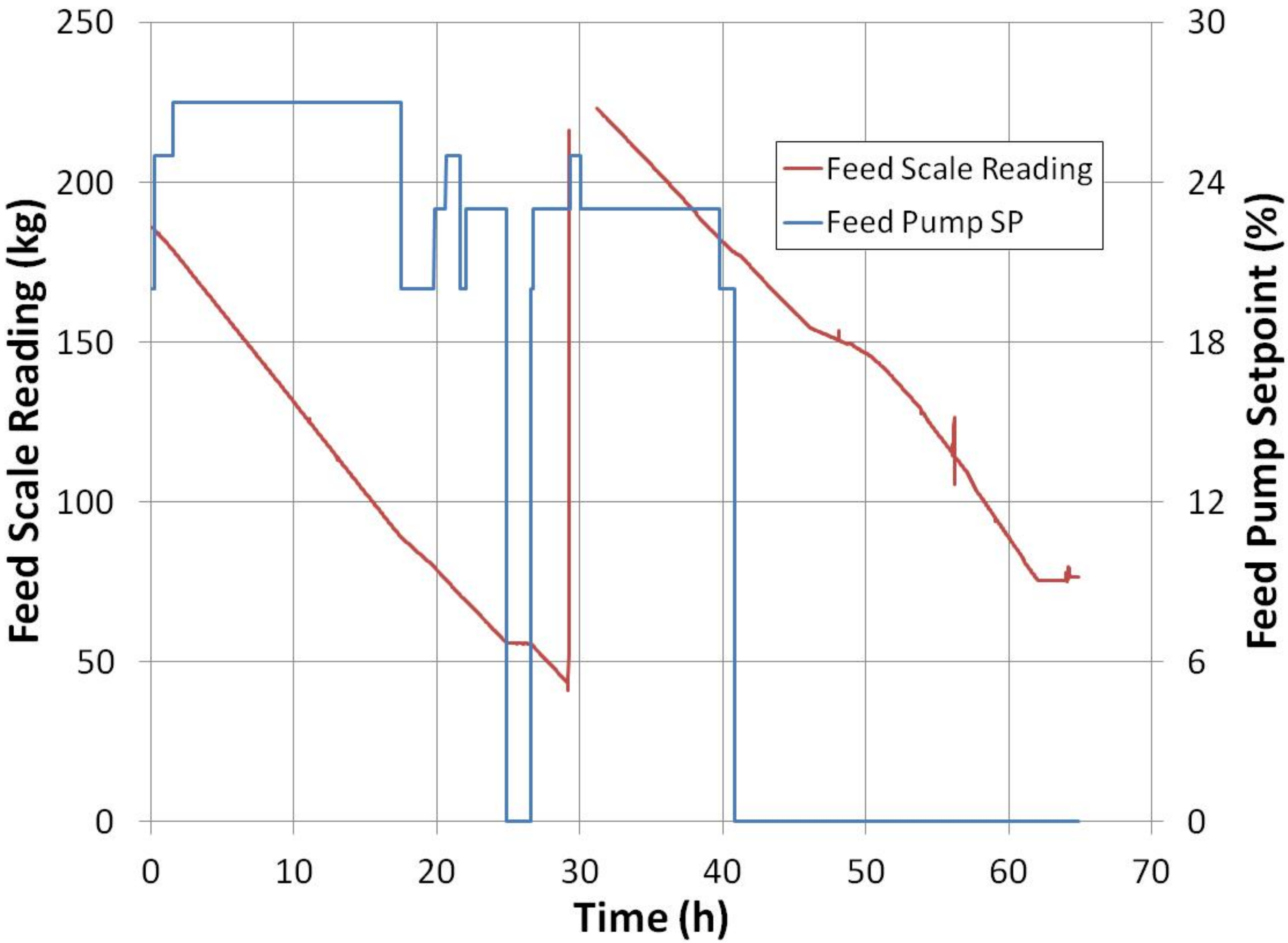


Figure A.2. Simulant Feed Pump Set Point and Balance Reading for RSM-10

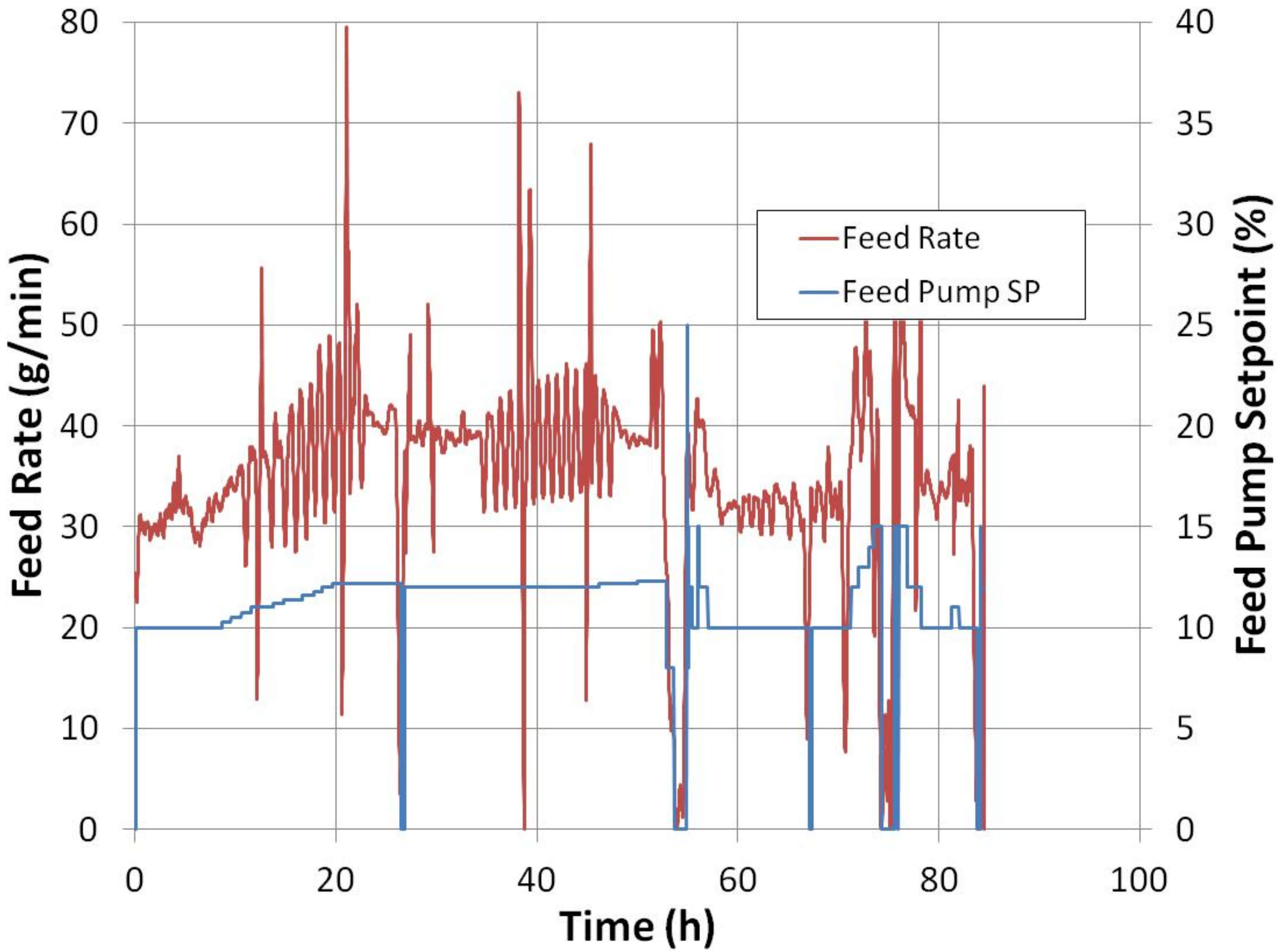


Figure A.3. Simulant Feed Pump Set Point and Calculated Feed Rate (30 minute average) for RSM-6

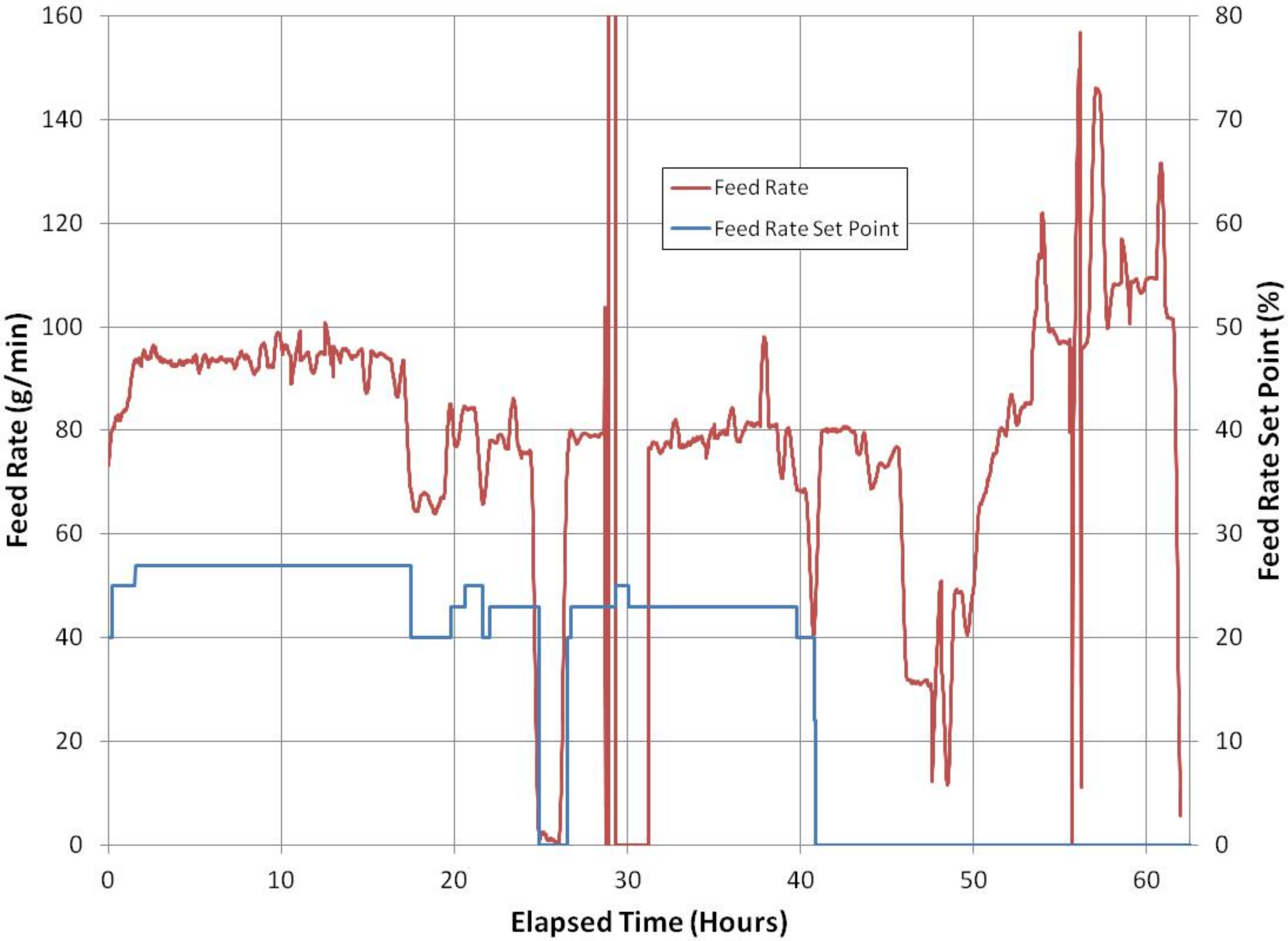


Figure A.4. Simulant Feed Pump Set Point and Calculated Feed Rate (30 minute average) for RSM-10

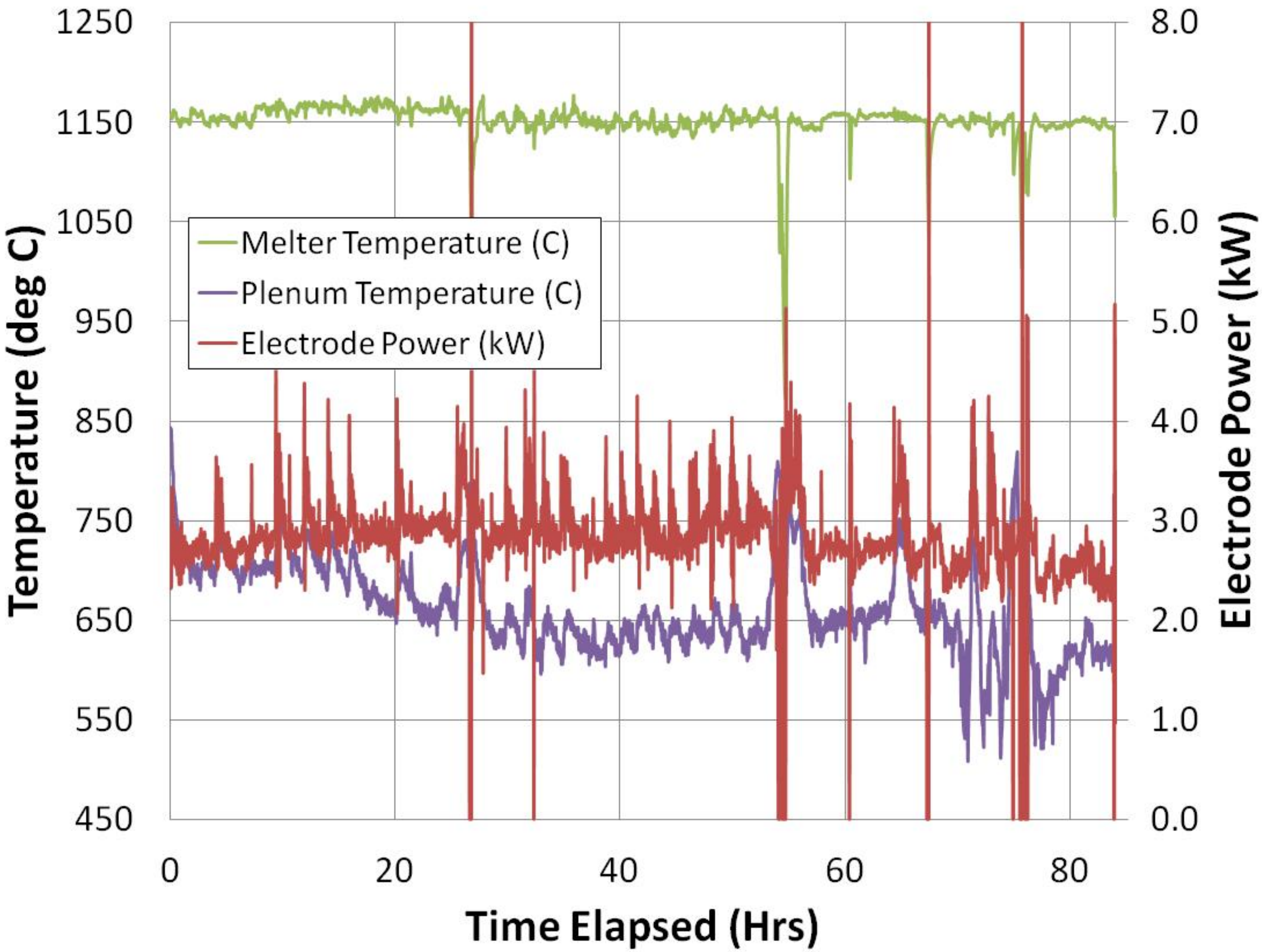


Figure A.5. Melter and Plenum Temperatures and Electrode Power for RSM-6

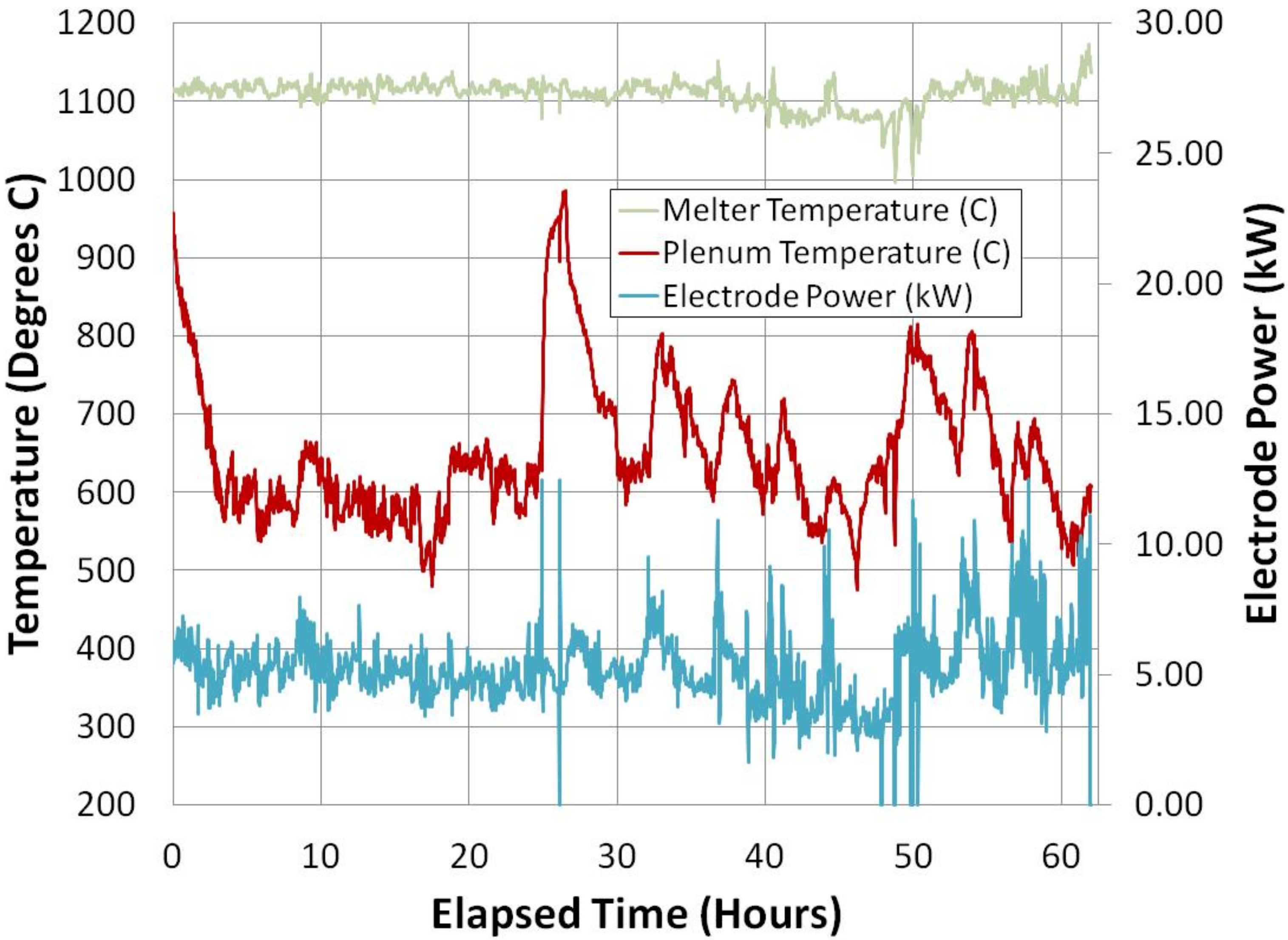


Figure A.6. Melter and Plenum Temperatures and Electrode Power for RSM-10

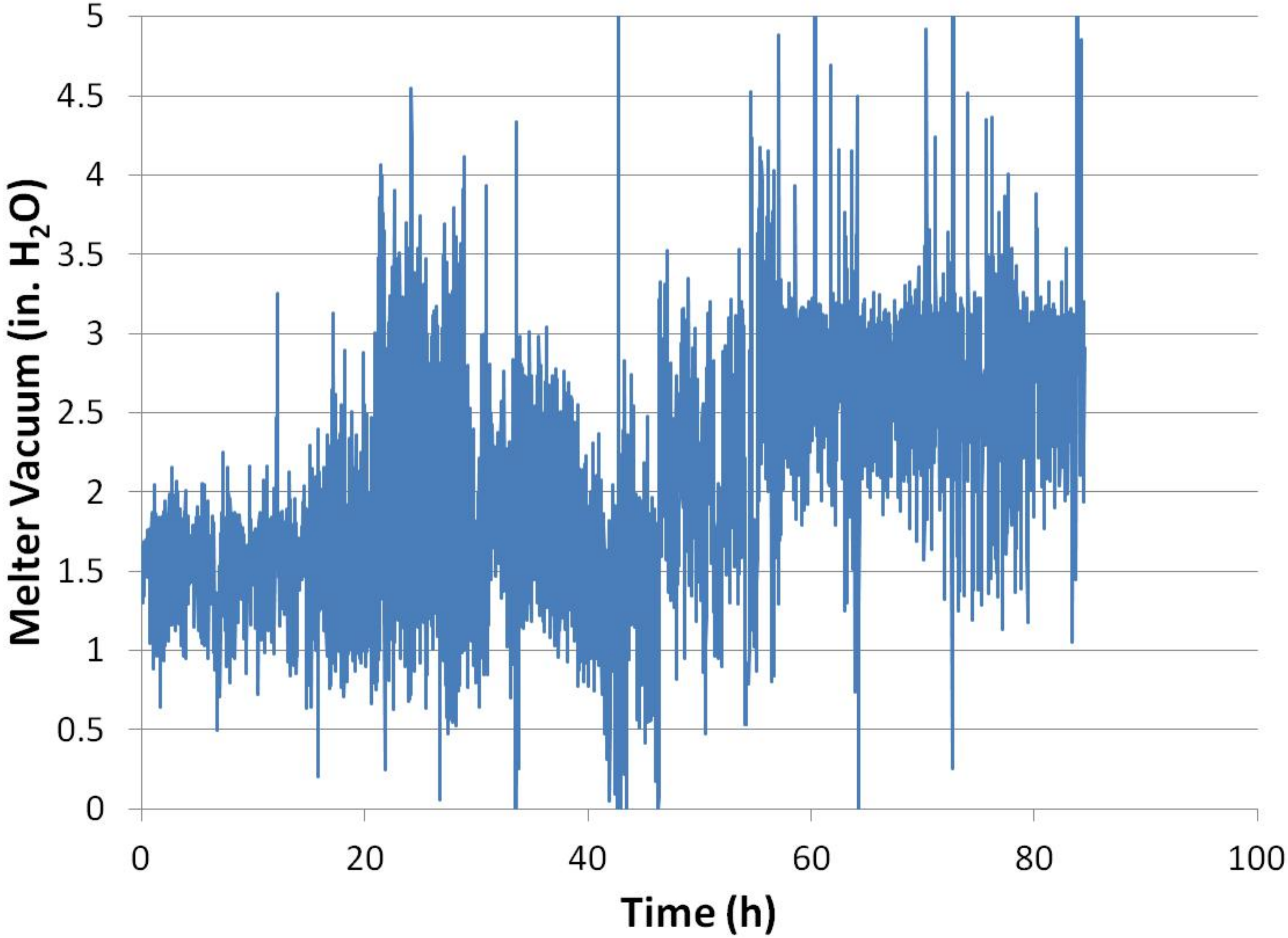


Figure A.7. Melter Vacuum in RSM-6

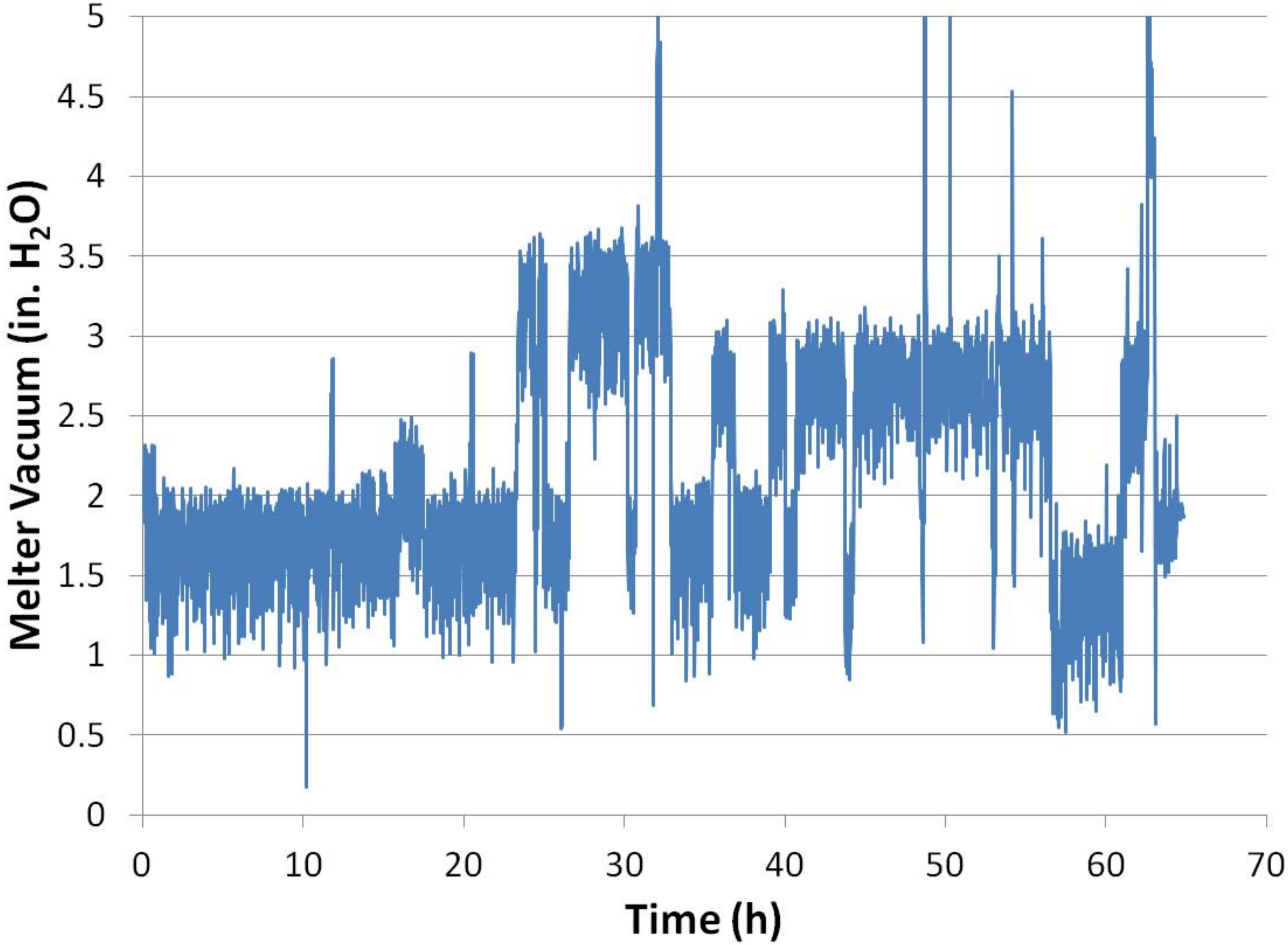


Figure A.8. Melter Vacuum in RSM-10

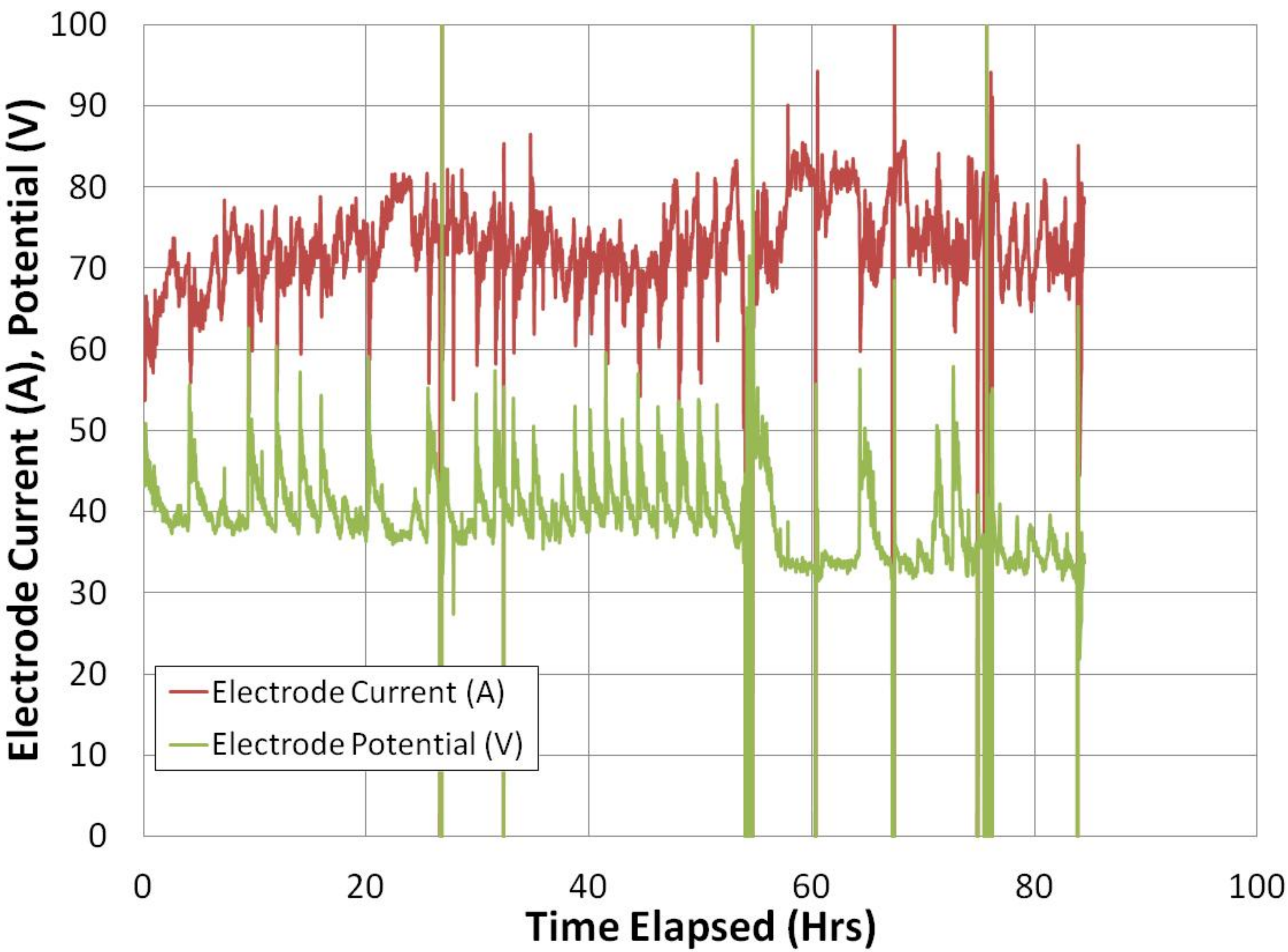


Figure A.9. Electrode Current and Potential for RSM-6

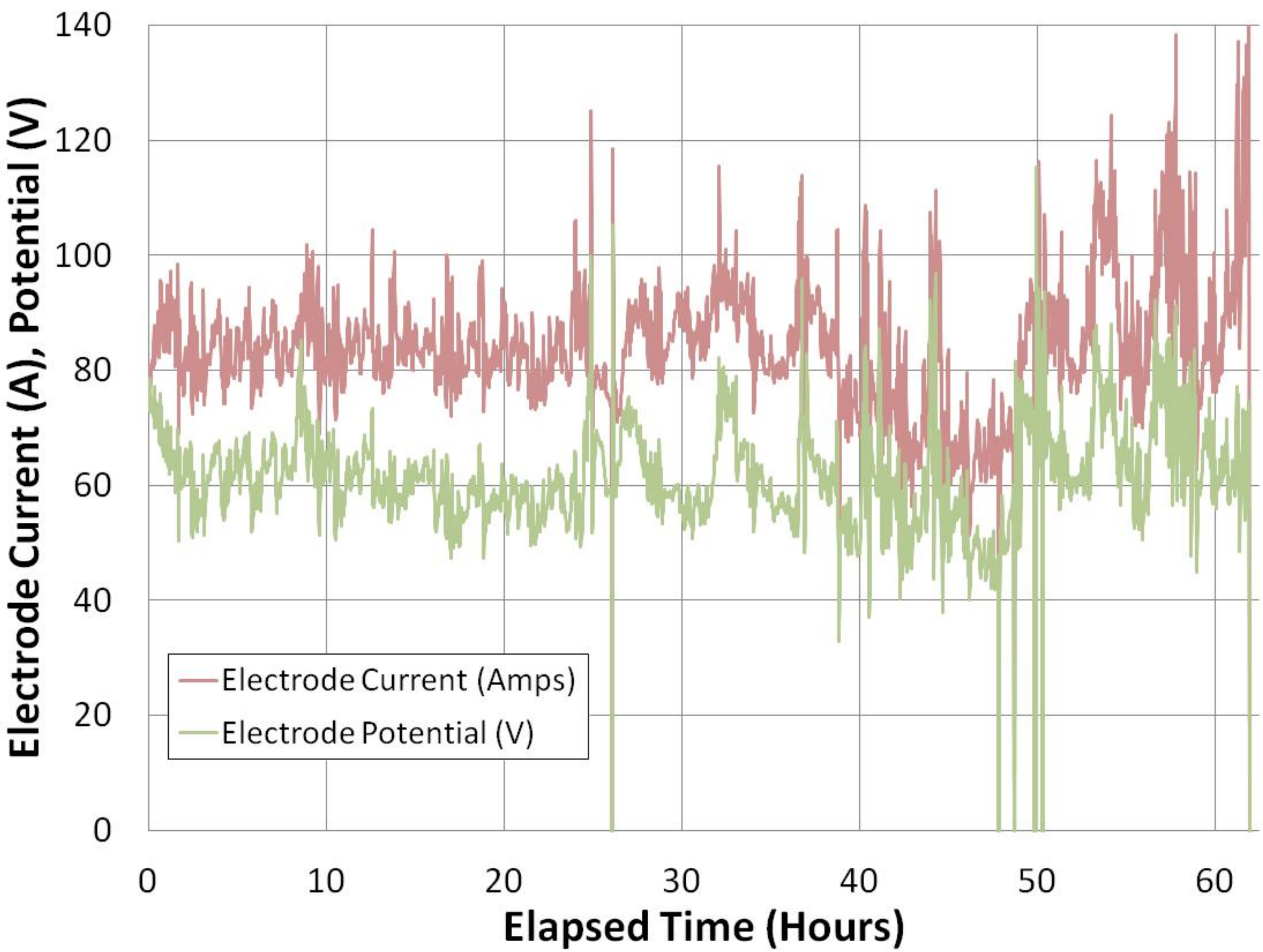


Figure A.10. Electrode Current and Potential for RSM-10

Distribution

ORP Personnel:

Elaine Diaz
Bill Hamel
Benton (Ben) Harp
Albert Kruger
Kevin Smith

EM Headquarters:

Rod Rimando

Idaho National Laboratory:

Donna Guillen

Prague (UCT):

Richard Pokorny

Pacific Northwest National Laboratory

Will Eaton
Jaime George
Pavel Hrma
Tony Jin
Dongsang Kim
Jarod Kroll
Seungmin Lee
Charmayne Lonergan
Benjamin McCarthy
David Peeler
Renee Russell
Steve Schlahta
Gary L. Smith (APEL/96)
Cody Stewart
John Vienna
Information Release

Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** *Since 1965*

U.S. DEPARTMENT OF
ENERGY

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99352
1-888-375-PNNL (7665)
www.pnl.gov