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Vitrification of Hanford Tank Wastes for Condensate Recycle and Feed Composition Changeover Testing

December 2021

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U.S. DEPARTMENT OF ENERGY

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Richland, Washington 99354

Summary

During the vitrification of Hanford Site nuclear waste at the Waste Treatment and Immobilization Plant (WTP), which is a part of the safe and efficient retrieval, treatment, and disposal mission of the U.S. Department of Energy Office of River Protection, the offgas condensate generated from the waste-to-glass conversion is currently planned to be concentrated by evaporation in the Effluent Management Facility (EMF). This concentrated condensate can then be recycled back to the incoming waste and vitrified.

To test the recycle process, an apparatus was designed and built to mimic the EMF evaporator and was then used to concentrate a volume of condensate produced during the vitrification of a sample of Hanford tank 241-AP-107 (referred to herein as AP-107) waste in a continuous laboratory-scale melter (CLSM). The concentrated condensate was added to an additional sample of AP-107 waste, to mimic one round of the recycle process, and the combined solution was vitrified, producing a second round of recycle condensate.

In the current study, the EMF test apparatus was used to concentrate the second-round recycle condensate under evaporation conditions (at 45 °C and 1.4 psia) designed to emulate EMF operation. The condensate was successfully concentrated by a factor of ~10 while retaining over 95 % of the technetium-99 (^{99}Tc), Cs, and I inventories in the concentrate. Another portion of AP-107 waste was retrieved by Washington River Protection Solutions, LLC (WRPS) and transferred to Pacific Northwest National Laboratory (PNNL), where it was pretreated and then combined with the second-round recycle AP-107 condensate concentrate and glass-forming chemicals (GFCs) to form the two-time recycle AP-107 melter feed, approximating a second round to the recycling action to be performed at the WTP. A portion of AP-105 waste was also retrieved by WRPS and provided to PNNL for pretreatment and combining with GFCs to form AP-105 melter feed.

The two-time recycle AP-107 and AP-105 melter feeds were processed consecutively in the CLSM. The CLSM run proceeded for 13.63 hours, producing 9.70 kg of glass for an average glass production rate of 1464 kg m^2 d^{-1} during the two-time recycle AP-107 feed charging and 1568 kg m^2 d^{-1} during the AP-105 feed charging. The rate during AP-107 charging was essentially equivalent to the rate when processing no-recycle AP-107 feed[1] and lower than that achieved when processing one-time recycle AP-107 feed.[2] However, all rates were within the potential range of variability when processing melter feeds with similar composition in the CLSM. Likewise, the rate during AP-105 charging was higher than the previous rate[3] processing AP-105, but within the potential CLSM range. The cold-cap characteristics changed from the typically thin AP-107 cold cap to a foamy-edged cold cap as previously seen with AP-105 shortly after transitioning to the AP-105 melter feed.

The glass produced during the CLSM run was within 10 % of its target composition for the primary glass components. The CaO and Li_2O targets varied by more than 1 wt% between the two-time recycle AP-107 and AP-105 glass targets and it took about 2 turnovers of the CLSM glass inventory to reach a relative chemical steady state in the glass for CaO and Li_2O after the melter feed inputs were switched.

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[1] Dixon et al. 2019. *Vitrification of Hanford Tank Waste 241-AP-107 in a Continuous Laboratory-Scale Melter*. PNNL-28361, Rev. 0 (RPT-DFTP-014, Rev. 0). Pacific Northwest National Laboratory, Richland, Washington.

[2] Dixon et al. 2020. *Vitrification of Hanford Tank 241-AP-107 with Recycled Condensate*. PNNL-30189, Rev. 0 (RPT-DFTP-024, Rev. 0). Pacific Northwest National Laboratory, Richland, Washington.

[3] Dixon et al. 2018. *Vitrification of Hanford Tank Waste 241-AP-105 in a Continuous Laboratory-Scale Melter*. PNNL-27775 (RPT-DFTP-010, Rev. 0). Pacific Northwest National Laboratory, Richland, Washington.

The ^{99}Tc and total cesium content in the melter feeds were maintained at concentrations expected to be experienced at the WTP. During the CLSM run, while processing the two-time recycle AP-107 melter feed at a relative chemical steady state, the ^{99}Tc/Cs ratio was 10, and 34% of ^{99}Tc and 74% of Cs were retained in the glass. These values were higher than those measured in the CLSM run with one-time recycle AP-107 melter feed. After the transition to processing the AP-105 melter feed, when the production reached a relative chemical steady state, the ^{99}Tc/Cs ratio was 77 while 44% of ^{99}Tc was retained in the glass. The Cs retention during this time frame reached 200% due to the excess Cs in the glass after the target content decreased to 15% of its initial level in the two-time recycle AP-107 melter feed to the lower target in the AP-105 melter feed. While iodine was below inductively coupled plasma mass spectrometry analytical reporting limits in the melter feed and glass samples, it was detected in quantities above the analytical reporting limits in the liquid and filter samples collected from the CLSM offgas treatment system. The behavior of iodine in the CLSM offgas treatment system followed a similar pattern to those of ^{99}Tc and Cs.

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Acronyms and Abbreviations

APEL	Applied Process Engineering Laboratory
ARL	analytical reporting limit
CA	contamination area
CLSM	continuous laboratory-scale melter
DF	decontamination factor
DFLAW	direct-feed low-activity waste
DM10	DuraMelter10
DOE-ORP	U.S. Department of Energy Office of River Protection
EMF	Effluent Management Facility
ETF	Effluent Treatment Facility
GFC(s)	glass-forming chemical(s)
HCA	high contamination area
HEPA	high-efficiency particulate air (filters)
HLW	high-level waste
IC	ion chromatography
ICP-AES	inductively coupled plasma atomic emission spectroscopy
ICP-MS	inductively coupled plasma mass spectrometry
LAW	low-activity waste
PES	polyethersulfone
PNNL	Pacific Northwest National Laboratory
psia	pounds per square inch absolute
QA	quality assurance
R	retention
R&D	research and development
Rec	recovery
RPL	Radiochemical Processing Laboratory
SBS	submerged-bed scrubber
sccm	standard cubic centimeters per minute
SwRI	Southwest Research Institute
TC	thermocouple
TIC	total inorganic carbon
TOC	total organic carbon
TSCR	Tank Side Cesium Removal
WRPS	Washington River Protection Solutions, LLC
WTP	Waste Treatment and Immobilization Plant
WWFTP	WRPS Waste Form Testing Program

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1.0 Introduction

It is the responsibility of the U.S. Department of Energy-Office of River Protection (DOE-ORP) to safely and efficiently retrieve, treat and dispose approximately 56 million gallons of radioactive waste from 177 underground tanks located on the Hanford Site in Washington State. The Hanford waste tanks are currently operated and managed by Washington River Protection Solutions, LLC (WRPS). As part of tank farm operations, WRPS supports DOE-ORP's waste retrieval mission. An important element of the DOE-ORP mission is the construction and operation of the Waste Treatment and Immobilization Plant (WTP), which will process and stabilize tank waste. Currently, the first phase of the planned WTP startup and operation, called direct-feed low-activity waste (DFLAW), involves directly processing only the liquid supernatant portion of the waste by vitrification in electric melters in the WTP low-activity waste (LAW) facility without full pretreatment (Bernards et al. 2017). A second portion of the tank waste, called high-level waste (HLW), is set to contain most of the radioactivity inventory (Bernards et al. 2017).

To meet the acceptance criteria at the WTP LAW facility, WRPS is designing a Tank Side Cesium Removal (TSCR) system to remove suspended solids and cesium (Cs/^{137}Cs) from the supernatant (Bernards et al. 2017). After these processes, the waste will be combined with glass-forming chemicals (GFCs) to form a mixed aqueous and solid slurry, called melter feed, that can be charged into the melters. During vitrification, a stable glass is produced for disposal while water, volatile waste components, and a portion of semi-volatiles from the waste-to-glass conversion process escape to the offgas treatment system, where they are captured, primarily as condensate. This offgas condensate will then be concentrated by evaporation in the Effluent Management Facility (EMF) and recycled back to the LAW facility to be incorporated into the melter feed. Recycled radionuclides technetium-99 (^{99}Tc) and iodine-129 (^{129}I) are expected to accumulate in the offgas treatment waste stream. Under normal operations, the evaporator bottoms will be returned to the LAW melter facility but could also be returned to the tank farm without evaporation when the EMF evaporator is unavailable. The evaporator overhead condensate will be sent to the Effluent Treatment Facility (ETF).

A test program was established at Pacific Northwest National Laboratory (PNNL) to conduct scaled unit operation process steps with actual Hanford tank waste (Peterson et al. 2017). To facilitate this program, the Radioactive Waste Test Platform was established to allow for baseline and alternative flowsheets and unit operations to be tested in comparable tests where both the direct effect of changes and the downstream effects of changes could be evaluated. As a part of this platform, a continuous laboratory-scale melter (CLSM) system was designed and constructed in the Radiochemical Processing Laboratory (RPL) at PNNL for vitrifying treated tank waste samples. An identical, duplicate CLSM system (to process and study non-radioactive waste simulants) was also constructed in the Applied Process Engineering Laboratory (APEL) at PNNL, and a study was performed to evaluate the system performance (Dixon et al. 2020a).

The first portion of waste received for vitrification was from tank 241-AP-105 (hereafter called AP-105). The AP-105 waste was filtered to remove solids (Geeting et al. 2018a), it underwent ion exchange to remove cesium (Fiskum et al. 2018), had GFCs added and was vitrified in the CLSM (Dixon et al. 2018), and the condensate produced from vitrification was concentrated and converted to a non-glass waste form based on the Cast Stone waste form formulation (Cantrell et al. 2018). A second portion of waste received was the supernatant from Hanford tank 241-AP-107 (hereafter called AP-107). This AP-107 waste went through solids removal by filtration (Geeting et al. 2018b) and cesium removal by ion exchange (Westesen et al. 2021). After these activities, the Kim et al. (2012) model for WTP baseline glass formulation was used to calculate the mass of GFCs to be added to the AP-107 waste to form the AP-107 melter feed, which was vitrified in the CLSM (Dixon et al. 2019). In a subsequent study, the offgas condensate produced from the AP-107 vitrification was concentrated in an evaporator and added to a new portion of AP-107 waste, after the waste went through solids removal by filtration (Geeting et al. 2019) and cesium removal by ion exchange (Fiskum et al. 2019), to mimic the recycle action of the EMF and LAW facility (Dixon et al. 2020b). This combined AP-107 recycle composition was entered into the Kim et al. (2012) model to determine the appropriate GFCs and a new glass formulation (termed AP-107-R1), which was vitrified in the CLSM (Dixon et al. 2020b).

In the current study, the condensate collected in the CLSM offgas treatment system during vitrification of the AP-107 recycle composition was concentrated in an evaporator test apparatus and combined with a new portion of AP-107 waste (from which solids and cesium had been removed, as in previous testing) to emulate a two-cycle recycle action. This two-time recycle waste composition was estimated based on the known compositions of the previous AP-107 samples and the condensate samples with the target concentrating ratio planned for evaporation. A two-time recycle AP-107 melter feed was then prepared using this waste composition estimate and the Kim et al. (2012) model to determine the appropriate GFC additions and glass formulation. A sample of AP-105 waste that had been processed through solids removal by filtration (Allred et al. 2020) and cesium removal by ion exchange (Fiskum et al. 2021), also had GFCs added, as calculated by the Kim et al. (2012) model, to form an AP-105 melter feed.

The purpose of the test described in this report was to assess the effect of recycle on dynamic melter processing and glass chemistry and evaluate the change of feed composition during continuous operation on those same factors. To do this, the CLSM system in the RPL was used to vitrify the two-time recycled AP-107 melter feed, immediately followed by the AP-105 melter feed, and generate product samples with processing results. Such results were then compared with previous AP-107 (with and without recycle) and AP-105 tests to contribute towards confirming the fraction of waste components assumed to partition into the offgas system and evaluate melter processing characteristics during the feed composition transition. Ultimately, the CLSM has demonstrated the ability to support future WTP programmatic needs regarding cold-cap behavior under different melter feed compositions and the effects of condensate recycle on component volatility into the offgas.

2.0 Quality Assurance

All research and development (R&D) work at PNNL is performed in accordance with PNNL's Laboratory-Level Quality Management Program, which is based on a graded application of NQA-1-2000, *Quality Assurance Requirements for Nuclear Facility Applications* (ASME 2000), to R&D activities. To ensure that all client quality assurance (QA) expectations were addressed, the QA controls of the PNNL's WRPS Waste Form Testing Program (WWFTP) QA program were also implemented for this work. The WWFTP QA program implements the requirements of NQA-1-2008, *Quality Assurance Requirements for Nuclear Facility Applications* (ASME 2008), and NQA-1a-2009, *Addenda to ASME NQA-1-2008* (ASME 2009). These are implemented through the *WWFTP Quality Assurance Plan* (QA-WWFTP-001) and associated QA-NSLW-numbered procedures that provide detailed instructions for implementing NQA-1 requirements for R&D work.

The work described in this report was assigned the technology level "Applied Research" and was planned, performed, documented, and reported in accordance with procedure QA-NSLW-1102, *Scientific Investigation for Applied Research*. All staff members contributing to the work received proper technical and QA training prior to performing quality-affecting work.

3.0 Experimental

This section describes the experimental process used to concentrate the AP-107 condensate from the previous CLSM run (Dixon et al. 2020b) and prepare the two-time recycle AP-107 and AP-105 melter feeds. The CLSM system in the RPL, which was used to vitrify the melter feeds, and the analyses of the resultant samples are also explained.

3.1 EMF Evaporator Testing

This section describes the evaporator feed preparation, test conditions, operation of the EMF evaporator test apparatus, and the resultant compositions of the evaporator feed and evaporator effluent (condensate and concentrate; the tops and bottoms, respectively) using offgas condensate from the CLSM system.

3.1.1 Evaporator Feed Preparation

Bench-scale EMF testing with approximately 7.5 L of AP-107 offgas condensate generated from the CLSM system (Dixon et al. 2020b) was conducted to determine component partitioning between the evaporator condensate and evaporator concentrate. During the CLSM run (Dixon et al. 2020b), the feed to the CLSM vessel was blended with evaporator concentrate from the previous AP-107 run (Dixon et al. 2019) so offgas produced during testing was a second recycle of this process. The AP-107 offgas condensate used as the evaporator feed was slightly yellow but transparent with rust-colored solids settled at the bottom. Per the WTP plan of operations (Bernards et al. 2017), all effluents coming to the EMF evaporator feed tank are filtered before being discharged into the feed tank along with the addition of 5 M caustic to maintain the feed tank pH above 10. Accordingly, solids in the AP-107 offgas condensate were filtered out using a 5-µm polyethersulfone (PES) filter paper and collected for the chemical analyses described in Section 3.4. The filtered AP-107 offgas condensate was pH adjusted from 1.0 to 12.8 using 580 mL of 5 M NaOH to finish its preparation as evaporator feed. The evaporator feed density was measured to be 1.086 g mL^{-1}.

3.1.2 Design Configuration and Test Conditions

A schematic of the EMF evaporator test apparatus used for this testing is shown in Figure 3.1. All testing was conducted inside a radiological contamination area (CA) fume hood in the RPL at PNNL. The apparatus was constructed almost entirely from glass, with the reaction vessel consisting of a cylindrical flat-bottom 1-L glass beaker. The evaporator feed in the vessel was heated using a fabric heating mantle, controlled with a Model 270 temperature controller (J-KEM Scientific, Inc., St. Louis, Missouri), and continuously stirred using a Teflon-coated magnetic stir bar on a stir plate. A pressure transducer was installed on top of the reaction vessel to record pressure during evaporation. Temperatures and pressures within the reaction vessel were recorded electronically at 1-minute intervals.

The EMF evaporator test apparatus operating conditions were prototypic to the full-scale EMF evaporator, which is designed to operate at 45 °C and 1.4 psia. As the evaporator feed boiled in the reaction vessel, the vapors travelled unrestricted through the glass condenser and drained into the glass condensate flask. The condenser was continuously operated with chilled water set to 8.5 °C. Any vapors that passed through the condenser and condensate flask were condensed in the gas washing bottle. The vacuum in the system was created by a Vacuubrand Chemistry vacuum pump, model MZ 2C NT. Figure 3.2 shows a photograph of the EMF evaporator test apparatus components as they were arranged in a CA fume hood.

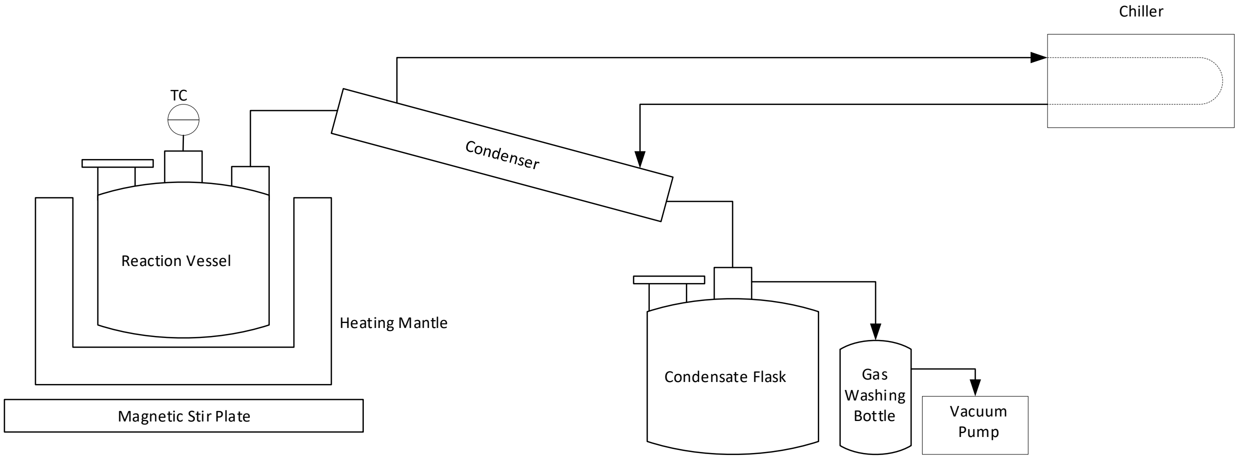

Figure 3.1. EMF evaporator test apparatus system schematic.

Figure 3.2. EMF evaporator test apparatus as arranged in a CA fume hood.

Initially, 1 L of evaporator feed was loaded into the reaction vessel. To achieve the desired concentration of 10.7 g g^{-1} (evaporator feed-to-concentrate ratio), and mimic a semi-continuous process, a volume balance of nominally 1 L of liquid was maintained between the reaction vessel and condensate flask throughout the experiment until all 8.7 kg of evaporator feed was evaporated down to an equivalent 707 g. At the end of testing, density for the evaporator condensate and evaporator concentrate were measured to be 1.01 and 1.38 g mL^{-1}, respectively. Solids were found in the evaporator concentrate but were not removed as had been done previously by Dixon et al. (2020b). Instead, the slurry was captured and collected for the chemical analyses described in Section 3.4.

3.1.3 Evaporator Operation and Contents Compositions

The EMF evaporator test apparatus was operated under vacuum at an average pressure around 1.4 psia with minor variance over the duration of the test, shown in Figure 3.3, leading to a standard deviation of 0.4 psia and a range from 0.8 to 2.2 psia. The evaporator feed boiled at an average temperature of approximately 42 °C during testing where, after initial heating, minor fluctuations in temperature readings ranged from 34.0 to 49.6 °C, observed in Figure 3.3, for a standard deviation of 5.0 °C. The boil-off rate of the evaporator condensate was approximately 2.62 mL min^{-1}.

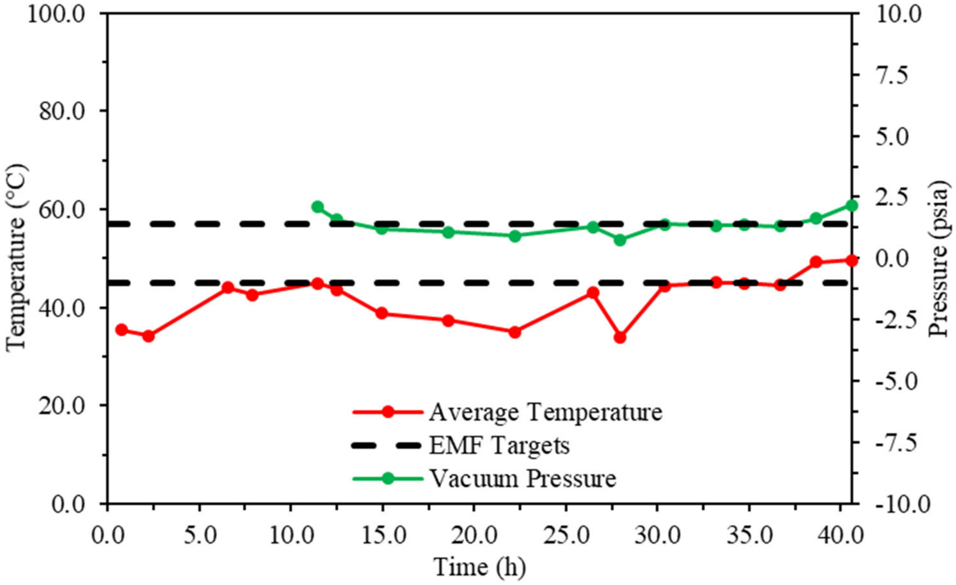

Figure 3.3. Temperature and pressure of the EMF evaporator test apparatus during operation.

The reaction vessel was typically insulated with glass wool that was periodically moved to allow for visual observation. The condensate would continue to boil during these brief observational periods and removing the insulation to observe the vessel contents did not interrupt the test.

The test apparatus was temporarily shut down to collect evaporator condensate from the condensate flask and replenish the reaction vessel with evaporator feed. The evaporator condensate was clear and colorless with a pH of 9. The evaporator concentrate after evaporation was dark brown and contained insoluble solids that had settled to the bottom of the reaction vessel. The concentrate slurry was pH tested to be 13. The final measured concentration factor, based on mass, was found to be 11.3 g g^{-1} (evaporator feed-to-concentrate ratio). A photo of a subsample from each processing solution is shown in Figure 3.4.

The solids in the AP-107 offgas condensate were filtered using a 5-µm PES filter paper and were analyzed; results are shown in Table 3.1. No ion chromatography (IC) anions were detected. Trace amounts (<0.01%) of Cs were present but the majority of constituents were made up of Si, Fe, Al, Zr, and Ti. The analytes found in the solid sample are consistent with those of glass formers and are likely present due to the vitrification process that typically causes trace amounts of melter feed to enter into the offgas treatment system. The percent of each component in the solids makeup was calculated as a ratio of the amount found for that component over the total mass of all components measured in the solid sample.

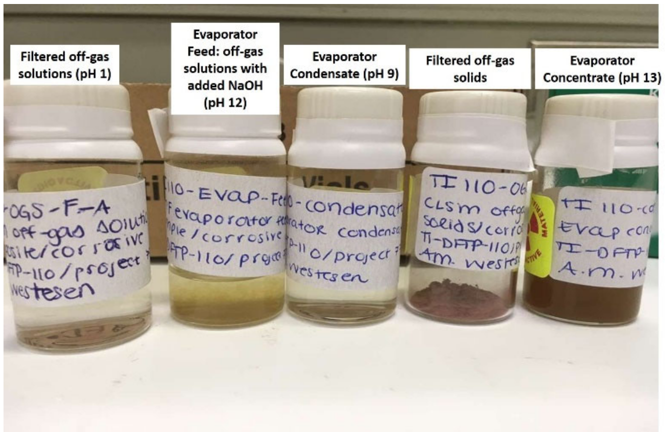

Figure 3.4. Labeling of evaporator solutions.

Table 3.1. Filtered Offgas Solids Composition

Analysis Method	Component	Filtered Offgas Solids (mg kg^{-1})	% in Solids (wt%)
Inductively coupled plasma mass spectrometry (ICP-MS)	Total Cs	45.4	0.01
	Total I	--	--
	^{99}Tc	--	--
Inductively coupled plasma atomic emission spectroscopy (ICPAES)	Al	49400	9.8
	Ba	16.0	--
	Cr	837	0.2
	Cu	12.8	--
	Fe	81900	16.3
	Li	49.8	--
	Mn	58.3	--
	Mo	146	--
	P	127	--
	K	298	0.1
	Si	318000	63.2
	Na	978	0.2
	Sr	3.06	--
	Sn	54.2	--
	Ti	10400	2.1
	W	774	0.2
	V	26.7	--
	Y	12.2	--
	Zn	4.40	0.1
	Zr	39400	7.8
Total			100.0

"--" = not applicable; analysis not performed, or component comprises < 0.1% of total

The compositions of the evaporator feed, condensate, concentrate, and collected concentrate solids were evaluated to understand component mass fractionations. Analysis results are shown in Table 3.2.

Component results shown with a "<" indicate that the value was below the analytical reporting limit (ARL); therefore, the fractionation result could not be calculated for these analytes.

Table 3.2. Evaporator Feed, Concentrate, and Condensate Compositions

Analysis Method	Component	Evaporator Feed (mg kg^{-1})	Concentrate (mg kg^{-1})	Condensate (mg kg^{-1})	Fraction in Concentrate (%)	Fraction in Condensate (%)	Total Recovered (%)
	Total inorganic carbon (TIC)	19.6	108	19.9	49	87	136
	Total organic carbon (TOC)	30.9	224	20.1	64	56	120
ICP-MS	Total Cs	0.734	8.25	<0.025	99	--	99
	Total I	0.793	8.99	0.0243	100	2.6	102
	^{99}Tc	2.20	24.4	0.00205	96	--	96
IC	Br$^-$	<9.84	19.9	<9.96	--	--	--
	Cl$^-$	456	5260	<9.96	102	--	102
	F$^-$	42.8	456	<9.96	94	--	94
	NO_3^-	4590	50500	<9.97	97	--	97
	NO_2^-	<9.84	45.3	<9.96	--	--	--
	PO_4^{3-}	<9.84	52.9	<9.96	--	--	--
	SO_4^{2-}	187	2110	<9.96	99	--	99
ICP-AES	Al	28.8	346	<5.02	106	--	106
	B	232	2480	28.3	94	10.5	105
	Ca	40.1	457	<2.51	100	--	100
	Cr	32.8	373	<0.251	100	--	100
	Co	<0.244	<0.195	<0.251	--	--	--
	Fe	53.9	632	<5.02	103	--	103
	Li	13.9	151	<0.753	96	--	96
	Mo	1.15	14.2	<0.502	109	--	109
	Ni	0.295	3.45	<0.251	103	--	103
	K	64.1	676	<7.53	93	--	93
	Si	141	1480	36.9	92	22.5	115
	Na	9160	103000	36.9	99	0.3	99
	S	68.6	749	<3.77	96	--	96
	Ti	3.28	38.4	<0.251	103	--	103
	Zn	65.0	759	<0.377	103	--	103
	Zr	4.39	51.3	<0.753	103	--	103

< values indicate the associated sample results were less than the analytical reporting limit (ARL).
"--" = not applicable; value not reported, or fractionation cannot be calculated with a less-than value.

Each component fractionation was calculated as the ratio of the total component measured in the evaporator feed to the total component collected in the evaporator effluent according to Eq. (3.1),

$$\frac{C_{Ea} \times M_E}{C_{Fa} \times M_F} = F_{Ea} \qquad (3.1)$$

where:

C_{Ea} = concentration of component *a* in the evaporator effluent (condensate, concentrate, concentrate solids)

M_E = mass of evaporator effluent (6897 g for condensate; 706.5 g for concentrate)
C_{Fa} = concentration of component *a* in the evaporator feed
M_F = mass of evaporator feed (8022 g)
F_{Ea} = fraction of component *a* in the evaporator effluent (condensate, concentrate, concentrate solids)

The concentrations in the evaporator concentrate showed a nominal 10× increase compared to the evaporator feed. Recovered fractions of at least 92% for Cs, I, and ^{99}Tc along with all detected anions were reported in the evaporator concentrate. As expected, no (or extremely low) concentrations of most components were measured in the evaporator condensate. The recoveries of TIC/TOC above 100% by nominally 25% is hypothesized to be contamination from vacuum grease. Three analytes were detected by ICP-AES in the condensate: B at 28.3 mg kg^{-1}, Si at 36.9 mg kg^{-1}, and Na at 36.9 mg kg^{-1}. Silicon detected is possibly due to leaching from the glassware and the contamination from vacuum grease, whereas B and Na are semi-volatile from the evaporator feed.

3.2 Melter Feed Preparation

The AP-107 waste received after solids and cesium removal had a density of 1.253 kg L^{-1} and totaled 6.786 kg. A total of 0.525 kg of evaporator concentrate, prepared as described in Section 3.1, was combined with the AP-107 waste for a volume ratio of 7.0 % evaporator concentrate to waste. This ratio was less than the target ratio of 8.5% because the evaporator concentrate was denser than estimated based on a previous concentrate value (Dixon et al. 2020b). By combining the AP-107 waste and this concentrated AP-107 offgas condensate solution, two rounds of the process of recycling the condensate from the EMF to the waste incoming to the LAW facility was replicated. The estimated component concentrations of this combined AP-107 waste plus concentrate solution, referred to as the two-time recycle AP-107 waste, were used in the Kim et al. (2012) glass models to calculate a glass composition to satisfy the WTP baseline requirements and the mass of GFC additions needed to achieve this composition.

The AP-105 waste received after solids and cesium removal had a density of 1.285 kg L^{-1} and totaled 7.504 kg. The component concentrations of the AP-105 waste (Fiskum et al. 2021) were used in the Kim et al. (2012) glass models to calculate a glass composition to satisfy the WTP baseline requirements and the mass of GFC additions needed to achieve this composition. This process of analyzing the combined waste stream and using a model to determine the GFC additions is the same process currently planned for melter feed preparation at the WTP (Bernards et al. 2017).

The GFC minerals and the mass of each addition for the two-time recycle AP-107 waste and AP-105 waste are given in Table 3.3. These GFCs were added to their respective waste solutions to form the 'two-time recycle AP-107 melter feed' and 'AP-105 melter feed', which had target glass yields of 685.8 g-glass L-feed^{-1} and 699.8 g-glass L-feed^{-1}, respectively. The composition of the glass to be produced from the two-time recycle AP-107 melter feed, referred to as AP-107-2R, is shown in Table 3.4 along with the compositions of the AP-107 one-time recycle glass from Dixon et al. (2020b), referred to as AP-107-1R, the AP-107 glass from Dixon et al. (2019), referred to as AP-107, and the AP-107 glass from Matlack et al. (2018), referred to as AP107WDFL. The composition of the glass to be produced from the AP-105 melter feed, referred to as AP-105, is also shown in Table 3.4 along with the composition of the AP-105 glass from Matlack et al. (2017), referred to as WDFL1.

Table 3.3. GFCs Masses Added to Each Portion of Waste

GFCs	Mass Added to Two-Time Recycle AP-107 Waste (g)	Mass Added to AP-105 Waste (g)	Mineral Source
Kyanite	401.36	310.72	Kyanite Mining Corporation
Boric Acid	965.72	991.43	Alfa Aesar
Wollastonite	486.89	319.65	NYCO Mineral
Iron Oxide	286.23	292.96	JT Baker
Lithium Carbonate	143.39	--	Foote Mineral Company
Olivine	160.91	166.84	Unimin Corporation
Silica	1938.06	2116.64	Sil-Co-Sil 75
Rutile	76.22	79.18	Chemalloy
Zinc Oxide	192.33	196.34	Noah
Zircon	244.95	249.84	Prince Minerals
Sucrose	349.44	372.73	C+H Sugar
Total	5245.50	5096.33	

Table 3.4. Target Glass Compositions for AP-107 and AP-105 Wastes

Component	**AP-107-2R (wt%)**	AP-107-1R[(a)] (wt%)	AP-107[(b)] (wt%)	AP107WDFL[(c)] (wt%)	**AP-105 (wt%)**	WDFL1[(d)] (wt%)
Al_2O_3	**6.13**	6.13	6.12	6.10	**6.13**	6.10
B_2O_3	**9.95**	9.95	9.95	10.00	**9.95**	10.00
CaO	**4.09**	4.53	3.69	3.94	**2.64**	2.08
Cl	**0.19**	0.18	0.15	0.42	**0.22**	0.45
Cr_2O_3	**0.08**	0.07	0.08	0.08	**0.06**	0.05
F	**0.03**	0.03	0.00	0.04	**0.00**	0.01
Fe_2O_3	**5.52**	5.52	5.52	5.50	**5.52**	5.50
K_2O	**0.49**	0.36	0.47	0.38	**0.49**	0.41
Li_2O	**1.06**	1.52	0.50	0.89	**0.00**	--.--
MgO	**1.49**	1.49	1.49	1.48	**1.49**	1.48
Na_2O	**16.89**	16.34	17.49	17.20	**19.35**	21.00
NiO	**0.01**	0.01	0.01	0.00	**0.02**	0.00
P_2O_5	**0.16**	0.19	0.15	0.13	**0.11**	0.17
SO_3	**0.38**	0.39	0.37	0.44	**0.33**	0.30
SiO_2	**45.60**	45.36	46.08	45.50	**45.76**	44.54
TiO_2	**1.40**	1.40	1.40	1.40	**1.40**	1.40
ZnO	**3.51**	3.51	3.51	3.50	**3.51**	3.50
ZrO_2	**3.02**	3.02	3.01	3.00	**3.02**	3.00
Total	**100.0**	100.0	100.0	100.0	**100.0**	100.0

Information for the glasses associated with the testing described in this report is shown in **boldface** type.

(a) Dixon et al. (2020b)

(b) Dixon et al. (2019)

(c) Matlack et al. (2018)

(d) Matlack et al. (2017)

3.3 CLSM System

This section describes the CLSM system as assembled in a high contamination area (HCA) fume hood in the RPL at PNNL with supporting equipment located in an adjacent CA fume hood and the surrounding areas, as well as the details of the operating conditions for system performance.

3.3.1 System Design and Configuration

The CLSM system was designed to collect samples of glass, offgas solids, and offgas condensate without upsetting continuous operation. The CLSM was not designed to be fully prototypic of the WTP LAW melters, but to reproduce the feed-to-glass conversion process performed in the melters. A simplified flow diagram of the CLSM system is shown in Figure 3.5.

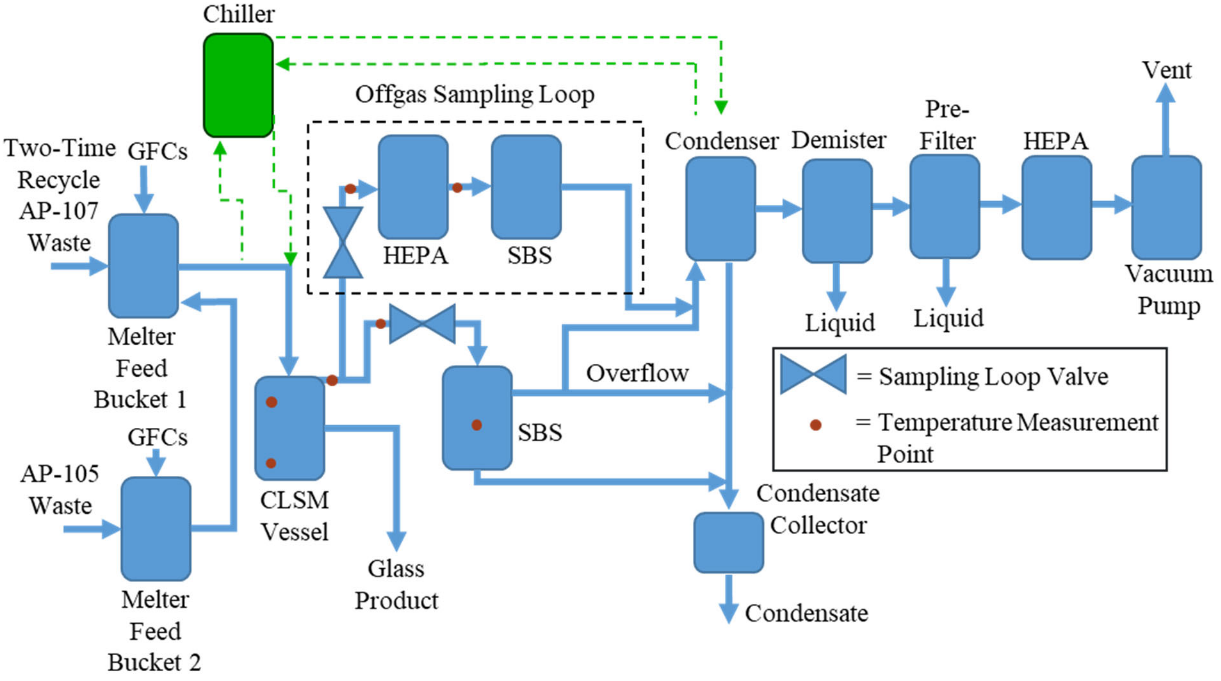

Figure 3.5. Simplified flow diagram of the CLSM system.

The two-time recycle AP-107 melter feed was placed in the 'Melter Feed Bucket 1' position and the AP-105 melter feed was placed in the 'Melter Feed Bucket 2' position. Each bucket was agitated by an overhead stirrer for at least 24 hours prior to processing in the CLSM system and remained continuously agitated during testing. A peristaltic pump was used to move the melter feed from Bucket 2, in the CA fume hood, into Bucket 1, in the HCA hood, when desired. The melter feed was pumped from Bucket 1 to the CLSM vessel by a progressive cavity pump through quarter-inch, stainless-steel tubing, which could produce a continuous drip of melter feed at a steady rate. The stainless-steel feed tubing that entered the CLSM vessel was water-cooled to prevent evaporation of the melter feed in the tubing that could result in feed line blockage.

The CLSM vessel was fabricated as an octagonal cross-sectional design using Inconel 690 plate and sized to an equivalent cylindrical diameter of approximately 12.0 cm (4.7 inches), resulting in a cross-section and glass surface area of 0.0113 m^2 and plenum volume of 0.0018 m^3. A newly fabricated CLSM vessel was used for this study. The glass inventory in the CLSM vessel was approximately 2.0 kg, resulting in a glass melt pool depth of ~6.4 cm (2.5 inches).

As seen in Figure 3.6, the lid of the CLSM vessel contained eight access ports: three for thermocouples ("TC" in the figure), one for an air bubbler, one for the feed tubing, one for a sight glass into the vessel ("Viewport" in the figure), one for the connection to the offgas system, and one for pressure relief ("Back-Up Offgas" in the figure). Heat was supplied externally to the CLSM vessel by a surrounding furnace. The hot zone of the furnace was located below and around the glass melt pool while the offgas head space, called the plenum, of the CLSM vessel was surrounded by insulation. The CLSM achieved continuous operation by periodically pouring glass out of the melt pool to a glass discharge box located below the CLSM vessel. Pouring was achieved by lowering the vacuum maintained on the CLSM vessel by the offgas system, which allowed glass to pour by rising through a discharge riser and passing over an overflow weir.

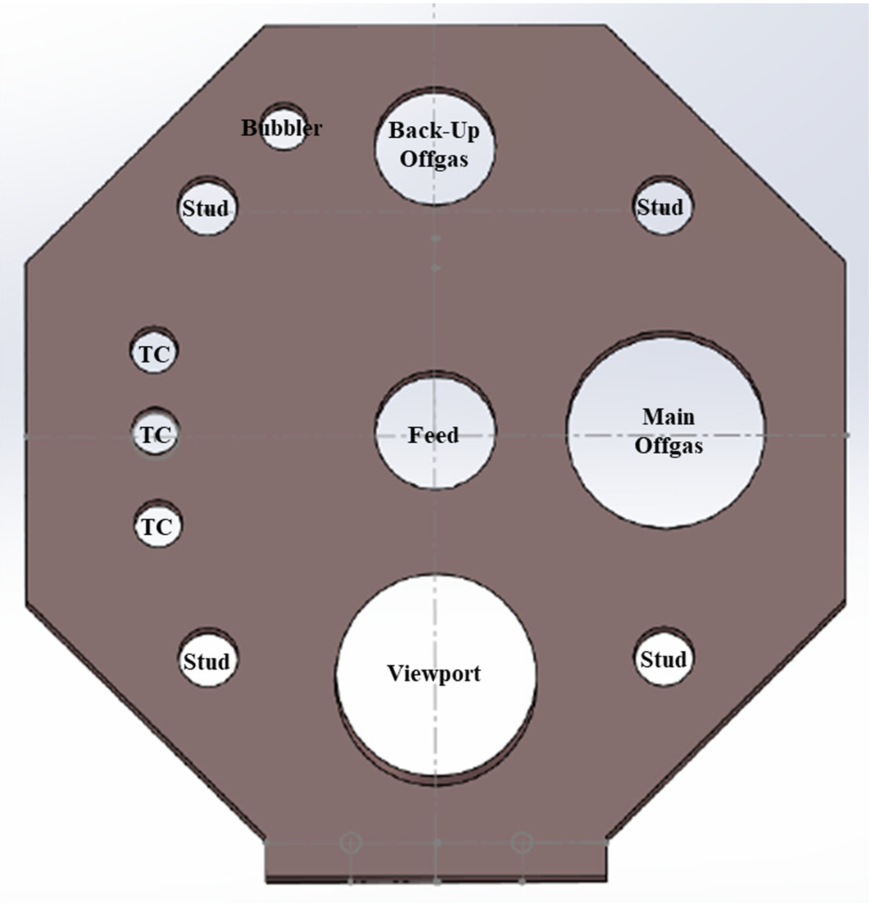

Figure 3.6. CLSM vessel lid and identified ports. The designation ‘TC’ stands for a thermocouple port.

The offgas produced by the conversion of melter feed to molten glass was drawn off from a port in the CLSM vessel lid into the offgas system with a vacuum pump. The offgas system was constructed of stainless-steel piping and the units described subsequently. Except when the offgas stream was sampled, the offgas would flow through the primary pathway in the offgas system, which consisted of a submerged-bed scrubber (SBS; referred to as the primary SBS), a condenser, a demister, a polypropylene pre-filter, and a high-efficiency particulate air (HEPA) filter (referred to as the primary HEPA filter). The primary SBS and the condenser worked together to both cool the offgas, causing condensation of steam, and perform scrubbing to remove other soluble gases and aerosols as much as possible. The cool liquid from the condenser along with the liquid overflow from the primary SBS drained into a collector where this condensate liquid could be drained periodically. Offgas from the condenser passed through a demister, that allowed any remaining liquid to accumulate before the pre-filter and primary HEPA filter captured any remaining difficult-to-remove particulates. After HEPA filtration, the offgas flowed through the vacuum pump and was released to the HCA fume hood ventilation system. When needed, the pre-filter and primary HEPA filter could be bypassed and the offgas would flow directly from the demister to the vacuum pump.

The total offgas stream could be sampled by closing the sampling valve in the primary offgas pathway to divert the full offgas flow through a sampling loop containing heated HEPA filters (referred to as the sampling HEPA filters) followed by an SBS (referred to as the sampling SBS). This sampling train consisted of three parallel housings, each with a sampling HEPA filter. Each housing was available for a discrete sampling evolution. The sampled offgas stream was then released back into the primary offgas pathway before the condenser unit. Sampling of the total offgas stream avoided the inherent issues with offgas piping geometry and design that are encountered with slip-stream sampling and ensured that the sample was representative. Offgas sampling durations were typically 10-30 minutes or until the sampling HEPA filters became impassable.

The CLSM system consisted of commercially available as well as custom-built parts. In addition to the CLSM system described above (shown in Figure 3.5), supporting equipment included a controller for the furnace; a water chiller pumping system to cool all of the necessary locations in the CLSM system, such as the condenser and the primary SBS, with a separate liquid pump plumbed into the chiller line to transport cooling water to the feed nozzle at a controlled rate; a water flush pump for washing out the melter feed pumping system; a controller for the heat trace around the sampling and primary HEPA filters; and a computer for controlling the CLSM system while continuously recording process data. An image of the CLSM system layout in the fume hoods in RPL is shown in Figure 3.7.

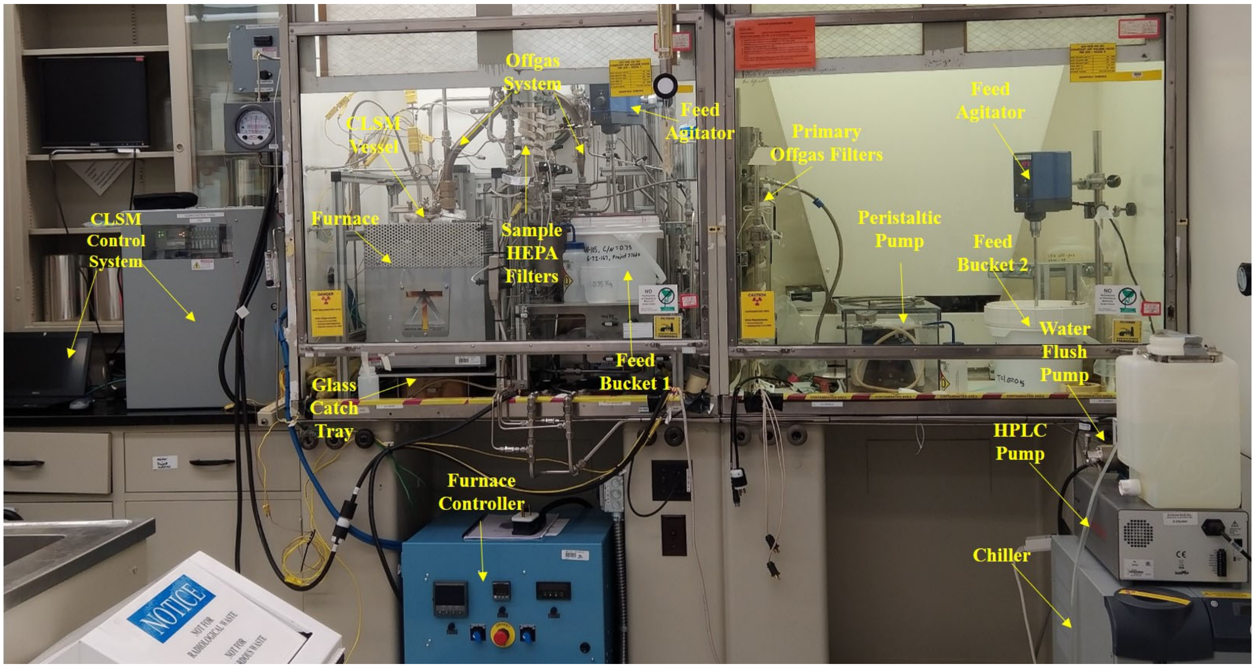

Figure 3.7. CLSM system layout in the RPL HCA (left) and CA (right) fume hoods.

3.3.2 Test Conditions

The CLSM was operated to maintain a glass melt pool temperature of 1150 °C (± 30 °C) by manually adjusting the control temperature of the surrounding furnace as necessary. During feeding operations, the melter feed was charged onto the glass melt surface in the CLSM vessel, forming a batch blanket, called a cold cap, where the feed was heated and converted to glass (Dixon et al. 2015). The feeding rate (governed by the progressive cavity pump with an operational range from 0-36 revolutions per minute) and air bubbling rate (governed by a mass flow controller that could deliver air at 50-3000 standard cubic centimeters per minute [sccm] through a high-temperature 600 nickel alloy tube that was submerged in the glass melt pool) were varied to maintain a target cold-cap coverage over the glass melt surface of 75-95%. The cold-cap coverage was determined to be in the appropriate range when the temperature in the plenum fell into the 500-700 °C range and this could be confirmed through visual observation (by visually estimating the cold-cap coverage) in the viewport of the CLSM vessel lid. The CLSM briefly did produce glass melt pool and plenum temperatures above and below the target ranges.

Typical of slurry-fed melters, the plenum temperature and cold-cap coverage were influenced by many factors, including feed composition and component concentrations, which may vary between different melter feeds (Matlack et al. 2011). The target production rate ranges for the first feed to be processed in the CLSM, the two-time recycle AP-107 melter feed, were derived from the previous processing of AP-107 melter feed in the CLSM (Dixon et al. 2019) and the processing of AP-107 melter feed in the DuraMelter10 (DM10) melter system operated by the Vitreous State Laboratory of The Catholic University of America (Matlack et al. 2018), and are listed in Table 3.5. These values align with the designed operation rates at the WTP of 15 metric tons of glass per day [MTG d^{-1}] of immobilized LAW (Bernards et al. 2017).

Table 3.5. Target CLSM Operating Conditions

Parameter	CLSM Target Range	DM10 Processing[(a)]
Target glass production rate, kg m^{-2} d^{-1}	1500 – 2000	1974
Melt surface area, m^2	0.0113	0.021
Target feeding rate, kg-feed h^{-1}	1.59 – 2.12	3.98
Target feeding rate, L-feed h^{-1}	1.01 – 1.35	2.65
Bubbling rate, sccm	50 – 2000	1400
Target glass melt temperature, °C	1150 ± 30	1150
Plenum temperature range, °C	500 – 700	580
Plenum vacuum normal operation, in-H_2O	2 – 4	--
Offgas piping temperature range, °C	< 500	--
Primary SBS temperature, °C	15 – 35	--

[(a)] Matlack et al. (2018)

Values marked with '--' were not comparable due to differences in the offgas systems.

The condenser in the offgas system was operated with chilled water and the condensate drained periodically from a collector vessel. The liquid level in the primary SBS was maintained by overflow so that the pressure-drop across the primary SBS remained relatively constant; the temperature was maintained by circulating chilled water through cooling coils in the primary SBS. In the offgas sampling loop, the sampling HEPA filters were wrapped with heat trace and covered with insulation to maintain an elevated temperature (>100 °C) and prevent/reduce condensation prior to the sampling SBS. The offgas system vacuum pump was operated such that it pulled a vacuum on the CLSM vessel during feeding operation. The nominal operating vacuum was 2–4 in-H_2O. As described in Section 3.3.1, the CLSM vessel vacuum was reduced periodically to pour glass. At the end of the run, the bubbler air and viewport purge air were adjusted to increase the pressure in the melter, pouring controlled volumes of glass from the CLSM vessel until the remainder of the glass inventory had exited the vessel.

3.4 Sample Analysis Methods

The mass of each component in the formation of melter feeds was totaled to determine the initial mass in each bucket. The masses of all product streams were weighed after the run; these included the glass from each pour, the total condensate, the final sump contents from both the sampling SBS and primary SBS (the SBS sumps contained only the liquid from the final capacity of each SBS since, during operation, the SBS liquid would overflow into the condensate collector), the liquid in the demister, the liquid that had accumulated in the pre-filter housing, the pre-filter, the primary HEPA filters, and the sampling HEPA filters. Approximately 10-mL or 10-g samples (for liquid or solid streams, respectively) were taken of the melter feed and from selected product streams. Appropriate product streams were selected by the operational team to gain insight about the operational behavior of each CLSM run. These selected samples, and whole primary/sampling HEPA filters, were sent to the Southwest Research Institute (SwRI) for cation and anion chemical analysis. The analysis methods employed by SwRI and each component measured using each method are given in Table 3.6.

Table 3.6. Sample Chemical Analysis Methods and Components Scanned

Analysis	Methods	Component
Cations	ICP-AES or ICP-MS for Cs, I, and ^{99}Tc	Ag, Al, As, B, Ba, Bi, Ca, Cd, Co, Cr, Cs, Cu, Fe, I, La, Li, K, Mg, Mn, Mo, Na, Ni, P, Pb, S, Si, Sr, Sn, ^{99}Tc, Ti, V, W, Y, Zn, and Zr
Anions	IC or Ion-Specific Electrode	Chloride, Chromate, Fluoride, Nitrate, Nitrite, Phosphate, and Sulfate
Radionuclides	Alpha Spectroscopy	^{241}Am, ^{242}Cm, $^{243/244}$Cm, ^{237}Np, ^{238}Pu, $^{239/240}$Pu, and ^{244}Pu
TOC	TOC Analyzer	Total organic carbon
TIC	TIC Analyzer	Total inorganic carbon

4.0 Results

This section describes the operation of the CLSM in RPL on May 25, 2021 for the processing of the two-time recycle AP-107 melter feed and AP-105 melter feed. The production and chemical analysis results are also detailed.

4.1 Operational Description

During set-up of the CLSM system, approximately 2.0 kg of previously prepared AP-107 glass (without Cs, I, or ^{99}Tc spikes) were loaded into the CLSM vessel as the initial glass inventory. The furnace surrounding the CLSM vessel was heated from room temperature to 1250 °C at 10 °C min^{-1}. The CLSM run then began by charging the two-time recycle AP-107 melter feed at 10:15 AM, when the glass temperature had reached its desired range. The time (using the start of feeding as 0.00 h), mass, and cumulative mass of each glass pour during the run are given in Table 4.1. Following the termination of feeding, the cold cap burned off (all remaining melter feed in the cold cap and plenum walls was converted into glass) and the glass inventory was poured out of the CLSM vessel, corresponding with the final glass pour reported for the run. Given the total mass of glass poured and the initial inventory of glass in the CLSM vessel, the mass of glass produced during the CLSM run was 9.70 kg, corresponding to nearly 5 turnovers of the 2.0 kg glass inventory. Other notes about the performance during the run follow.

- Around hour 4.00, the cold cap was observed to “roll over,” or suddenly become submerged, exposing fresh molten glass to the plenum space. This resulted in an increase in the plenum temperature until the cold cap structure was stable again after about 15 minutes. This temperature excursion is shown in Figure 4.1.

- From hour 7.47 to 8.15, the cold cap was able to burn off and the CLSM system remained in idling conditions because a plug in the feed transfer line between melter feed bucket #1 and melter feed bucket #2 (see Figure 3.5) necessitated a pause to clear the line.

- The cold-cap behavior for the two-time recycle AP-107 melter feed (thin and quick to react to operator input) seemed consistent with previous AP-107 runs (Dixon et al. 2019 and 2020b), and after hour 8.15, the cold-cap behavior changed quickly to the behavior typical of AP-105 feeds characterized as having persistent foam around the edges and being sluggish to respond to operator input (Dixon et al. 2018).

Table 4.1. Timing and Mass of Glass Pours During the CLSM Run of the Two-Time Recycle AP-107 and AP-105 Melter Feeds

Pour Time (h)	Glass Mass (g)	Cumulative Glass Mass (kg)	Melter Feed Charged
0.17	97.24	0.10	Two-Time Recycle AP-107
0.80	227.21	0.32	
1.27	262.99	0.59	
1.78	310.86	0.90	
2.27	299.45	1.20	
2.77	559.24	1.76	
3.23	202.74	1.96	
3.95	217.90	2.18	
4.63	501.61	2.68	
4.83	387.59	3.07	
5.57	442.78	3.51	
6.05	343.05	3.85	
6.53	292.37	4.15	
7.13	241.82	4.39	
8.75	594.14	4.98	AP-105
9.25	465.32	5.45	
9.75	437.36	5.88	
10.23	360.07	6.24	
10.73	370.81	6.61	
11.25	335.35	6.95	
11.75	316.65	7.27	
12.25	452.55	7.72	
12.80	281.43	8.00	
13.25	288.28	8.29	
13.75	303.34	8.59	
14.27	346.27	8.94	
14.31	2736.59	11.68	

4.2 Production Results

The production results from the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds are given in Table 4.2 for the total run and for times of processing each melter feed. The production results include the total values of the feeding time (and low flow duration within the feeding time), operational downtime, measured mass of glass produced, calculated mass of melter feed consumed, and average values for the glass production rate, feeding rate, glass temperature, and plenum temperature. The mass of melter feed consumed was calculated based on the amount of glass produced and the target glass yield of each melter feed, given in Section 3.2. The data logger that recorded processing information during the CLSM run failed during the middle portion of the run, thus making the average bubbling flux rate (bubbling rate in L min^{-1}, scaled by the glass surface area of the CLSM vessel) not able to be calculated.

The processing values recorded during the CLSM run are displayed in Figure 4.1. These results include the glass and plenum temperatures, the average glass production rate during the portion of the run with each melter feed, the bubbling flux rate, and the melter vessel vacuum measurements. The glass and plenum temperatures were monitored by thermocouples with dual reading capabilities, one recorded by the CLSM data acquisition system and the other by a calibrated handheld device, both of which are reported in Figure 4.1. Three offgas samples were collected during the CLSM run, one during steady processing of the two-time recycle AP-107 melter feed, one during transition to the AP-105 melter feed, and one during steady processing of the AP-105 melter feed. The occurrence of each offgas sample in the timeline of the run is shown in relation to the processing values in Figure 4.1. Fewer data were recorded during the middle of the run due to the failure of the data logger.

Table 4.2. CLSM Production Results During the AP-107 Recycle CLSM Run

Parameter	Total CLSM Run	Two-Time Recycle AP-107	AP-105
Test Date	May 25, 2021	May 25, 2021	May 25, 2021
Feeding Duration, h	13.63	7.47	6.16
Low Flow Duration, h	--	--	--
Downtime, h	0.68	--	--
Glass Produced, kg	9.70	5.15	4.55
Melter Feed Consumed (Calculated), kg	22.13	11.84	10.29
Average Glass Production Rate, kg m^{-2} d^{-1}	1511	1464	1568
Average Feeding Rate, kg h^{-1}	1.62	1.59	1.67
Average Bubbling Flux Rate(a), L m^{-2} min^{-1}	--	--	--
Average Glass Temperature, °C	1158	1157	1159
Average Plenum Temperature, °C	705	733	670

(a) Values not reported due to data loss.

Values marked with '--' did not occur or could not be reported.

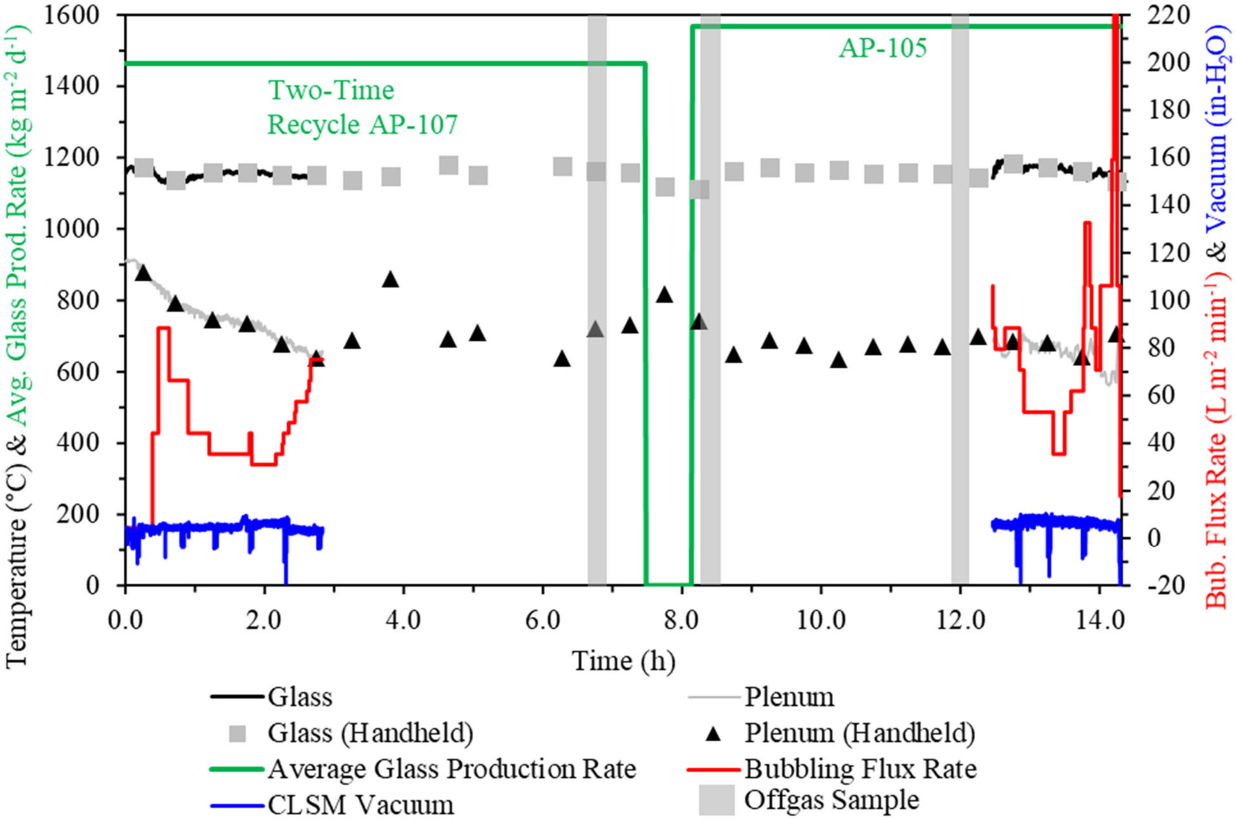

Figure 4.1. Processing values (glass and plenum temperatures, effective glass production rate, bubbling flux rate, and melter vacuum measurements) and offgas sample timing recorded during the CLSM run with two-time recycle AP-107 and AP-105 melter feeds. Gaps in the plots of bubbling flux rate and CLSM vacuum during the middle of the run are due to failure of the data logger.

4.3 Sample Chemical Analysis

The samples selected for chemical analysis from the CLSM run are listed in Table A.1 in Appendix A along with the total mass of each sample stream and the concentration of each analyzed cation, anion, and radionuclide. The samples of condensate were combined into three portions: 1) all condensate produced while feeding the two-time recycle AP-107 melter feed from the start of the run to hour 7.47; 2) all condensate produced during the downtime from hour 7.47 to 8.15; and 3) all condensate produced while feeding the AP-105 melter feed from hour 8.15 to the end of melter feed charging. Similarly, the liquid that accumulated in the demister was combined into two portions: 1) all liquid produced while feeding the two-time recycle AP-107 melter feed from the start of the run to hour 7.47; and 2) all liquid produced while feeding the AP-105 melter feed from hour 8.15 to the end of melter feed charging. Liquid accumulated in the pre-filter housing during the run, so it was drained from the housing and collected. The sump from the primary SBS was drained after the run and collected. Aliquots of all the liquid portions described were sent for chemical analysis. For the collection of each offgas sample, the complete sampling HEPA filters were digested and analyzed together. Due to its size, the pre-filter had to be split into 3 portions for shipment, but all 3 portions were digested and combined for analysis.

5.0 Discussion

This section discusses the insights gained from the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds and compares data to the previous AP-107 CLSM runs (Dixon et al. 2019 and 2020b) and AP-105 run (Dixon et al. 2018) in the RPL at PNNL.

5.1 Component Decontamination Factor, Retention, and Recovery

The decontamination factor (DF) of any component through any unit in a melter system is described as the mass flow rate of the component into the unit divided by the mass flow rate of the component out of the unit in the secondary product stream. In the CLSM system, there is one incoming mass flow stream, the melter feed, and there are two output mass flow streams, the glass (primary product stream) produced from the CLSM and the offgas (secondary product stream) exiting the CLSM. The CLSM offgas is comprised of gaseous mass exiting the system, vapor which is condensed by the offgas system as condensate, and solids that settle or are filtered. Thus, the DF of any component through the CLSM vessel is defined as the mass flow rate of that component in the melter feed divided by the mass flow rate of that component in the offgas stream. Given a state of no component accumulation in the CLSM vessel, the mass flow rate in the offgas is equal to the mass flow rate in the melter feed minus the mass flow rate in the glass, meaning the DF for a component in the CLSM vessel can be given by Eq. (5.1):

$$\mathrm{DF}_i = \frac{\dot{m}_{i,feed}}{\dot{m}_{i,feed} - \dot{m}_{i,glass}} \tag{5.1}$$

where $\dot{m}_{i,feed}$ is the mass flow rate [mg min^{-1}] of a component (i) in the melter feed and $\dot{m}_{i,glass}$ is the mass flow rate [mg min^{-1}] of the same component in the glass product.

The retention (R) of any component in the glass produced from the CLSM vessel is then defined as the mass flow rate of that component in the glass product divided by the mass flow rate of the same component in the melter feed and this value can be determined by Eq. (5.2):

$$R_i = \frac{\dot{m}_{i,glass}}{\dot{m}_{i,feed}} \tag{5.2}$$

The R_i value can be reported as a fraction or percentage (if Eq. (5.2) is multiplied by 100).

Finally, the recovery (Rec) of any component in the CLSM system is defined as the mass flow rate of the component out of the system in the summation of the glass and offgas divided by the mass flow rate of the same component into the system via the melter feed. The Rec_i value can be reported as a fraction or percentage (if multiplied by 100) and is defined in Eq. (5.3):

$$\mathrm{Rec}_i = \frac{\dot{m}_{i,glass} + \dot{m}_{i,offgas}}{\dot{m}_{i,feed}} \tag{5.3}$$

where $\dot{m}_{i,offgas}$ is the mass flow rate [mg min^{-1}] of a component (i) in the offgas as recovered by the units in the CLSM offgas system. For Eq. (5.1), Eq. (5.2), and Eq. (5.3), if the values are calculated for a fixed amount of time (e.g., the offgas sampling times or the total runtime) mass flow rates become total mass values (m_i; [mg]).

The components of primary interest in the CLSM glass product, in addition to the components in the target glass compositions, are ^{99}Tc, Cs, and I. Given the demonstrated volatility behavior of meta-stable technetium, ^{99m}Tc, from an idling glass melt (Matlack et al. 2010; Pegg 2015) and the potential unsteady incorporation of components into the glass melt while the cold cap varies from its target coverage and thickness, the R_{99Tc}, R_{Cs}, and R_I values were calculated both during the total runtime and during the offgas sampling timeframes when the cold-cap characteristics were believed to be steady.

5.2 Glass Composition

This section discusses the glass product from the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds and breaks down the analysis into the different types of components in the glass composition.

5.2.1 Primary Glass Components

Table 5.1 compares the compositions of the glass produced during the CLSM run with the respective target compositions. Since the initial glass in the CLSM vessel prior to processing was akin to AP-107-2R, the measured composition of glasses produced while processing the two-time recycle AP-107 melter feed (Glass Pours 0.17 through 7.13 in Table A.1 of Appendix A) was averaged (by converting the analyzed component concentrations in each glass sample listed to their associated oxides and averaging based on the mass of glass poured with each composition) and compared with the target AP-107-2R glass composition. For each primary glass component (present in >1.00 wt% amounts), the percent differences between the measured composition and the target composition are reported in Table 5.1.

Table 5.1. Comparison of CLSM Run AP-107-2R and AP-105
Glass Products with Target Compositions

Component	Target AP-107-2R	Average Measured AP-107-2R	% Diff. Target to Measured AP-107-2R	Target AP-105	Final Measured AP-105	% Diff. Target to Measured AP-105
	wt%	wt%	%	wt%	wt%	%
Al_2O_3	6.13	6.00	-2.1	6.13	5.99	-2.3
B_2O_3	9.95	9.62	-3.3	9.95	9.69	-2.6
CaO	4.09	4.08	-0.2	2.64	2.99	13.4
Cl	0.19	0.27	--	0.22	0.35	--
Cr_2O_3	0.08	0.11	--	0.06	0.09	--
F	0.03	0.05	--	0.00	0.03	--
Fe_2O_3	5.52	5.36	-2.9	5.52	5.45	-1.2
K_2O	0.49	0.40	--	0.49	0.45	--
Li_2O	1.06	1.15	8.9	0.00	0.32	--
MgO	1.49	1.46	-2.1	1.49	1.45	-2.6
Na_2O	16.89	17.85	5.6	19.35	18.75	-3.1
NiO	0.01	0.02	--	0.02	0.02	--
P_2O_5	0.16	0.09	--	0.11	0.06	--
SO_3	0.38	0.42	--	0.33	0.32	--
SiO_2	45.60	45.67	0.2	45.76	46.54	1.7
TiO_2	1.40	1.38	-1.9	1.40	1.36	-3.0
ZnO	3.51	3.27	-6.8	3.51	3.38	-3.9
ZrO_2	3.02	2.81	-6.8	3.02	2.75	-8.8
Total	100.0	100.0		100.0	100.0	

Values marked with '--' were not calculated because the component target concentrations were <1%.

When the AP-105 melter feed began processing, the glass composition was estimated to take two turnovers of the CLSM vessel inventory before arriving at a chemical steady state expected of the different melter feed composition (Dixon et al. 2020a). Thus, only the final measured glass pour composition (Glass Pour 14.31 in Table A.1 of Appendix A) was compared with the target AP-105 glass composition and the percent differences displayed in Table 5.1.

Compositional trends for each primary component measured in the glass product from the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds are displayed in Figure 5.1 and Figure 5.2, with respect to the amount of glass discharged. Each graph shows the measured component content in the glass as black squares (■), the anticipated component content in the glass based on the target glass composition as a black line (━), and the expected component content in the glass based on the analyzed melter feed samples as a red line (—). Reasons for slight differences between the target glass compositions and the compositions expected based on the melter feed sample analysis will be discussed in Section 5.3.

For the primary components that were similar in content for both the AP-107-2R and AP-105 glass compositions, the measured values were consistently within ±10 % of their targets as has previously been shown for a variety of glass compositions produced in the CLSM (Dixon et al. 2020a and 2020b). The most frequent reason for a difference between the measured and target values was that the melter feed was slightly deficient (for Al_2O_3, Fe_2O_3, ZnO, and ZrO_2) or abundant (for SiO_2 and Na_2O) compared to the target. Two primary components, CaO and Li_2O, were greater in the AP-107-2R target composition than the AP-105 target composition, see Table 5.1. The compositional trends of CaO and Li_2O in Figure 5.2 revealed that turning over the glass inventory in the CLSM vessel twice, by discharging ~4 kg of glass, while processing a melter feed with a different composition brought the glass composition near the changed target desired from the new melter feed composition. This glass-equilibration-in-two-turnovers characteristic was observed previously for minor impurity spikes in the initial glass in the CLSM vessel (Dixon et al. 2020a and 2020b), so the CaO and Li_2O behavior in this CLSM run begins to affirm that characteristic for components present in greater quantities in the glass.

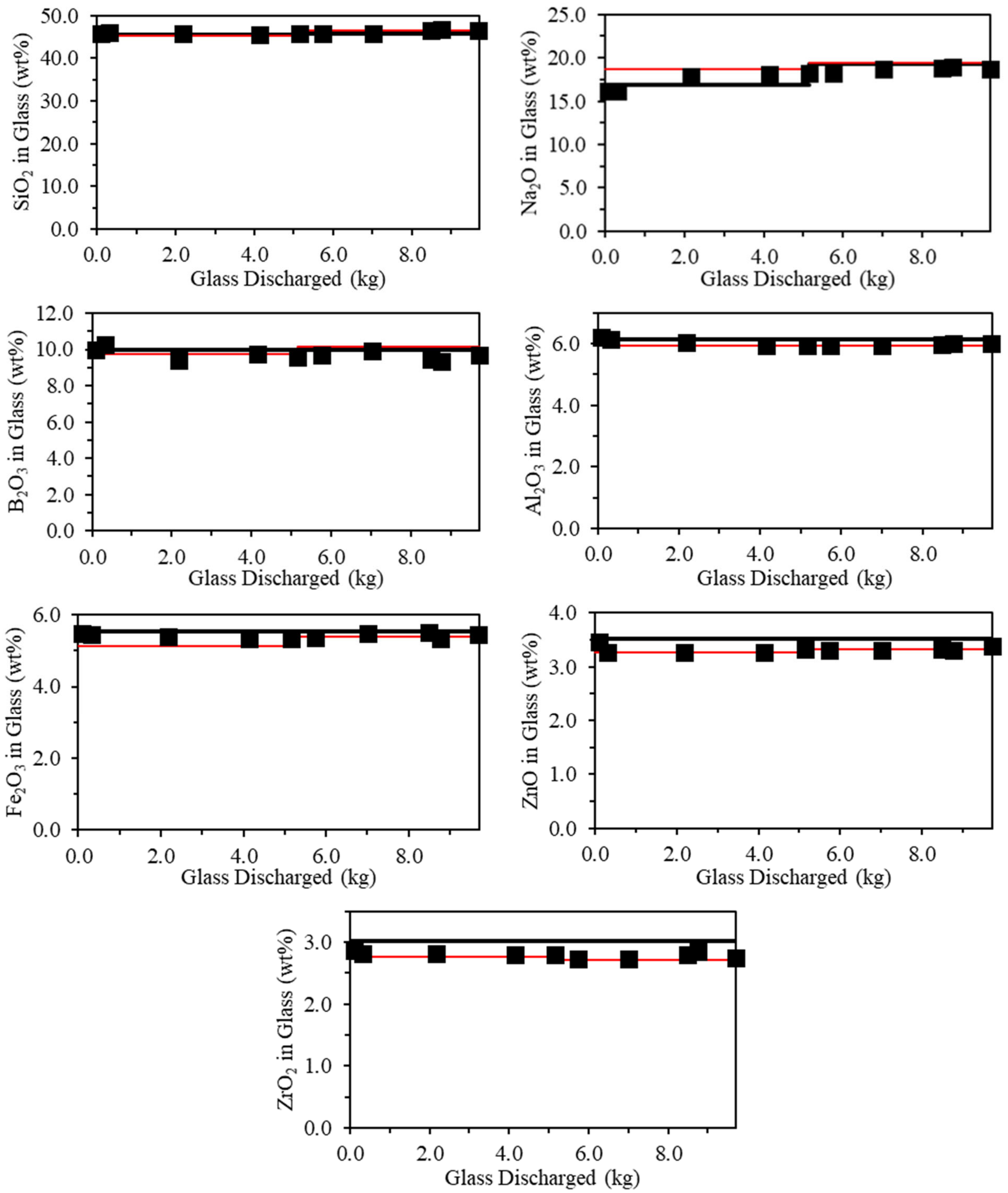

Figure 5.1. Content of a set of primary glass components (SiO_2, Na_2O, B_2O_3, Al_2O_3, Fe_2O_3, ZnO, and ZrO_2) in the glass produced during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds. The values in the black squares were measured in the glass, the black lines were the target in the glass, and the red lines were the expected value from analysis of the melter feeds (two-time recycle AP-107 from 0.00 – 5.15 kg and AP-105 from 5.15 – 9.70 kg).

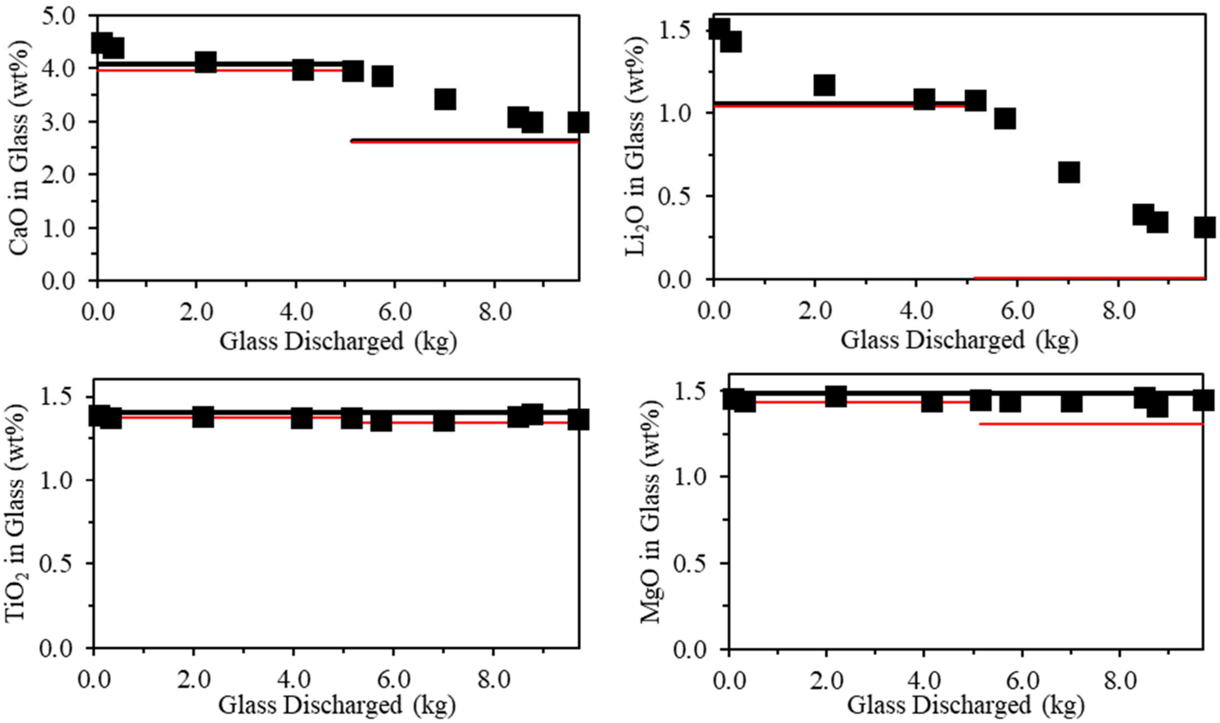

Figure 5.2. Content of a set of primary glass components (CaO, Li_2O, TiO_2, and MgO) in the glass produced during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds. The values in the black squares were measured in the glass, the black lines were the target in the glass, and the red lines were the expected value from analysis of the melter feeds (two-time recycle AP-107 from 0.00 – 5.15 kg and AP-105 from 5.15 – 9.70 kg).

5.2.2 Minor Glass Components

The compositional trends for each minor component (<1.00 wt%) measured in the glass product from the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds are displayed in Figure 5.3, with respect to the amount of glass discharged. Each graph shows the measured component content in the glass as black squares (■), the anticipated component content in the glass based on the target glass composition as a black line (━), and the expected component content in the glass based on the analyzed melter feed samples as a red line (━).

The trends for Cr_2O_3 and NiO in Figure 5.3 revealed a spike in their content above the glass target and melter feed levels at the start of the run, followed by a decrease with each subsequent glass pour. These trends indicated that when the glass inventory was idling in the CLSM vessel, during heat-up of the system or idling periods as previously observed (Dixon et al. 2020a and 2020b), Cr and Ni from the walls of the CLSM vessel were incorporated into the glass melt due to corrosion of the vessel. The CLSM vessel is constructed from Inconel-690[4], an alloy with relatively high Ni (minimum of 58.0 %) and Cr (a range from 27.0 to 31.0 %), with the balance provided by several additional components (Fe range from 7.0 to 11.0 %, Si at 0.50 % maximum, Mn at 0.50 % maximum, S at 0.015 % maximum, and Cu at 0.50 % maximum). A similar phenomenon has been observed in the DM10 melter, which is lined with refractory with high Cr levels and heated by Inconel-690 electrodes, after idling periods (Matlack et al. 2010, 2011, and 2018).

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[4] American Special Metals, Corp., Miami, Florida.

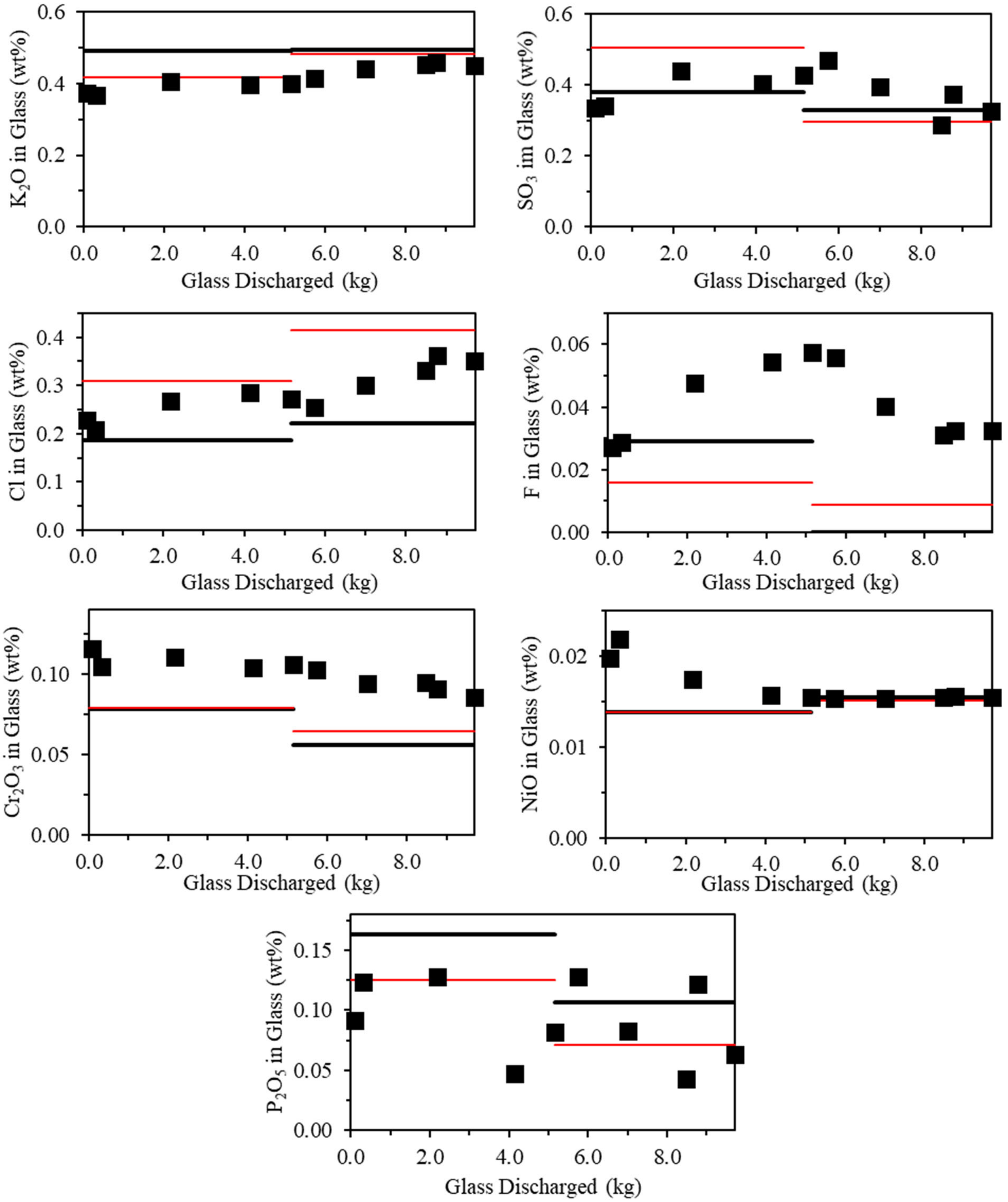

Figure 5.3. Content of minor glass components (K_2O, SO_3, Cl, F, Cr_2O_3, NiO, and P_2O_5) in the glass produced during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds. The values in the black squares were measured in the glass, the black lines were the target in the glass, and the red lines were the expected value from analysis of the melter feeds (two-time recycle AP-107 from 0.00 – 5.15 kg and AP-105 from 5.15 – 9.70 kg).

Other minor glass components present in the target glass compositions (SO_3, K_2O, P_2O_5, Cl, and F) varied by more than 10% from their glass target values in individual glass products due to reasons including, but not limited to, fluctuations in the melter feeds resulting in the actual target varying from the glass target, irregular volatility from the glass melt or in the cold cap, and analytical uncertainty due to the low concentrations compared to the primary components. The behavior of S, K, Cl, and F in the CLSM offgas treatment system will be discussed further in Section 5.4.

5.2.3 Minor Impurity Components

Several components were present as minor (300 ppm or less) impurities in the melter feeds and glass product during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds. The compositional trends for each minor impurity are shown in Figure 5.4 and Figure 5.5 with respect to the amount of glass discharged. Each graph shows the measured component content in the glass as black squares (■) and the expected component content in the glass based on the analyzed melter feed samples (if above the analytical detection limit) as a red line (—).

The general trend for the impurity components revealed that the components increased from their initial minimal level in the CLSM glass inventory to their greater expected values, based on their measured concentrations in the melter feeds, and then their concentrations plateaued at their expected values. For these components, their concentrations reached their expected values after 4 kg of glass had been poured from the CLSM. This result indicates that, in the CLSM system, impurities at greater concentrations in the incoming melter feed than in the glass product will reach their new expected value after 2 turnovers of the glass inventory. This phenomenon was also observed in the CLSM previously when processing the one-time recycle AP-107 melter feed (Dixon et al. 2020b).

Several components were exceptions to the general impurity trend. The concentration of Cu in the initial glass inventory was equal to the expected in the melter feed, so it did not increase, but remained at that level throughout the CLSM run. The concentrations of V, Y, and Sn in the glass product increased during the first 4 kg of glass pouring, in accordance with the general impurity trend, but the concentrations at which they plateaued for the remainder of the glass pouring were greater than their expected values in the melter feeds. These plateau values (~50 ppm for V, ~40 ppm for Y, and ~25 ppm for Sn), were similar to the concentrations of the components analyzed in the glass product from previous CLSM runs (Dixon et al. 2019, 2020a, and 2020b). The source of V, Y, and Sn at these levels in the glass products of different compositions is likely leaching from the material of the CLSM vessel, as described regarding the Cr_2O_3 and NiO content in the glass product in Section 5.2.2.

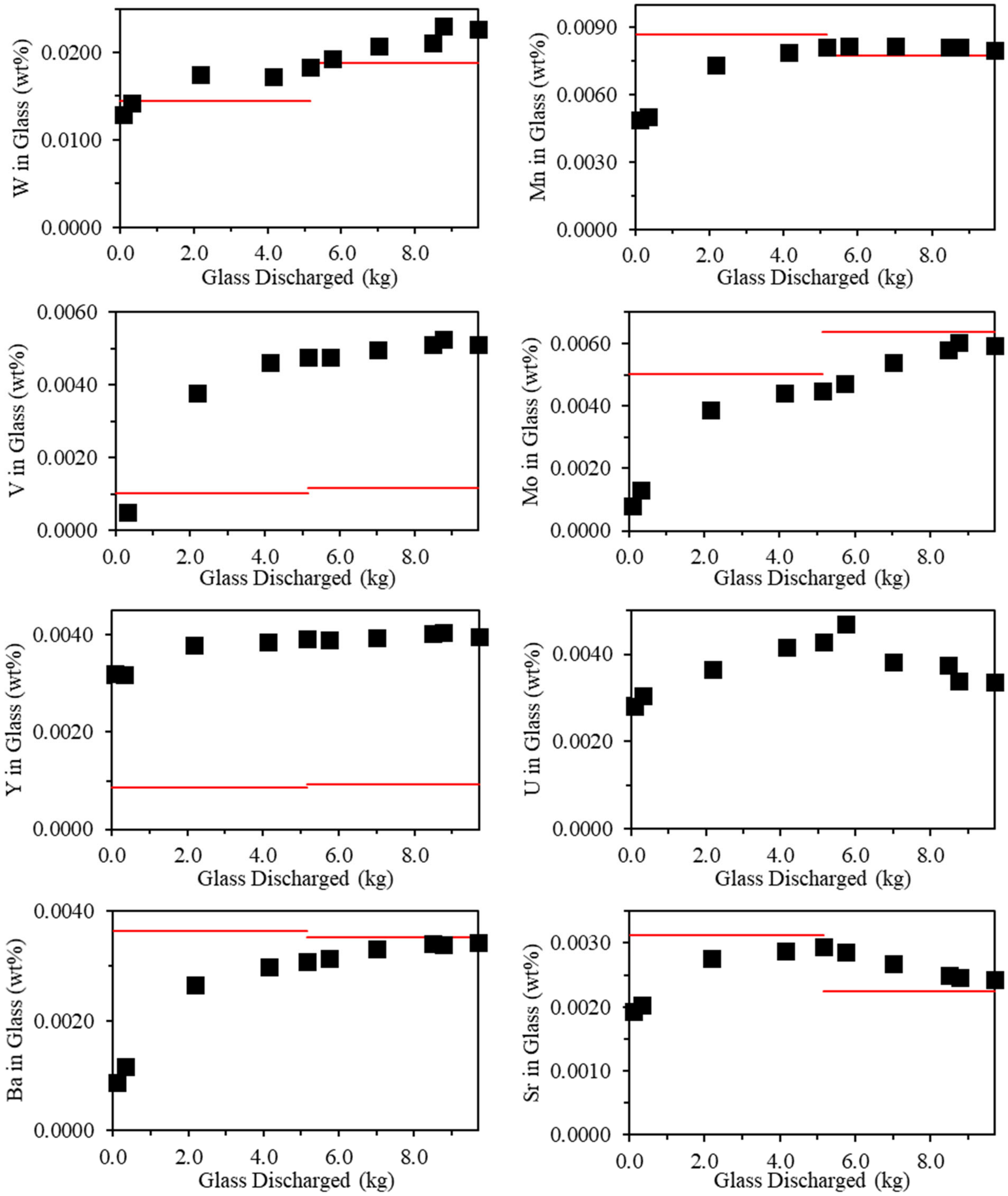

Figure 5.4. Content of a set of minor glass impurities (W, Mn, V, Mo, Y, U, Ba, and Sr) in the glass produced during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds. The values in the black squares were measured in the glass and the red lines were the expected value from analysis of the melter feeds (two-time recycle AP-107 from 0.00 – 5.15 kg and AP-105 from 5.15 – 9.70 kg).

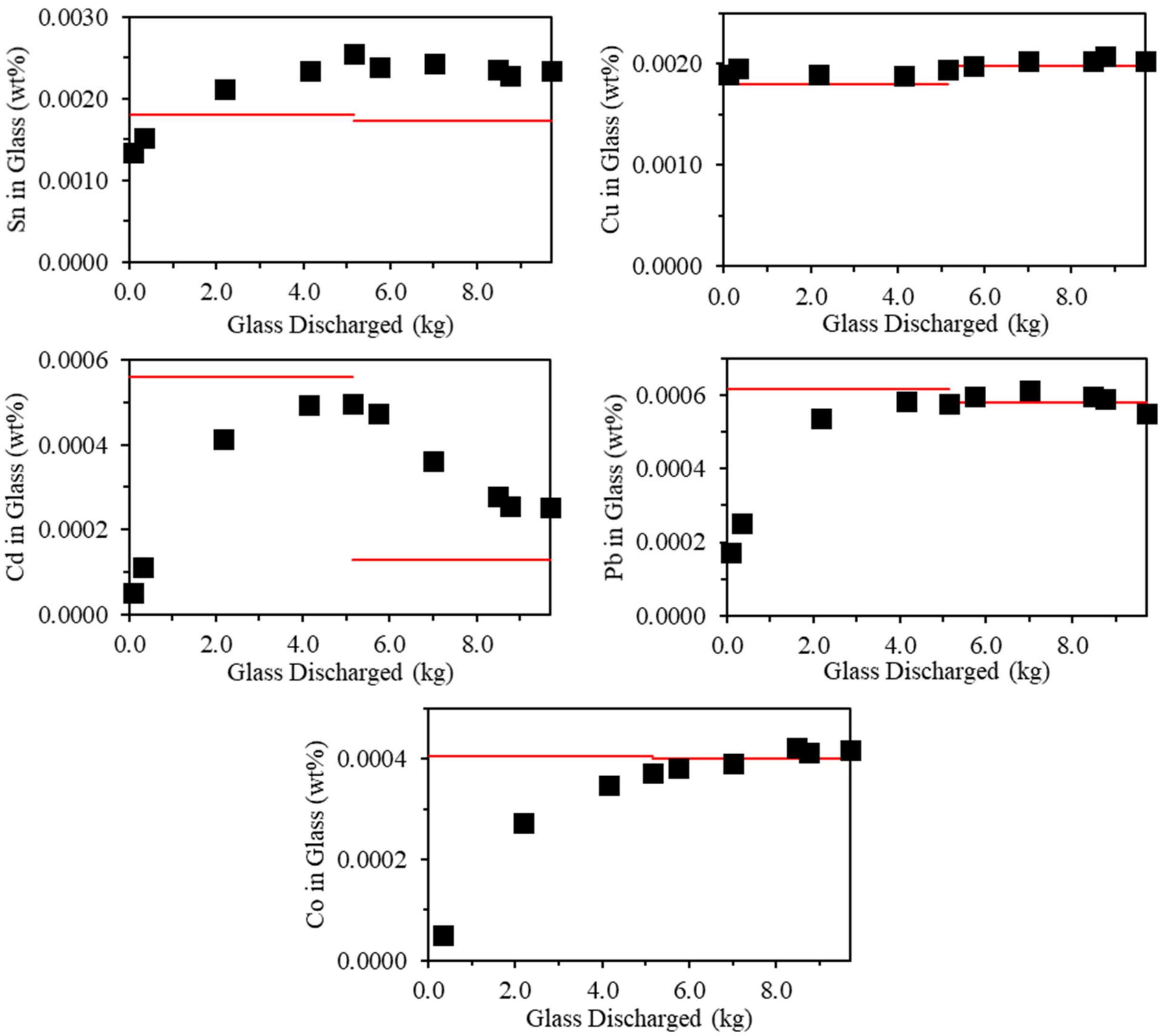

Figure 5.5. Content of a set of minor glass impurities (Sn, Cu, Cd, Pb, and Co) in the glass produced during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds. The values in the black squares were measured in the glass and the red lines were the expected value from analysis of the melter feeds (two-time recycle AP-107 from 0.00 – 5.15 kg and AP-105 from 5.15 – 9.70 kg).

The measured activity of each analyzed radionuclide in the glass products from the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds, with respect to the mass of glass discharged, is shown in Figure 5.6. The radionuclides in the glass product nominally followed the general impurity trend and were effectively retained in the glass product, with less than 1% of their inventories discovered in the offgas products.

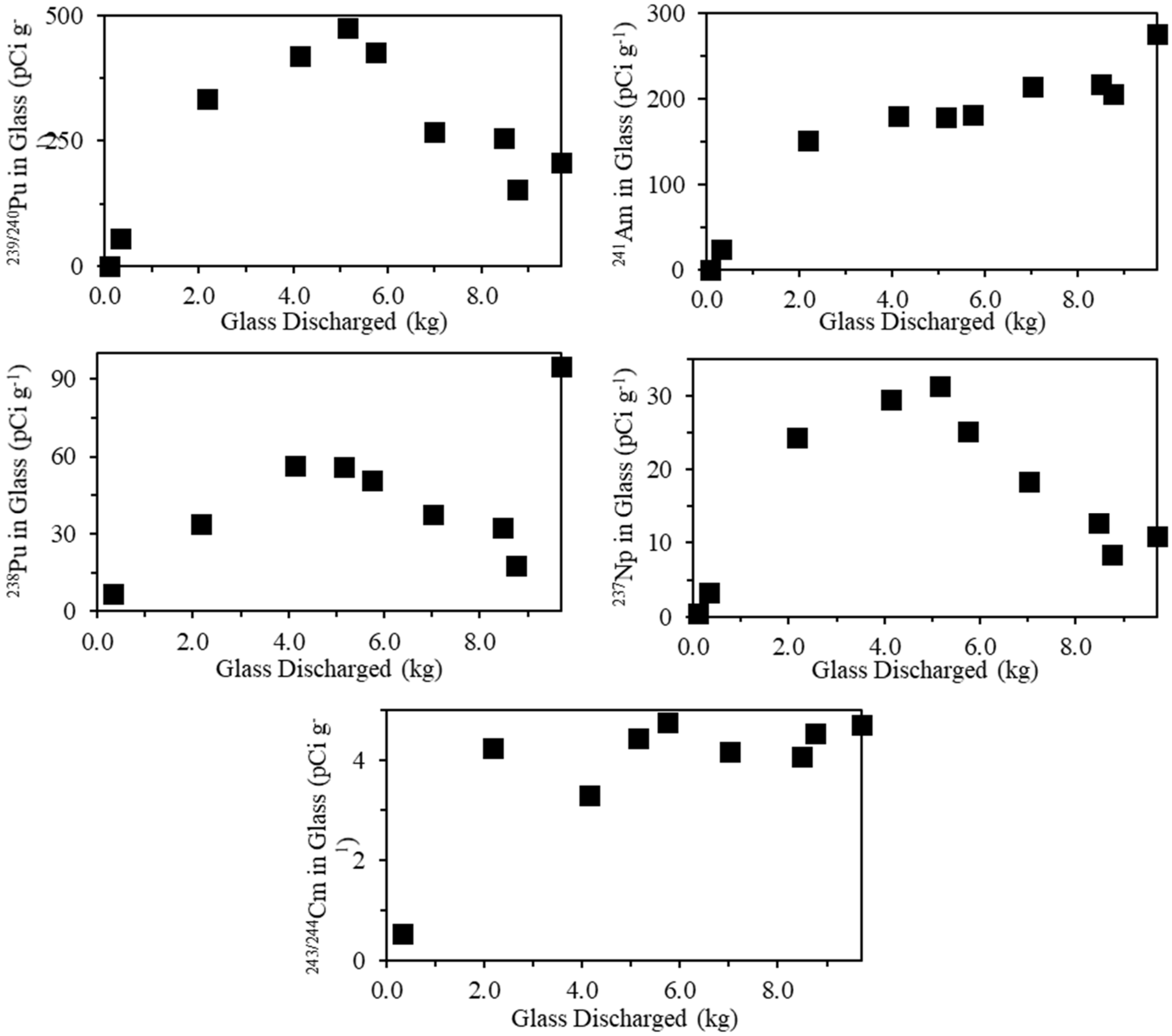

Figure 5.6. Activity of radionuclides ($^{239/240}$Pu, ^{241}Am, ^{238}Pu, ^{237}Np, and $^{243/244}$Cm) in the glass produced during the CLSM run with the two-time recycle AP-107 (from 0.00 – 5.15 kg) and AP-105 (from 5.15 – 9.70 kg) melter feeds.

5.3 DF, R, and Rec Calculations

For each component of the AP-107-2R and AP-105 glass compositions captured in the CLSM glass product (Table 5.1), and the radionuclides ^{99}Tc, Cs, and I where applicable, the following mass flow rates were calculated:

- Input into the CLSM vessel from the melter feed; $\dot{m}_{i,feed}$. Calculated during the run by dividing the total mass of each component (given the melter feed component concentrations listed in Table A.1 and the total mass of each melter feed composition calculated to have been consumed during the run, shown in Table 4.2) by the total runtime of 13.63 h.

- Output from the CLSM vessel in the glass product; $\dot{m}_{i,glass}$. Calculated from the glass component concentrations and the total glass mass produced of 9.70 kg, with the amount of each component present in the initial glass subtracted from the total mass.

- Output from the CLSM vessel in the captured offgas; $\dot{m}_{i,offgas}$. Calculated from the summation from all the offgas units, primarily the collected condensate of 6.50 kg, demister liquid of 3.19 kg, primary SBS sump of 1.03 kg, and filters, with the amount of every component in the appropriate number of blank HEPA filters subtracted from the total mass.

The mass flow rate data for the entire runtime of the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds are given in Table 5.2. Note that the chemical analysis results for Cs in the AP-105 melter feed and for I in both melter feeds and most glass pour samples were below the analytical reporting limits for those analytes, thus the reporting limit values, given in Table A.1 of Appendix A, were used for calculations where necessary and all related results should be treated as best estimates. From these mass flow rates, the DF_i, R_i, and Rec_i values, the latter two reported as percentages, were calculated as shown in Equations (5.1), (5.2), and (5.3), respectively. The values were calculated for the entire runtime of the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds, and are reported in Table 5.2.

Table 5.2. Component Mass Flow Rates, DFs, Retentions, and Recoveries During the CLSM Run with the Two-Time Recycle AP-107 and AP-105 Melter Feeds

Component	$\dot{m}_{i,feed}$	$\dot{m}_{i,glass}$	$\dot{m}_{i,offgas}$	Melter DF	R	Rec
Units	mg min^{-1}	mg min^{-1}	mg min^{-1}		%	%
^{99}Tc	0.10	0.06	0.04	2.2	55	97
Total Cs	0.01(a)	0.01	0.00	-20.3(b)	105(b)	159(b)
Total I	0.32(a)	0.18(a)	0.02	2.3(b)	56(b)	61(b)
Al	382	477	0	-4.0	125	125
B	376	452	2	-4.9	120	121
Ca	290	365	0	-3.9	126	126
Cl	44	49	8	-8.8	111	129
Cr	6	10	0	-1.5	167	170
F	2	7	1	-0.4	350	377
Fe	447	575	1	-3.5	129	129
K	45	55	1	-4.5	122	124
Li	32	42	0	-3.2	131	132
Mg	101	133	0	-3.2	132	132
Na	1718	2109	11	-4.4	123	123
Ni	1	2	0	-1.0	200	204
P	5	5	0	--	100	100
S	20	23	1	-6.7	115	119
Si	2604	3271	0	-3.9	126	126
Ti	99	124	0	-4.0	125	125
Zn	321	402	0	-4.0	125	125
Zr	247	311	0	-3.9	126	126
Total	6740	8411	25	-4.0(c)	125(c)	125(c)

(a) Component concentrations were below analytical reporting limits for the given stream and values should be considered best estimates.

(b) Values were calculated using best estimates for mass flow rates and should therefore be considered best estimates.

(c) Values were calculated using the mass flow rate totals from previous columns.

For most components, the $\dot{m}_{i,glass}$ values were greater than the $\dot{m}_{i,feed}$ values, resulting in negative DF_i values and R_i and Rec_i values that were above 100 %, generally by ~25 %. This indicates that the samples of melter feed collected and sent for analysis were more dilute than the actual melter feed charged during the CLSM run. This is most likely due to the sampling method employed to collect the samples, which required the stirring of the melter feed to be stopped temporarily during collection. The deficiencies of this collection method have been identified and an improved method will be used for future testing.

Despite the ~25% differences in the total values between the melter feed and glass mass flow rates due to the dilute melter feed samples collection, the ratios of components within the melter feeds seemed to be appropriately representative. Table 5.3 displays the comparison between the AP-107-2R and AP-105 target glass compositions and the compositions of glass expected from the conversion of the analyzed melter feeds. Most of the primary glass components (those present in >1.00 wt% amounts) were within 10% difference between target glass and melter feed analyzed compositions.

Table 5.3. Comparison of AP-107-2R and AP-105 Target Glass Compositions with the Compositions of the Analyzed Melter Feeds

Component	Target AP-107-2R	Analyzed Two-Time Recycle AP-107 Melter Feed	% Diff. Target to Analyzed AP-107-2R	Target AP-105	Analyzed AP-105 Melter Feed	% Diff. Target to Analyzed AP-105
	wt%	wt%	%	wt%	wt%	%
Al_2O_3	6.13	5.94	-3.1	6.13	5.93	-3.3
B_2O_3	9.95	9.76	-1.9	9.95	10.17	2.2
CaO	4.09	3.95	-3.5	2.64	2.61	-0.8
Cl	0.19	0.31	--	0.22	0.41	--
Cr_2O_3	0.08	0.08	--	0.06	0.06	--
F	0.03	0.02	--	0.00	0.01	--
Fe_2O_3	5.52	5.14	-6.9	5.52	5.38	-2.6
K_2O	0.49	0.42	--	0.49	0.48	--
Li_2O	1.06	1.04	-1.7	0.00	0.00	--
MgO	1.49	1.44	-3.4	1.49	1.31	-11.9
Na_2O	16.89	18.67	10.5	19.35	19.45	0.5
NiO	0.01	0.01	--	0.02	0.02	--
P_2O_5	0.16	0.13	--	0.11	0.07	--
SO_3	0.38	0.51	--	0.33	0.30	--
SiO_2	45.60	45.21	-0.9	45.76	46.42	1.4
TiO_2	1.40	1.37	-2.2	1.40	1.35	-4.2
ZnO	3.51	3.26	-7.1	3.51	3.32	-5.5
ZrO_2	3.02	2.77	-8.2	3.02	2.71	-10.1
Total	100.0	100.0		100.0	100.0	

Values marked with '--' were not calculated because the component target concentrations were <1%.

Two primary glass components in the AP-105 melter feed varied by more than 10% from the target AP-105 glass composition: MgO and ZrO_2. Both components, MgO (Dixon et al. 2020a) and ZrO_2 (Dixon et al. 2020b), were lower than their targets, as have previously been analyzed in other melter feed compositions, due to their heavy presence as minerals, olivine and zircon, with a tendency to settle if not well-agitated. The one component in the two-time recycle AP-107 melter feed that varied by more than 10% from the target AP-107-2R glass composition was Na_2O. The reason for the variation can be traced to the estimated composition for the recycled condensate that was used in the Kim et al. (2012) glass models to calculate the AP-107-2R glass composition. The analyzed composition for the recycled condensate, given as the 'Concentrate' values in Table 3.2 of Section 3.1.3, was much higher in Na concentration than expected due to the amount of NaOH needed to adjust the pH of the condensate to its appropriate value. This difference caused the calculated glass target to have a lower Na_2O content than the actual melter feed prepared.

5.4 Offgas Analysis

The seven components detected in appreciable quantities in the samples collected from the various units in the CLSM offgas treatment system during prior runs (Dixon et al. 2020b) were ^{99}Tc, Cs, I, S, K, Cl, and F. These components were likewise present in the samples collected from the offgas treatment system during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds. The total quantities of these components collected in the CLSM vessel output streams, the glass, and 4 collective units in the offgas treatment system (the sampling loop, primary SBS sump, accumulated condensate, and filters) are shown in Table 5.4.

Table 5.4. Quantities of Selected Components in CLSM Output Streams During the CLSM Run with the Two-Time Recycle AP-107 and AP-105 Melter Feeds

	^{99}Tc	Cs	I	Cl	F	K	S
Units	mg	mg	mg	mg	mg	mg	mg
Glass	45.6	5.28	110[(b)]	39673	5344	44950	18735
Wash + Sampling[(a)]	8.31 (1.66)	0.637 (0.192)	2.94 (0.531)	1423 (463)	109 (10.3)	157 (60.6)	159 (26.6)
SBS Sump	2.21	0.161	1.29	586	25.7	45.4	36.2
Condensate	12.0	1.19	5.63	2559	251	233	280
End Filters	19.0	1.20	4.86	2563	160	347	320

[(a)] Values outside of parentheses were calculated to consider the amount of each component expected to be present during an offgas piping wash; the values inside of parentheses were calculated only from the sampling HEPA filters.

[(b)] Iodine concentrations in glass samples were below analytical reporting limits; the value should be considered a best estimate.

The measured sampling loop values are given in parentheses in the ‘Wash + Sampling’ row of Table 5.4. From previous offgas analysis in the simulant CLSM system (Dixon et al. 2020a and 2021), when the offgas piping from the CLSM vessel to the sampling loop was washed upon the conclusion of each CLSM run, about 20% of the inventory of each component (Re, S, K, Cl, and F) recovered in the offgas system was found in the offgas piping wash. Since the radioactive CLSM system in RPL cannot be disassembled and washed for analysis due to safety restrictions, the same recovery principle was applied as an assumption for all the components recovered in the offgas system during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds. Thus, the estimated quantities of each component, factoring up to 20% in a theoretical offgas piping wash, are shown without parentheses in the ‘Wash + Sampling’ row of Table 5.4.

The quantities of each component in the output streams from Table 5.4 were converted to percentages of the total quantities in the output streams via Eq. (5.4):

$$Output\%_{i,s} = \frac{m_{i,s}}{\sum_s m_{i,s}} \times 100 \tag{5.4}$$

where $Output\%_{i,s}$ is the percentage (%) of the quantity of a component (i; ^{99}Tc, Cs, I, Cl, F, K, S) in each output stream (s; Glass, Wash + Sampling, SBS Sump, Condensate, End Filters) and $m_{i,s}$ is the mass of a component (i) in stream (s). The $Output\%_{i,s}$ is displayed in Figure 5.7a for each of the streams in the offgas treatment system during the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds.

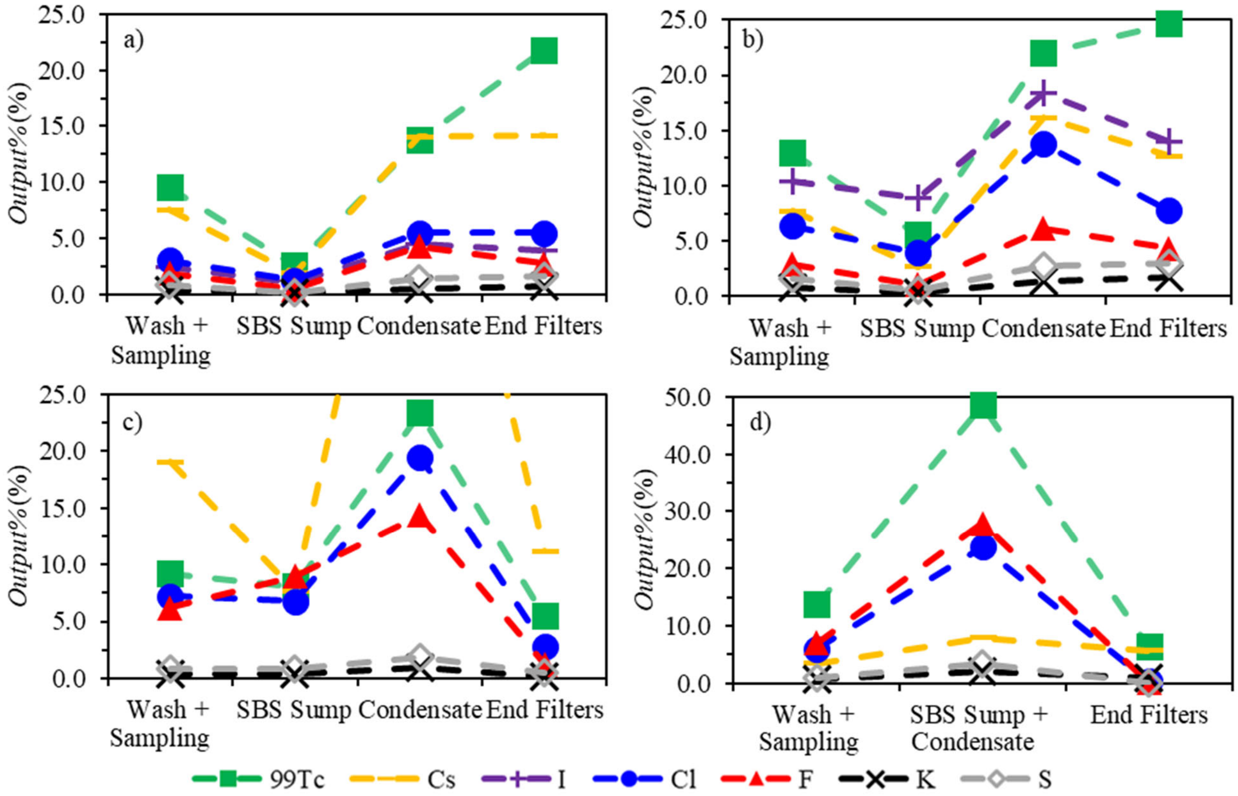

Figure 5.7. Percentage of the quantity of each component in the output stream in the CLSM offgas treatment system during runs with a) the two-time recycle AP-107 and AP-105 melter feeds, b) one-time recycle AP-107 melter feed (Dixon et al. 2020b), c) AP-107 melter feed (Dixon et al. 2019), and d) AP-105 melter feed (Dixon et al. 2018).

In addition, the $Output\%_{i,s}$ was calculated for all prior CLSM runs with Hanford tank wastes, all of which are also displayed in Figure 5.7. The pre-filter unit in the offgas treatment system was not a part of the design for the CLSM runs with AP-107 melter feed (Dixon et al. 2019; shown in Figure 5.7c) and AP-105 melter feed (Dixon et al. 2018; shown in Figure 5.7d) and lower quantities of each component were captured in the end filters as a result during those runs. An unexpected spike of Cs in the initial glass present in the vessel for the CLSM run with AP-107 melter feed caused greater quantities to be gathered in the offgas treatment system during the run (Dixon et al. 2019). The SBS sump and condensate were collected and analyzed together for the CLSM run with AP-105 melter feed (Dixon et al. 2018). Considering these factors, the behavior of the components captured in appreciable quantities in the CLSM offgas treatment system did not vary considerably between the melter feeds with different compositions.

5.5 ^{99}Tc and Cs Retention and Analysis

The measured content of ^{99}Tc and Cs in the glass product, with respect to the mass of glass discharged, and their expected component content in the glass based on the analyzed melter feed samples if 100% retained are marked by the red inset line in Figure 5.8. The Cs concentration in the AP-105 melter feed, displayed in the Cs graph in Figure 5.8 as the inset red line from 5 to 9.7 kg of glass discharged, was below analytical reporting limits and should be considered a best estimate. The Tc and Cs retention values, R_{99Tc} and R_{Cs}, calculated for each glass pour are also displayed in Figure 5.8 with respect to the mass of glass discharged. The characteristic relationships between ^{99}Tc and Cs are shown in Table 5.5 for the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds.

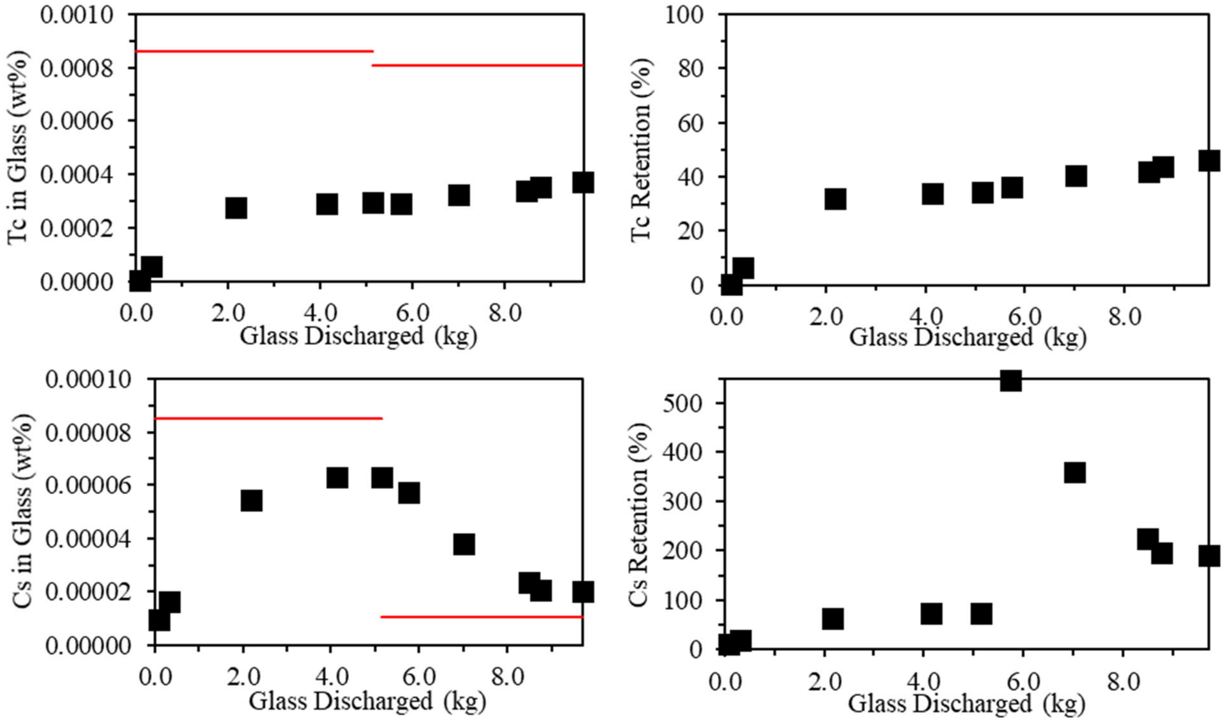

Figure 5.8. Measured ^{99}Tc and Cs content and retention in the glass product from the CLSM run with the two-time recycle AP-107 (from 0.00 – 5.15 kg) and AP-105 (from 5.15 – 9.70 kg) melter feeds.

Table 5.5. ^{99}Tc and Cs Relationships During the CLSM Run with the Two-Time Recycle AP-107 and AP-105 Melter Feeds

^{99}Tc Glass Target (wt%)	Cs Glass Target (wt%)	^{99}Tc/Cs Target Mass Ratio (mg mg^{-1})	Glass Pour Time (h)	Glass Discharged (kg)	^{99}Tc Glass Actual (wt%)	Cs Glass Actual (wt%)	^{99}Tc Retention (%)	Cs Retention (%)
8.63E-04	8.49E-05	10.2	0.17	0.10	1.14E-06	9.49E-06	0	11
8.63E-04	8.49E-05	10.2	0.80	0.32	5.39E-05	1.62E-05	6	19
8.63E-04	8.49E-05	10.2	3.95	2.18	2.77E-04	5.44E-05	32	64
8.63E-04	8.49E-05	10.2	6.53	4.15	2.92E-04	6.30E-05	34	74
8.63E-04	8.49E-05	10.2	7.13	5.15	2.97E-04	6.30E-05	34	74
8.06E-04	1.05E-05	76.6	8.75	5.75	2.92E-04	5.74E-05	36	546
8.06E-04	1.05E-05	76.6	10.23	7.01	3.25E-04	3.78E-05	40	359
8.06E-04	1.05E-05	76.6	12.25	8.48	3.38E-04	2.37E-05	42	225
8.06E-04	1.05E-05	76.6	12.80	8.76	3.53E-04	2.05E-05	44	195
8.06E-04	1.05E-05	76.6	14.31	9.70	3.74E-04	2.01E-05	46	191
						Total, R_i	44[(a)]	84[(a)]
						Total, Rec_i	84[(a,b)]	135[(a,b)]

[(a)] Values were calculated based on the conversion of melter feed into glass and thus are different from the calculated values in Table 5.2.

[(b)] Values reported include the estimate for mass recovered in offgas piping wash, described in Section 5.4.

Both ^{99}Tc and Cs were present at minimum quantities in the initial glass in the CLSM vessel at the start of the run. Once melter feed charging began and both components began to be integrated into the glass product, it took about 1 turnover of the glass inventory (2 kg of glass poured) for both to reach a relative chemical steady state that held while the two-time recycle AP-107 melter feed was being charged. When the melter feed being processed changed to AP-105, between 5.15 and 5.75 kg of glass discharged, the ^{99}Tc content in the melter feed remained relatively constant while the content in the glass, and thus the R_{99Tc} values, increased for about 2 glass turnovers before reaching a relative chemical steady state. In the AP-105 melter feed, the Cs content was ~15% of the content in the two-time recycle AP-107 melter feed. When this target changed, the Cs content in the glass decreased for about 2 glass turnovers before reaching a relative chemical steady state. At this relative chemical steady state, with a ^{99}Tc/Cs mass ratio of 76.6, the Cs was still retained at a high level in the glass and did not volatilize at a dramatically increased level as theorized from previous melter runs with LAW simulant (Matlack et al. 2004).

The ^{99}Tc/Cs mass ratio during the two-time recycle AP-107 melter feed charging (see glass pours through 5.15 kg discharged in Table 5.5) was 10.2, while the twice-recycle retention values were R_{99Tc} = 34 % and R_{Cs} = 74 % when their contents reached a relative chemical steady state. The average glass production rate during the two-time recycle AP-107 melter feed charging (see Table 4.2) was essentially equivalent to the average rate during the non-recycle AP-107 melter feed charging in the CLSM (1477 kg m^{-2} d^{-1}; Dixon et al. 2019) but was less than the average rate during the one-time recycle AP-107 melter feed charging in the CLSM (1739 kg m^{-2} d^{-1}; Dixon et al. 2020b). The twice-recycle R_{99Tc} value at the relative chemical steady state during the two-time recycle AP-107 melter feed charging was ~5% less than the value during the non-recycle AP-107 melter feed charging in the CLSM (39%; Dixon et al. 2019) but was ~2% greater than the average rate during the one-time recycle AP-107 melter feed charging in the CLSM (32%; Dixon et al. 2020b). Atypical charging conditions and glass settings in the CLSM vessel, where the glass level in the CLSM vessel was below the target 2 kg inventory, during the CLSM run with the one-time recycle AP-107 melter feed were speculated to have affected the glass production and retention characteristics during that run (Dixon et al. 2020b), and the CLSM run with the two-time recycle AP-107 melter feed seemed to confirm this assumption by lying closer to the non-recycle production results.

The ^{99}Tc/Cs mass ratio during the AP-105 melter feed charging (see glass pours after 5.15 kg discharged in Table 5.5) was 76.6, while the single-pass retention values were R_{99Tc} = 44 % and R_{Cs} = 185 % when their contents reached a relative chemical steady state. The average glass production rate during the AP-105 melter feed charging (see Table 4.2) was greater than the average rate during the previous run with AP-105 in the CLSM (1330 kg m^{-2} d^{-1}; Dixon et al. 2018), but within the typical range when processing the same melter feed in the CLSM (Dixon et al. 2020a). The single-pass R_{99Tc} value at the relative chemical steady state during the AP-105 melter feed charging was ~20% greater than the value during the previous run with AP-105 in the CLSM (24%; Dixon et al. 2019), though difficulties controlling the system during the previous run were believed to have reduced the ^{99}Tc retention compared to the expectation if charging was consistent.

6.0 Conclusions

During the previous processing of AP-107 waste with recycled condensate in the CLSM, 7.5 L of offgas condensate were collected during vitrification. A test apparatus was designed to operate like the EMF evaporator and was used to successfully concentrate the AP-107 condensate by a factor of 10 while retaining over 95 % of the ^{99}Tc, Cs, and I inventory. A sample of AP-107 waste was retrieved by WRPS and received at the RPL by PNNL. After undergoing solids filtration and cesium removal by ion exchange, a portion of the waste was combined with the AP-107 condensate concentrate to approximate a second cycle of the recycle action to be performed at the WTP. In addition, a sample of AP-105 waste was retrieved by WRPS, received at the RPL by PNNL, and sent through solids filtration and cesium removal. Glass compositions were calculated from the Kim et al. (2012) glass models based on the expected composition of the combined AP-107 and condensate concentrate and the analyzed AP-105 composition, referred to as AP-107-2R and AP-105, respectively. GFCs were added to the two wastes and the resultant melter feeds were processed in the CLSM.

Over 13.63 hours of processing, 9.70 kg of glass were produced, for an average glass production rate of 1464 kg m^2 d^{-1} for AP-107-2R and 1568 kg m^2 d^{-1} for AP-105. The production rate for AP-107-2R was less than the processing rate of the AP-107 waste with one-time recycle, but nearly equivalent to that of AP-107 without recycle. The production rate for AP-105 was greater than previous processing, but within the typical range when processing the same melter feed in the CLSM. Other conclusions from this melter run include:

- No strange or challenging cold-cap behavior or processing operations were observed to be caused from the transition from the AP-107 to AP-105 melter feed composition; however, there were noticeable differences in cold-cap behavior under each melter feed composition in alignment with what had been seen in previous runs with AP-107 and AP-105 wastes.

- All the primary components in the glass product from the CLSM were within 10 % of their targets based on the AP-107-2R and AP-105 glass compositions.
- Two components, CaO and Li_2O, had targets greater in the AP-107-2R than in the AP-105 glass composition. After the change in melter feed being processed, each component reached within 10% of its target value after two turnovers of the CLSM vessel glass inventory (4 kg glass discharged).
- Components recovered in the CLSM offgas system (^{99}Tc, Cs, I, S, K, Cl, and F) were recovered in similar proportions in each unit of the offgas system compared to previous CLSM runs.
- While the two-time recycle AP-107 melter feed was being charged at a relative chemical steady state, 34% of the ^{99}Tc and 74% of the Cs were retained in the glass product.
- Both the ^{99}Tc and Cs retentions in the AP-107-2R glass were greater than in the previous run with the one-time recycle glass, but the ^{99}Tc retention was less than in the no-recycle run.
- While the AP-105 melter feed was being charged at a relative chemical steady state, 44% of the ^{99}Tc and 185% of the Cs were retained in the glass product, the latter due to a much lower amount of Cs in the AP-105 melter feed than in the two-time recycle AP-107 melter feed.
- The ^{99}Tc retention in the AP-105 glass was greater than in the previous AP-105 run because a relative chemical steady state was not properly achieved in the previous run.
- Cs results indicated that Cs did not dramatically volatilize from the glass melt when the ^{99}Tc/Cs mass ratio increased from 10 to 77.

7.0 References

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Appendix A – Chemical Analysis of Samples Collected from the CLSM Run with the Two-Time Recycle AP-107 and AP-105 Melter Feeds

The table in this section displays the complete chemical analytical results for all samples from the CLSM run with the two-time recycle AP-107 and AP-105 melter feeds that were sent to SwRI for analysis. Values noted with "<" indicate the associated sample results were less than the analytical reporting limit (ARL). Values marked with "--" denote that the analysis was not performed for a specific sample.

Table A.1. Chemical Analysis of Samples Produced During the CLSM Run with the Two-Time Recycle AP-107 and AP-105 Melter Feeds

	Sample Mass	Component Concentration (mg kg^{-1})										
Sample Name	(kg)	^{99}Tc	Total Cs	Total I	Al	Ba	B	Cd	Ca	Cr	Co	Cu
2× Recycle AP-107 Melter Feed	11.84	3.90	0.384	<11.6	14200	16.5	13700	2.53	12750	244	1.84	8.14
AP-105 Melter Feed	10.29	3.60	<0.0470	<11.9	14000	15.7	14100	0.572	8340	197	1.79	8.82
Glass Pour 0.17	0.16	0.0113	0.0943	<12.1	32700	8.67	30800	0.501	31900	785	<0.493	18.9
Glass Pour 0.80	0.29	0.541	0.163	<11.5	32600	11.7	32000	1.13	31550	720	0.501	19.7
Glass Pour 3.95	2.40	2.78	0.545	<12.1	32000	26.6	29200	4.13	29600	758	2.72	19.0
Glass Pour 6.53	2.43	2.92	0.629	<11.5	31400	29.8	30300	4.92	28400	708	3.46	18.8
Glass Pour 7.13	0.31	2.98	0.631	<11.6	31500	30.8	29700	4.96	28400	724	3.73	19.5
Glass Pour 8.75	0.73	2.91	0.573	17.7	31400	31.3	30000	4.73	27500	702	3.80	19.8
Glass Pour 10.23	1.53	3.24	0.376	<11.6	31200	32.9	30600	3.61	24400	639	3.88	20.2
Glass Pour 12.25	1.81	3.39	0.238	<11.3	31700	34.1	29500	2.78	22100	648	4.23	20.4
Glass Pour 12.80	0.35	3.53	0.205	<12.1	31700	33.9	29000	2.56	21300	622	4.13	20.7
Glass Pour 14.31	4.34	3.76	0.202	<11.3	31900	34.4	30300	2.55	21500	587	4.19	20.4
Pre-filter	0.43	18.0	0.591	1.06	9.27	<0.321	264	<0.321	11.8	51.5	<0.321	0.449
Primary HEPA Filter A	0.01	1.42	0.132	11.2	12000	16300	11350	<0.491	4860	<47.5	<49.1	2.43
Sampling HEPA Filter 1	0.02	43.5	6.71	<11.9	11600	15500	12100	<0.485	4490	125	<48.5	1.48
Sampling HEPA Filter 2	0.02	35.7	2.31	<11.5	11700	15900	11800	<0.477	4620	48.7	<47.7	1.27
Sampling HEPA Filter 3	0.02	29.1	3.52	<11.3	11800	15600	11900	<0.444	4530	< 42.2	<44.4	1.19
Condensate A	3.28	1.59	0.186	0.733	10.7	< 0.248	127	<0.248	13.7	16.6	<0.248	<0.248
Condensate B	0.71	2.10	0.129	1.05	9.31	< 0.248	140	<0.248	12.2	8.49	<0.248	<0.248
Condensate C	2.51	2.09	0.193	0.988	12.4	< 0.249	141	<0.249	10.1	5.80	<0.249	<0.249
Demister A	0.72	2.65	0.288	1.09	14.8	< 0.247	185	<0.247	18.8	25.2	<0.247	0.267
Demister B	2.48	2.13	0.193	1.05	13.6	< 0.249	138	<0.249	11.5	5.10	<0.249	<0.249
Primary SBS Sump	1.03	2.15	0.156	1.25	15.2	< 0.250	134	<0.250	8.44	2.83	<0.250	<0.250
Pre-filter Fluid	0.59	6.80	0.433	1.47	12.3	<0.235	216	<0.235	14.3	49.4	<0.235	0.539

Table A.1. Chemical Analysis of Samples Produced During the CLSM Run with the Two-Time Recycle AP-107 and AP-105 Melter Feeds (cont.)

	Component Concentration (mg kg^{-1})											
Sample Name	Fe	La	Pb	Li	Mg	Mn	Mo	Ni	P	K	Si	Na
2× Recycle AP-107 Melter Feed	16250	<1.43	2.79	2180	3920	39.1	22.7	49.0	247	1560	95550	62600
AP-105 Melter Feed	16800	<1.41	2.59	5.71	3530	34.5	28.4	53.1	139	1790	96900	64400
Glass Pour 0.17	38000	<1.48	1.70	6990	8730	48.6	7.93	154	396	3070	213000	119000
Glass Pour 0.80	38200	<1.43	2.54	6690	8735	50.4	12.9	172	542	3065	216000	120500
Glass Pour 3.95	37700	1.67	5.39	5440	8910	73.4	38.8	137	559	3370	214000	133000
Glass Pour 6.53	37300	1.59	5.83	5030	8700	78.6	44.0	123	204	3290	213000	134000
Glass Pour 7.13	37400	1.96	5.77	5020	8770	81.4	44.9	122	359	3330	214000	135000
Glass Pour 8.75	37400	1.90	5.94	4490	8700	81.5	46.9	120	558	3440	213000	135000
Glass Pour 10.23	38200	1.70	6.09	2980	8650	81.2	53.8	120	359	3650	213000	138000
Glass Pour 12.25	38700	1.61	5.98	1830	8850	81.4	58.2	122	186	3760	218000	140000
Glass Pour 12.80	37300	<1.40	5.88	1610	8510	81.1	60.3	122	529	3800	218000	140000
Glass Pour 14.31	38400	1.58	5.54	1480	8800	80.2	59.6	122	276	3760	219000	140000
Pre-filter	170	<0.963	<0.482	22.5	<3.21	0.921	2.13	2.46	<8.03	301	16.0	3605
Primary HEPA Filter A	387	<1.47	2.57	4.02	719	14.6	24.5	26.9	<12.3	9155	373000	25800
Sampling HEPA Filter 1	350	<1.46	2.89	75.4	666	5.22	44.4	6.81	29.8	9470	363000	33200
Sampling HEPA Filter 2	285	<1.43	3.51	40.4	666	4.90	25.1	3.27	16.8	9310	356000	31800
Sampling HEPA Filter 3	293	<1.33	2.26	21.8	668	7.24	27.2	5.43	18.9	9410	365000	32400
Condensate A	29.6	<0.745	<0.373	6.17	<2.48	0.314	<0.497	3.25	<6.21	32.5	37.8	462
Condensate B	22.7	<0.744	<0.372	6.52	<2.48	<0.248	<0.496	0.884	<6.20	37.7	35.7	576
Condensate C	20.9	<0.746	<0.373	4.49	<2.49	<0.249	<0.497	0.422	<6.22	39.5	28.7	606
Demister A	62.3	<0.741	<0.371	9.85	2.74	0.825	1.11	6.50	<6.18	52.6	39.7	779
Demister B	21.9	<0.746	<0.373	3.81	<2.49	<0.249	<0.497	0.318	<6.22	40.8	29.5	633
Primary SBS Sump	7.05	<0.749	<0.374	2.16	<2.50	<0.250	<0.499	0.634	<6.24	44.1	18.6	655
Pre-filter Fluid	113	<0.708	<0.353	13.6	<2.36	1.79	3.05	15.1	<5.88	115	27.4	1450

Table A.1. Chemical Analysis of Samples Produced During the CLSM Run with the Two-Time Recycle AP-107 and AP-105 Melter Feeds (cont.)

	Component Concentration (mg kg^{-1})											
Sample Name	Sr	S	Sn	Ti	W	U	V	Y	Zn	Zr	Cl	F
2× Recycle AP-107 Melter Feed	14.1	916	8.15	3720	65.5	<23.9	4.66	3.88	11850	9265	1400	72.0
AP-105 Melter Feed	10.0	529	7.70	3600	84.1	<23.5	5.17	4.09	11900	8960	1850	39.7
Glass Pour 0.17	19.2	1330	13.3	8270	128	27.8	<0.987	31.8	27600	21100	2270	269
Glass Pour 0.80	20.4	1375	15.3	8265	142	30.6	4.87	31.9	26300	20900	2085	288
Glass Pour 3.95	27.6	1770	21.2	8290	175	36.7	37.9	38.1	26300	20900	2680	477
Glass Pour 6.53	28.7	1610	23.4	8240	172	41.6	46.1	38.5	26200	20700	2840	542
Glass Pour 7.13	29.5	1720	25.5	8270	183	42.9	47.7	39.3	26700	20800	2720	577
Glass Pour 8.75	28.5	1880	23.8	8120	192	46.9	47.5	38.8	26500	20200	2540	556
Glass Pour 10.23	26.7	1570	24.2	8090	206	38.0	49.3	39.3	26400	20100	2990	400
Glass Pour 12.25	25.0	1150	23.6	8290	212	37.6	51.2	40.4	26800	20800	3330	312
Glass Pour 12.80	24.5	1500	22.8	8370	230	33.9	52.3	40.5	26600	21100	3620	324
Glass Pour 14.31	24.4	1310	23.5	8220	228	33.9	51.3	39.8	27300	20500	3530	326
Pre-filter	<0.321	236	<1.28	0.654	<1.61	<16.1	0.886	<0.321	24.9	<0.963	556	26.6
Primary HEPA Filter A	234	267	5.00	19.8	47.3	<24.6	<0.982	2.27	10300	92.5	21.1	15.2
Sampling HEPA Filter 1	222	887	4.28	23.9	55.2	<24.3	<0.970	2.18	10300	91.1	10200	448
Sampling HEPA Filter 2	228	744	3.75	21.1	46.5	<23.9	<0.955	2.12	9990	90.2	9480	139
Sampling HEPA Filter 3	223	569	5.23	21.1	51.2	<22.2	<0.887	2.23	10300	90.9	10600	79.6
Condensate A	<0.248	45.6	<0.993	1.16	<1.24	<12.4	<0.248	<0.248	28.4	1.41	305	35.3
Condensate B	<0.248	44.1	<0.992	0.786	<1.24	<12.4	<0.248	<0.248	25.8	1.25	427	44.8
Condensate C	<0.249	39.5	<0.994	0.620	<1.24	<12.4	<0.249	<0.249	27.6	0.777	500	41.3
Demister A	<0.247	71.1	<0.988	1.77	<1.24	<12.4	<0.247	<0.247	40.5	1.90	541	44.9
Demister B	<0.249	37.6	<0.995	0.755	<1.24	<12.4	<0.249	<0.249	30.7	0.812	585	39.0
Primary SBS Sump	<0.250	35.1	<0.998	<0.250	<1.25	<12.5	<0.250	<0.250	31.3	<0.749	569	25.0
Pre-filter Fluid	<0.235	123	<0.941	1.39	<1.18	<11.8	<0.235	<0.235	34.5	1.43	823	33.9

Table A.1. Chemical Analysis of Samples Produced During the CLSM Run with the Two-Time Recycle AP-107 and AP-105 Melter Feeds (cont.)

	Component Concentration (mg kg^{-1})						Component Concentration (pCi g^{-1})				
Sample Name	N (Nitrate)	N (Nitrite)	Sulfate	Bromide	Phosphate	TOC	^{241}Am	$^{243/244}$Cm	^{237}Np	^{238}Pu	$^{239/240}$Pu
2× Recycle AP-107 Melter Feed	13700	7265	2530	<17.5	38.1	13000	9.31E+01	<1.33E+00	1.50E+01	2.52E+01	2.18E+02
AP-105 Melter Feed	12900	9070	1350	<37.1	32.5	14200	1.11E+02	<1.33E+00	<9.82E-01	<6.60E-01	8.48E+00
Glass Pour 0.17	--	--	3510	<242	--	--	<1.01E-01	<-9.99E-02	<5.27E-01	<-5.25E-01	<5.24E-01
Glass Pour 0.80	--	--	3945	<230	--	--	2.39E+01	<5.40E-01	3.32E+00	6.88E+00	5.72E+01
Glass Pour 3.95	--	--	3580	<242	--	--	1.52E+02	4.23E+00	2.44E+01	3.38E+01	3.34E+02
Glass Pour 6.53	--	--	3480	<230	--	--	1.80E+02	3.29E+00	2.95E+01	5.66E+01	4.18E+02
Glass Pour 7.13	--	--	3300	<233	--	--	1.79E+02	4.44E+00	3.13E+01	5.60E+01	4.75E+02
Glass Pour 8.75	--	--	3450	<237	--	--	1.82E+02	4.76E+00	2.51E+01	5.09E+01	4.27E+02
Glass Pour 10.23	--	--	4140	<233	--	--	2.15E+02	4.17E+00	1.83E+01	3.74E+01	2.69E+02
Glass Pour 12.25	--	--	2810	<227	--	--	2.17E+02	4.06E+00	1.27E+01	3.25E+01	2.55E+02
Glass Pour 12.80	--	--	2720	<242	--	--	2.06E+02	4.54E+00	8.46E+00	1.75E+01	1.53E+02
Glass Pour 14.31	--	--	2960	<226	--	--	2.77E+02	4.69E+00	1.09E+01	9.47E+01	2.06E+02
Pre-filter	25000	25.1	1010	<6.42	<6.42	--	1.41E-01	<6.43E-03	1.61E-02	5.19E-02	2.11E-01
Primary HEPA Filter A	392	<0.950	46.7	<0.950	<0.950	--	<1.47E-01	<-2.47E-01	8.93E-01	<6.68E-01	<3.09E-01
Sampling HEPA Filter 1	437	100	2210	61.9	6.05	--	<1.03E-01	<1.03E-01	<7.82E-02	<4.00E-01	2.60E+00
Sampling HEPA Filter 2	353	78.6	1180	67.7	2.58	--	<7.94E-01	<0.00E+00	<1.56E-01	<-3.81E-02	<7.62E-02
Sampling HEPA Filter 3	497	108	1150	67.4	1.39	--	2.72E+00	<-1.74E-01	<3.02E-01	<9.54E-02	<9.53E-01
Condensate A	1470	<9.80	168	<9.80	<9.80	181	<2.46E-02	<-1.71E-01	<-4.28E-02	<0.00E+00	<1.25E-01
Condensate B	1640	<9.82	159	<9.82	<9.82	269	<2.03E-01	<2.24E-02	<2.26E-02	<-4.86E-02	4.86E-01
Condensate C	1610	<9.86	133	<9.86	<9.86	346	<9.46E-02	<0.00E+00	<0.00E+00	<-8.04E-02	<1.07E-01
Demister A	4520	12.4	280	<9.77	<9.77	270	<1.73E-01	<-2.45E-02	<4.55E-02	<1.54E-01	4.62E-01
Demister B	2710	<9.84	142	<9.84	<9.84	394	<-4.61E-02	<-9.14E-02	<-4.69E-02	<1.07E-01	<1.07E-01
Primary SBS Sump	1180	<9.92	103	<9.92	<9.92	310	<1.62E-01	<0.00E+00	<0.00E+00	<2.67E-02	<1.07E-01
Pre-filter Fluid	25100	<16.1	485	<9.34	<9.34	336	<7.51E-02	<-1.10E-02	<3.34E-02	<9.95E-02	5.02E-01

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