PNNL-30189, Rev. 0
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Vitrification of Hanford Tank 241-AP-107 with Recycled Condensate

September 2020

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Pacific Northwest National Laboratory
Richland, Washington 99354

Summary

During the vitrification of nuclear waste at the Hanford Waste Treatment and Immobilization Plant (WTP) – the primary 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, a test apparatus was designed to mimic the EMF evaporator and used to concentrate a volume of condensate that had been previously produced during the vitrification of Hanford tank 241-AP-107 (referred to herein as AP-107) waste in a continuous laboratory-scale melter (CLSM). The test apparatus successfully concentrated the AP-107 condensate by a factor of 10 while retaining over 90 % of the technetium-99 (^{99}Tc), Cs, and I inventory. A second portion of AP-107 waste was retrieved by Washington River Protection Solutions, LLC, given to Pacific Northwest National Laboratory, and combined with the AP-107 condensate concentrate after undergoing solids filtration and cesium removal by ion exchange. This combination served to approximate the recycling action to be performed at the WTP.

After the addition of glass-forming chemicals (GFCs), the combined AP-107 waste and AP-107 condensate concentrate were processed in the CLSM to produce a glass, called AP-107-1R, that was designed to satisfy the WTP baseline requirements (Kim et al. 2012). During the 8.87 hours of processing, 7.27 kg of AP-107-1R glass were produced for an average glass production rate of 1739 kg m^{-2} d^{-1}. Compared to the previous run in the CLSM without recycled condensate, the run with the recycle had a greater average glass production rate, but the rate was within the potential range of variability when processing melter feeds with similar composition in the CLSM.

The glass produced from the AP-107 recycle run in the CLSM was within 10 % of the target AP-107-1R glass composition with respect to the primary glass components. Analysis of the minor component impurities revealed that their content in the glass product had approached their nominal target after 2 turnovers of the glass inventory in the CLSM while the activity of the minor radionuclides was retained in the glass product.

The ^{99}Tc and total cesium content in the combined AP-107 waste and recycled condensate were maintained at concentrations expected to be experienced at the WTP. During processing in the CLSM, at discrete sampling time periods, the target ^{99}Tc/Cs mass ratio in the glass formulation varied from 0.9 to 62.9. Across this range, the Cs retention in the glass ranged from 53 to 60 %, while the retention from the entire runtime totaled 68 %, values which align with Cs retention in other scaled melter systems while processing LAW melter feeds at ^{99}Tc/Cs mass ratios ranging from 1 to 100. The ^{99}Tc retention in the glass ranged from 22 to 32 %, primarily due to the cold-cap coverage on the glass melt surface, the area covered by reacting melter feed, varying from ~80 % to ~95 % during processing, demonstrating greater volatility of ^{99}Tc from the glass while more surface was exposed, as expected based on previous ^{99}Tc volatility studies.

Acknowledgements

The authors thank Jackie Ranger and the team at the Southwest Research Institute for performing the chemical analysis of all radioactive and non-radioactive products from the continuous laboratory-scale melter system. We thank Renee Russell for reviewing all the data, calculations, and figures associated with this technical report. The authors gratefully acknowledge the financial support for testing of the continuous laboratory-scale melter system provided by Washington River Protection Solutions, LLC and project direction provided by Kristin Colosi.

Acronyms and Abbreviations

APEL	Applied Process Engineering Laboratory
CA	contamination area
CLSM	continuous laboratory-scale melter
CST	crystalline silicotitanate
CUA	The Catholic University of America
DF	decontamination factor
DFLAW	direct-feed low-activity waste
DIW	deionized water
DOE-ORP	U.S. Department of Energy Office of River Protection
EMF	Effluent Management Facility
EQL	estimated quantitation limit
ETF	Effluent Treatment Facility
GFCs	glass-forming chemicals
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
ICP-OES	inductively coupled plasma optical emission spectroscopy
LAW	low-activity waste
MDL	method detection limit
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
RPM	revolutions per minute
SBS	submerged-bed scrubber
sccm	standard cubic centimeters per minute
SwRI	Southwest Research Institute
TC	thermocouple port
TOC	total organic carbon

TSCR	Tank Side Cesium Removal
VSL	Vitreous State Laboratory
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

The primary mission of the U.S. Department of Energy-Office of River Protection (DOE-ORP) is to retrieve and process approximately 56 million gallons of radioactive waste from 177 underground tanks located on the Hanford site. 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 the waste. Currently, the first phase of the planned WTP startup and operation, called direct-feed low-activity waste (DFLAW), involves directly feeding only the liquid supernatant portion of the waste to electric melters in the WTP low-activity waste (LAW) vitrification 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 (Bechtel 2015), WRPS is designing a Tank Side Cesium Removal (TSCR) system for suspended solids and cesium (Cs/^{137}Cs) removal from the LAW. 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 are driven into the offgas treatment system, a large fraction of which are captured as condensate. This offgas condensate is then concentrated by evaporation in the Effluent Management Facility (EMF) and will then be recycled back to the LAW vitrification 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 but may 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 portions of retrievable LAW 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 for the removal of solids (Geeting et al. 2018a), it underwent ion exchange for the removal of 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 (Rovira et al. 2018). 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 this 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 to mimic the recycle action of the EMF and LAW vitrification facility. The purpose of the test described in this report was to produce and report the results from the vitrification of the recycled AP-107 melter feed that can be used as input to future evaluations of impacts from offgas condensate recycle on the LAW vitrification facility, dynamic melter processing, and glass chemistry. It also served to provide process performance data and sample chemical analysis that could contribute towards confirming the fraction of waste components assumed to partition into the offgas system. Comparative analysis was also performed on the processing of AP-107 without and with recycle in the CLSM and with the processing of AP-107 with recycle in a similar scaled melter system (Matlack et al. 2018). Ultimately, the CLSM has demonstrated the ability to support future WTP programmatic needs regarding cold-cap behavior, glass processing operations, and an understanding of 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 Test Objectives and Success Criteria

Testing was conducted to fulfill the objectives listed in Table 3.1 that were deemed primary to the planned purpose of assessing the effect of offgas recycle on the dynamic melter process and glass chemistry.

Table 3.1. Test Objectives and Success Criteria

Test Objective	Success Criteria	Results	Results Reference
Collect glass samples.	Glass samples have been collected during the processing of the melter feed.	A total of 7.27 kg of glass were produced during the CLSM run, which were collected and catalogued according to their pour times.	Section 5.1
Collect samples of melter offgas semi-volatiles with high-efficiency particulate air (HEPA) filters.	Offgas samples have been taken through the sampling loop while the cold cap is at steady-state (mass basis) during feeding.	Two offgas samples were collected during feeding while the cold-cap conditions were judged to be steady.	Section 5.2
Collect melter offgas condensate.	Offgas condensate from the entire melter run has been collected.	Collected all offgas condensate from the CLSM run.	Section 5.3
Record operational data to allow comparative studies to be performed between the current and previous CLSM vitrification tests including the consideration of the condensate recycle components (primarily Tc and halides) on process performance.	Operational data have been recorded to allow comparative studies of current and previous AP-107 vitrification runs at RPL (Dixon et al.2019)	All processing information collected during the AP-107 recycle run are reported graphically in this test report, key characteristics were compared with the previous AP-107 vitrification run and discussed.	Sections 5.2 and 6.0
Analyze chemical composition of melter feed, glass product, offgas condensate, and HEPA filter element samples.	Analyses of all samples are completed.	Aliquots of selected portions of melter feed, glass product, offgas condensate, other offgas liquids, and HEPA filters were sent for analysis.	Section 5.3

4.0 Experimental

This section describes the experimental process used to concentrate the AP-107 condensate and prepare the subsequent melter feed with recycle. The CLSM system used to vitrify the melter feed and the analyses of the resultant samples are also specified.

4.1 EMF Evaporator Testing

This section describes the 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 AP-107 offgas condensate from the CLSM melter.

4.1.1 Evaporator Feed Preparation

Bench-scale EMF testing with approximately 3.4 liters of AP-107 offgas condensate generated from the CLSM system (Dixon et al. 2019) was conducted to determine component partitioning between the evaporator condensate and evaporator concentrate. 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 4-μm polymer filter paper and collected for analysis by inductively coupled plasma optical emission spectroscopy (ICP-OES) and ion chromatography (IC). The filtered AP-107 offgas condensate was pH adjusted from 1.34 to 12.15 using 160 mL of 5 M NaOH to finish its preparation as evaporator feed. The evaporator feed density was measured to be 1.021 g mL^{-1}.

4.1.2 Test Conditions

A schematic of the EMF evaporator test apparatus used for this testing is shown in Figure 4.1. The apparatus was constructed almost entirely from glass. All testing was conducted inside a radiological contamination area (CA) fume hood in the RPL at PNNL. The reaction vessel was a cylindrical flat-bottom 1-L glass beaker. The evaporator feed in the vessel was heated using a fabric heating mantle and continuously stirred using a Teflon-coated magnetic stir bar on a stir plate. The heating mantle was controlled with a Model 270 temperature controller (J-KEM Scientific, Inc., St. Louis, Missouri). Temperatures within the reaction vessel were recorded electronically at 1-minute intervals.

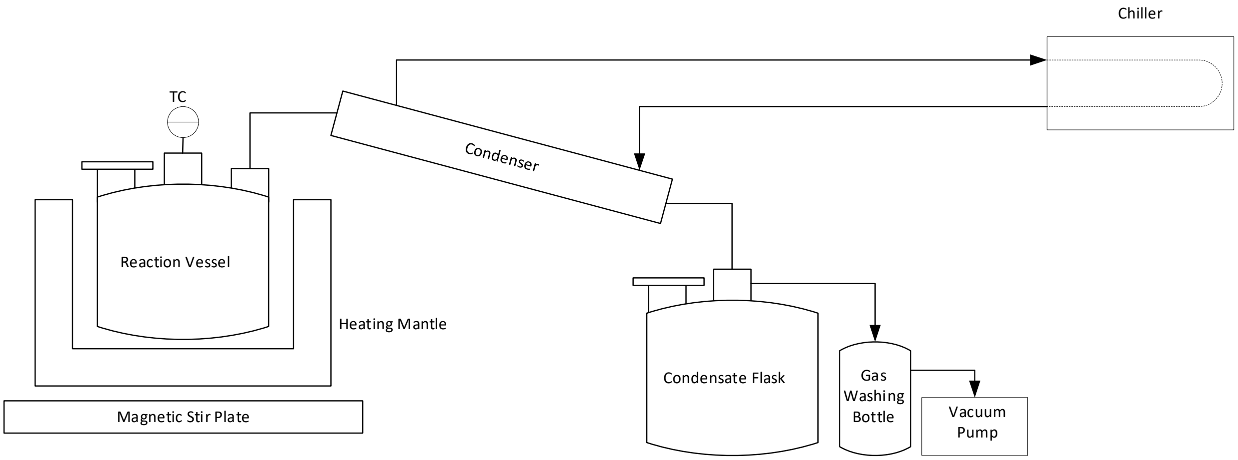

Figure 4.1. EMF evaporator test apparatus system schematic.

The evaporator feed temperature averaged 64 ± 3 °C during testing. This value is significantly higher than the prototypic EMF evaporator design of 50 °C due to the lower vacuum utilized in the test apparatus system. The EMF evaporator will operate with a targeted vacuum pressure of 28 in-Hg as compared to the test apparatus with test averages around 21 in-Hg; the greater absolute-pressure resulting in a higher boiling temperature. The effect of this temperature change on the partitioning of constituents to the evaporator condensate or evaporator concentrate is expected to be minimal and likely limited to the more volatile organic compounds. 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 °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 KNF vacuum pump, model N 86 KTP. Figure 4.2 shows a photograph of the EMF evaporator test apparatus components as they were arranged in a CA fume hood. Before running with the evaporator feed, water runs were performed to ensure that all the equipment and instruments were operating correctly.

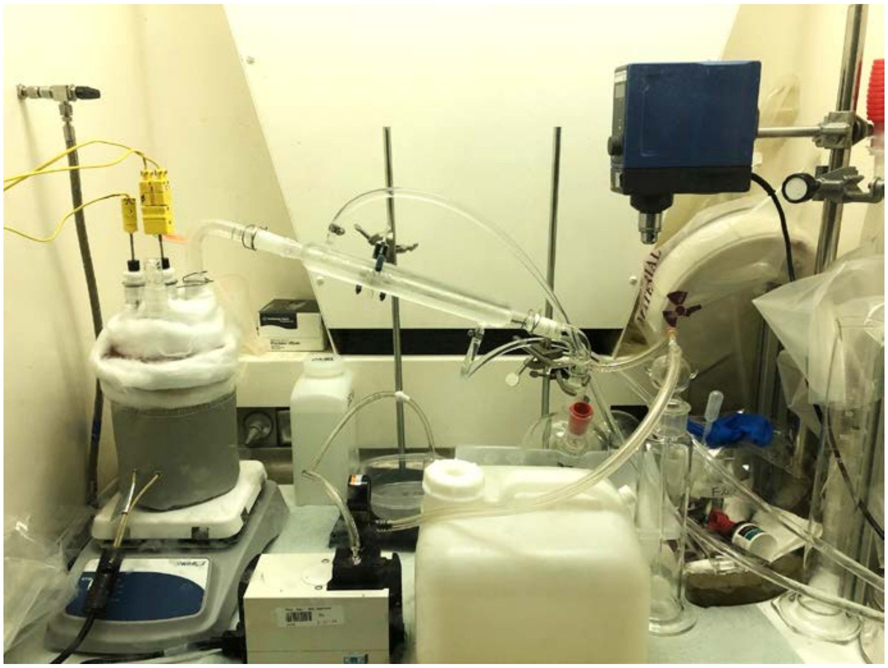

Figure 4.2. EMF evaporator test apparatus as arranged in a CA fume hood.

Initially, 1 L of evaporator feed was loaded into the reaction vessel. In order 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 3.6 kg of evaporator feed was evaporated down to an equivalent 339 g. At the end of testing, density for the evaporator condensate and evaporator concentrate were measured to be 1.03 and 1.15 g mL^{-1}, respectively. Solids were found in the evaporator concentrate and were filtered using a 0.45-µm polyethersulfone (PES) filter and collected for analysis by ICP-OES, IC, and inductively coupled plasma mass spectrometry (ICP-MS) for total cesium.

4.1.3 Evaporator Operation and Contents Compositions

The EMF evaporator test apparatus was operated under vacuum at around 21 in-Hg and boiled at approximately 64 °C during testing. The boil-off rate of the evaporator condensate was approximately 3 mL min^{-1}. The variance in pressure, shown in Figure 4.3, was minor (4 in-Hg) over the duration of the test; system pressure ranged from 19 to 23 in-Hg. Likewise, minor fluctuations in temperature were observed. After initial heating, temperature readings ranged from 57.7 to 67.6 °C.

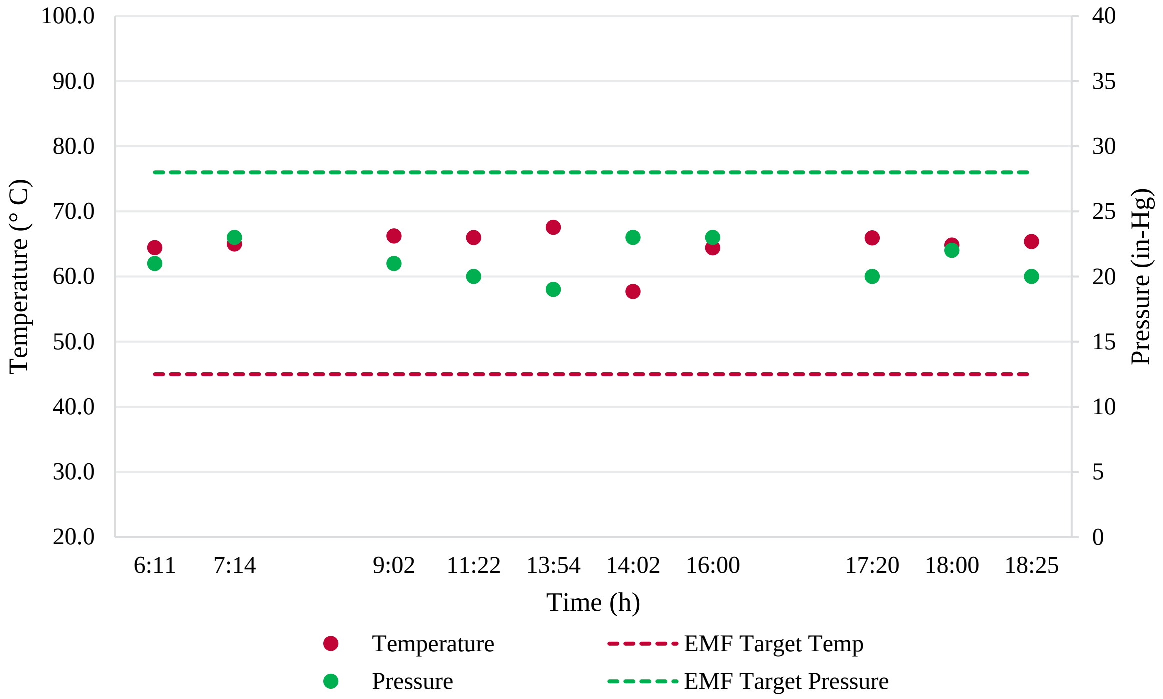

Figure 4.3. Temperature and pressure of the EMF evaporator test apparatus during operation.

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 did not interrupt the test.

The test apparatus was temporarily shut down ten times 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 contained insoluble solids that had settled to the bottom of the reaction vessel. The solids were filtered using a 0.45-µm PES filter so they could be analyzed. The resulting filtered evaporator concetrate was pH tested to be 12. The final measured concentration factor was based on mass and was found to be 10.3 g g^{-1} (evaporator feed-to-concentrate ratio).

The compositions of the evaporator feed, condensate, concentrate, and collected concentrate solids were evaluated to understand component mass fractionations. Analysis results of the test are shown in Table 4.1. Component results shown in brackets in Table 4.1 indicate that the analytical result was less than the estimated quantitation limit (EQL) but greater than or equal to the method detection limit (MDL) and the associated analytical uncertainty could be higher than ±15%. The fractionation result was placed in brackets, where it was calculated with one or more bracketed analytical value(s), to highlight the higher uncertainty.

Table 4.1. Evaporator Feed, Condensate, Concentrate, and Concentrate Solids Compositions

Analysis Method	Component	Evaporator Feed (mg kg^{-1})	Condensate (mg kg^{-1})	Concentrate (mg kg^{-1})	Concentrate Solids (mg kg^{-1})	Fraction in Condensate (%)	Fraction in Concentrate (%)	Fraction in Solids (%)
ICP-MS	Total Cs	10.8	[0.05]	103	226.0	--	92%	2%
	Total I	0.85	[0.05]	8.27	--	--	94%	--
	^{99}Tc	1.7	[0.001]	17.0	--	0.06%	95%	--
IC-anions	Br^-	[4.97]	[5.02]	[4.49]	36.8	--	--	--
	Cl^-	536	[5.02]	5500	8290	--	99%	1%
	F^-	24.4	[5.02]	228	441	--	91%	2%
	NO_3^-	2335	[5.02]	23000	35100	--	95%	1%
	NO_2^-	[4.97]	[5.02]	11.5	39.2	--	--	--
	PO_4^{3-}	[4.97]	[5.02]	[4.49]	77.2	--	--	--
	SO_4^{2-}	125	[5.02]	1240	1835	--	96%	1%
ICP-OES	Al	11.2	[5.02]	53.7	9200	--	46%	72%
	B	180	14.3	1680	2000	6.96%	90%	1%
	Ca	8.24	[2.51]	[3.62]	16200	--	4%	173%
	Cr	2.95	[0.25]	0.863	7410	--	3%	221%
	Fe	[5.91]	[5.02]	[4.46]	16400	--	--	244%
	Li	4.56	[0.5]	44.6	210	--	95%	4%
	Mo	[0.562]	[0.5]	5.84	20	--	101%	3%
	Ni	[0.378]	[0.25]	2.59	161	--	66%	37%
	K	38.5	[7.53]	394	827	--	99%	2%
	Si	33.8	14.6	97.1	33500	--	28%	87%
	Na	4820	[9.13]	46600	86000	--	94%	2%
	S	38.2	[3.76]	421	521	--	107%	1%
	Sn	[0.99]	[1.0]	[0.89]	[8.3]	--	9%	1%
	Ti	0.509	[0.25]	[0.22]	1420	--	4%	245%
	W	[1.24]	[1.25]	3.31	324	--	26%	23%
	Zn	25.2	[0.25]	25	41000	--	10%	143%

Bracketed values indicate the associated sample results were less than the EQL but greater than or equal to the MDL.
“--” = not applicable; value not reported, or fractionation cannot be calculated with a less-than value.

The component fractionations were calculated as ratios of the total component measured in the evaporator feed and the total component collected in the evaporator effluent according to Eq. (4.1),

$$\frac{C_{Ea} \times M_E}{C_{Fa} \times M_F} = F_{Ea} \tag{4.1}$$

where:

C_{Ea} = concentration of component *a* in the evaporator effluent (condensate, concentrate, concentrate solids)
M_E = mass of evaporator effluent (3149 g for condensate; 339.4 g for concentrate; 3.2 g for concentrate solids)
C_{Fa} = concentration of component *a* in the evaporator feed
M_F = mass of evaporator feed (3618 g)
F_{Ea} = fraction of component *a* in the evaporator effluent (condensate, concentrate, concentrate solids)

The concentrations in the evaporator concentrate show a nominal 10× increase compared to the evaporator feed. Recoveries greater than 90% of Cs, I, and ^{99}Tc along with all measured anions were reported in the evaporator concentrate. As expected, no (or extremely low) recoveries of most components were measured in the evaporator condensate. The only component detected by ICP-OES was boron at 14.3 mg kg^{-1}. In contrast to previous test results performed by the Vitreous State Laboratory (VSL) of The Catholic University of America (CUA) (Matlack et al. 2018), no measurable I was found in the evaporator condensate, indicating it is not volatile under the conditions in the EMF evaporator test apparatus. The solids recovered from the evaporator concentrate solids were determined to be primarily Ca, Cr, Fe, Ti, Si, and Zn.

4.2 Melter Feed Preparation

The AP-107 waste received after solids and cesium removal had a density of 1.27 kg L^{-1} and was split into two portions. The first portion, totaling 4.58 kg was combined with 0.34 kg of evaporator concentrate, prepared as described in Section 4.1, for a volume ratio of 7.5 % evaporator concentrate to total solution. By combining the AP-107 waste and concentrated AP-107 offgas condensate solutions, one round of the process of recycling the condensate from the EMF to the waste incoming to the LAW vitrification facility was replicated. The combined AP-107 waste plus concentrate solution, referred to as AP-107 waste recycle, was sampled for analysis by inductively coupled plasma atomic emission spectroscopy (ICP-AES), IC, ICP-MS, and determination of total organic carbon (TOC). The analytical results for the solution are listed in Table 4.2 and these results 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 GFCs addition needed to achieve this composition. This process of analyzing the combined waste stream and using a model to determine the GFCs addition is the same process currently planned for melter feed preparation at the WTP (Bernards et al. 2017).

The GFCs minerals and the mass of each addition are given in Table 4.3. These GFCs were added to the AP-107 waste recycle to form the ‘AP-107 recycle melter feed’ with a target glass yield of 699 g-glass L-feed^{-1}. The composition of the glass to be produced from the AP-107 recycle melter feed, referred to as AP-107-1R, is shown in Table 4.4 along with the composition of 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.

Table 4.2. AP-107 Waste Recycle Composition

Component	Analysis Method	Average Concentration (mg kg^{-1})
^{99}Tc	ICP-MS	5.30
Total I		2.16
Total Cs		6.18
Cl	IC	2465
F		304
NO_2		12700
NO_3		20850
Al	ICP-AES	7015
B		131
Ca		12.2
Cd		3.39
Co		0.324
Cr		349
Cu		0.346
Fe		10.8
K		2220
Li		2.30
Mo		32.5
Na		91600
Ni		15.1
P		623
Pb		1.44
S		1385
Si		55.4
Sn		6.20
Ti		0.381
V		0.411
W		53.5
Zn		13.0
Zr		1.63
TOC	TOC	1490

Table 4.3. GFCs Masses Added to Each Portion of AP-107 Waste

GFCs	Mass Added to AP-107 Waste Recycle (g)	Mass Added to AP-107 Waste Simulant Recycle (g)	Mineral Source
Kyanite	271.71	356.68	Kyanite Mining Corporation
Boric Acid	653.39	857.72	Alfa Aesar
Wollastonite	365.20	479.40	NYCO Mineral
Iron Oxide	193.09	253.47	JT Baker
Lithium Carbonate	139.90	183.66	Foote Mineral Company
Olivine	107.81	141.52	Unimin Corporation
Silica	1280.44	1680.86	Sil-Co-Sil 75
Rutile	51.49	67.59	Chemalloy
Zinc Oxide	129.80	170.39	Noah
Zircon	165.25	216.93	Prince Minerals
Sucrose	47.02	61.72	C+H Sugar
Total	3405.10	4469.94	

Table 4.4. Glass Compositions Based on AP-107 Waste

Component	AP-107-1R (wt%)	AP-107[a] (wt%)	AP107WDFL[b] (wt%)
Al_2O_3	6.13	6.12	6.10
B_2O_3	9.95	9.95	10.00
CaO	4.53	3.69	3.94
Cl	0.18	0.15	0.42
Cr_2O_3	0.07	0.08	0.08
F	0.03	0.00	0.04
Fe_2O_3	5.52	5.52	5.50
K_2O	0.36	0.47	0.38
Li_2O	1.52	0.50	0.89
MgO	1.49	1.49	1.48
Na_2O	16.34	17.49	17.20
NiO	0.01	0.01	0.00
P_2O_5	0.19	0.15	0.13
PbO	0.00	0.00	0.00
SO_3	0.39	0.37	0.44
SiO_2	45.36	46.08	45.50
TiO_2	1.40	1.40	1.40
ZnO	3.51	3.51	3.50
ZrO_2	3.02	3.01	3.00
Sum	100.00	99.99	100.00

(a) Dixon et al. (2019)
(b) Matlack et al. (2018)

The second portion of AP-107 waste after solids and cesium removal totaled 6.01 kg. To simulate the recycle process that was replicated with the first portion of AP-107 waste, a simulant of the evaporator concentrate was designed based on the composition, shown in Table 4.1, but with a target cesium concentration as low as possible. To create the evaporator concentrate simulant, the components listed in Table 4.5 were added to 500 mL of deionized water (DIW) and the resultant solution was shaken in batch contact with crystalline silicotitanate (CST) ion exchange media for approximately 120 hours. The density of the evaporator concentrate simulant was 1.09 g mL^{-1} and 0.42 kg were filtered from the CST media. Iodine was then added to the solution via 0.00448 g of KI and ^{99}Tc was added via 0.360 mL of a solution with a concentration of 20 mg-^{99}Tc mL^{-1}. The evaporator concentrate simulant was then added to a second portion of AP-107 waste to form the ‘AP-107 waste simulant recycle melter feed’ with a volume ratio of 7.5 %. The GFCs that were added to the AP-107 waste simulant recycle to form the AP-107 simulant recycle melter feed are shown in Table 4.3.

Table 4.5. Evaporator Concentrate Simulant Components

Chemical Additions	Mass (g)
$Al(NO_3)_3 \cdot 9H_2O$	0.43359
$Ca(NO_3)_2 \cdot 4H_2O$	0.01277
H_3BO_3	5.54557
SiO_2	0.12080
NaOH	34.41375
KOH	0.32611
$Na_3PO_4 \cdot 12H_2O$	0.08812
NaCl	5.23124
NaF	0.29277
Na_2SO_4	1.08271
$Zn(NO_3)_2 \cdot 6H_2O$	0.06621
$NaNO_2$	0.01027
$NaNO_3$	17.94380
$LiOH \cdot H_2O$	0.15503

4.3 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 area, and it details the operating conditions for system performance.

4.3.1 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 4.4.

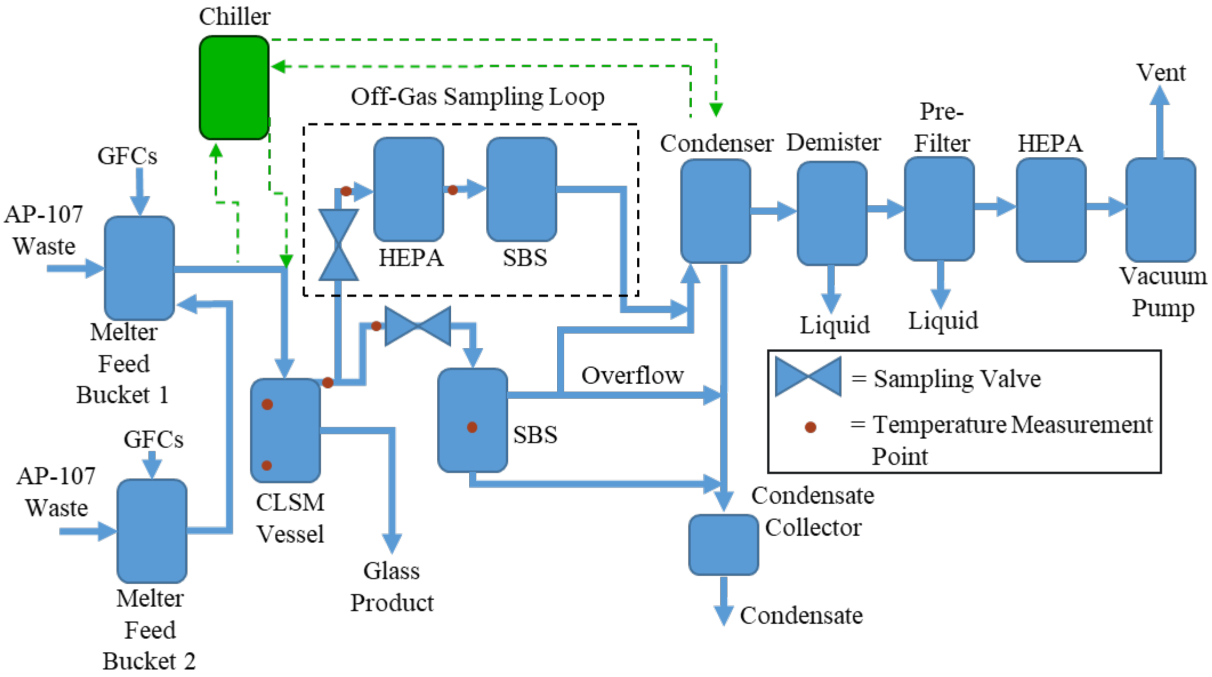

Figure 4.4. Simplified flow diagram of the CLSM system.

The AP-107 simulant recycle melter feed was placed in the 'Melter Feed Bucket 1' position and the AP-107 recycle 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 the action was 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 may result in feed line blockage.

The CLSM vessel was fabricated as an octagonal cross-sectional design using Inconel 690 plate. The vessel was 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 4.5, 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.

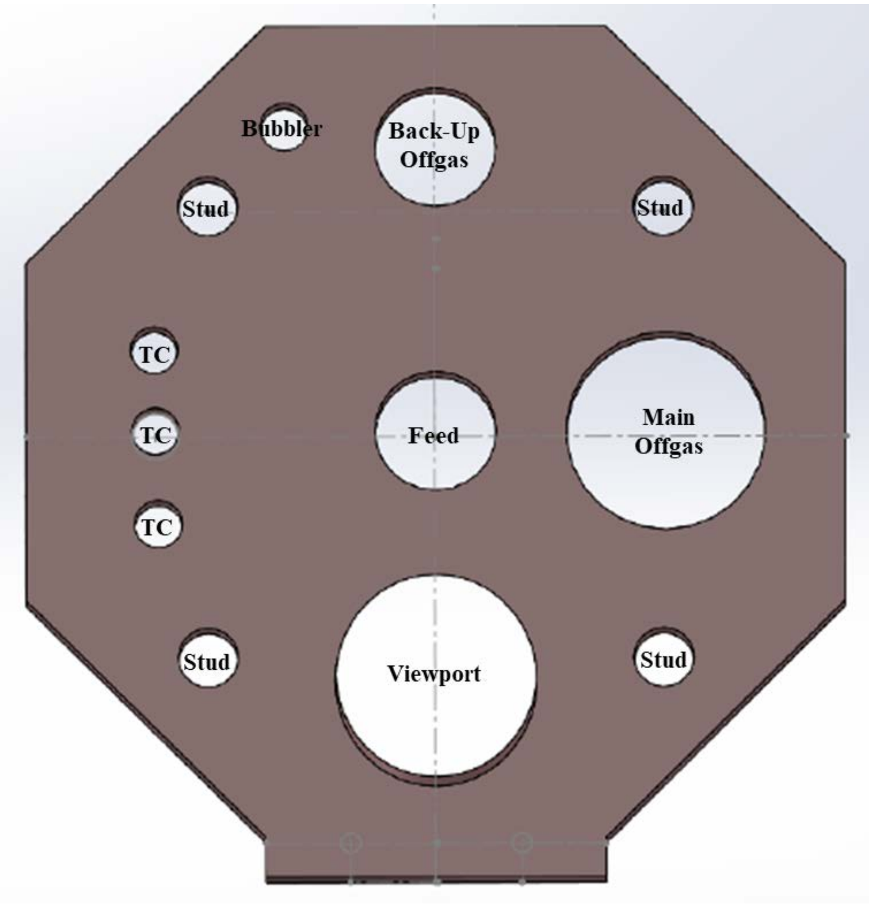

Figure 4.5. CLSM vessel lid and identified ports. The designation 'TC' stands for a thermocouple port.

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 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 banks of two sampling HEPA filters each. Each bank was available for discrete sampling evolutions. 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 with slip-stream sampling and ensured that the sample was representative. Offgas sampling durations were 10-30 minutes until the sampling HEPA filters became impassable.

The CLSM system consisted of commercially available as well as custom parts. In addition to the CLSM system described above (shown in Figure 4.4), 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. The CLSM system was assembled in an HCA fume hood in the RPL at PNNL. An image of the CLSM system layout in the fume hood in RPL is shown in Figure 4.6.

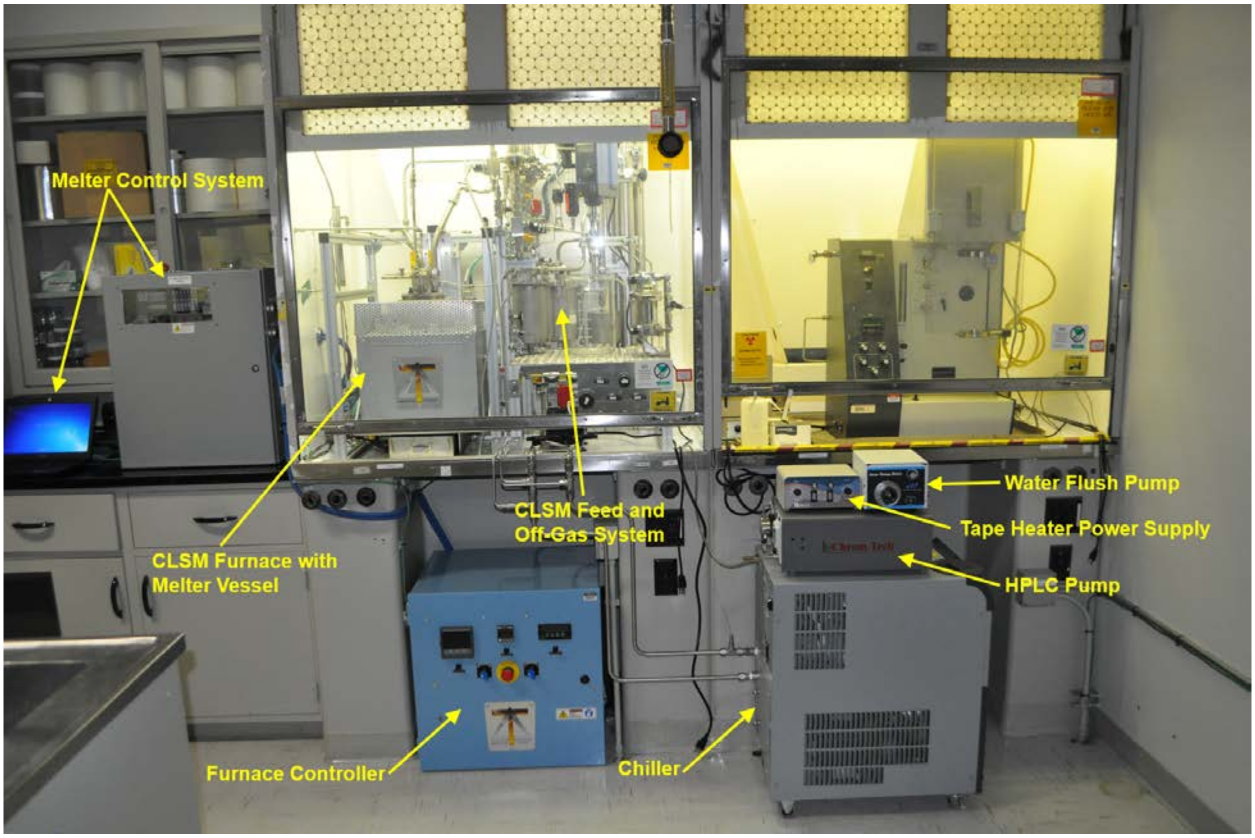

Figure 4.6. CLSM system layout in the RPL fume hood.

4.3.2 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 550-650 °C range and this could be confirmed through visual observation 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 other factors, including feed composition and concentration, which may vary between different melter feeds (Matlack et al. 2011). The target production rate ranges for the AP-107 recycle melter feeds in the CLSM, 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 DM10 melter system operated by VSL at CUA (Matlack et al. 2018), are listed in Table 4.6. 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 4.6. 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	450 – 650	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 and 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 4.3.1, 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.

4.4 Sample Analysis Methods

The mass of each component in the formation of melter feeds was totaled to determine the initial mass in each bucket; after the CLSM run, the remaining heels of melter feed in each bucket were combined and the total mass was recorded. The masses of all product streams were weighed after the run; these included the glass from each pour, the total condensate, the final sump 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 4.7.

Table 4.7. 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, Ba, Bi, B, Ca, Cd, Co, Cr, Cs, Cu, Fe, I, La, Li, K, Mg, Mn, Mo, Na, Ni, P, Pb, Si, Sr, S, Sn, ^{99}Tc, Ti, W, V, 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

5.0 Results

This section describes the operation of the CLSM in RPL on February 25, 2020 for the processing of the AP-107 simulant recycle melter feed and AP-107 recycle melter feed. The production and chemical analysis results are also detailed.

5.1 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 AP-107 recycle CLSM run then began charging the AP-107 simulant recycle melter feed at 12:34 PM on February 25, 2020, when the glass temperature had reached its desired range. The time, mass, and cumulative weight of each glass pour during the run are given in Table 5.1. Following the termination of feeding, the cold cap burned off (all remaining melter feed in the cold cap 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 AP-107 recycle CLSM run was 7.25 kg, corresponding to more than 3.5 turnovers of the CLSM glass inventory. Other notes about the performance during the run follow.

- The average plenum temperature during the first 4 hours of processing, 723 °C, was greater than the average for the entire run, see Section 5.2, due to the cold-cap coverage reaching only ~80 % based on the observation of the operational team. During this period, the bubbling gas was centered directly under the charging melter feed and this condition limited the cold-cap size.

- Around hour 4.50, the cold cap was observed to “roll over,” exposing the bottom of the cold cap to the plenum space. Following this action, vacuum was no longer able to be maintained in the CLSM vessel and glass began to pour. As a result, feeding was stopped, and attempts were made to return the target vacuum conditions in the CLSM vessel. After several minutes the vacuum was restored and the glass pouring stopped. In total 1.8 kg of glass were poured from the CLSM vessel, corresponding with the glass pour at hour 4.50 in Table 5.1, leaving an estimate of 250 g of glass inventory remaining in the vessel.

- From hour 4.63 to 6.56, the CLSM system remained in idling conditions while the AP-107 recycle melter feed was pumped from Melter Feed Bucket 2 (see Figure 4.4) into the heel of the AP-107 simulant recycle melter feed in Melter Feed Bucket 1.

- At hour 6.56, feeding was resumed, but glass was not poured from the CLSM vessel until the operational team determined that the glass level in the vessel had returned to its target of ~6.4 cm from the bottom of the vessel, indicating the glass inventory had been restored to its desired 2.0 kg capacity.

- Around hour 9.40, a portion of the cold cap was observed to roll over, but no vacuum issues arose after this event.

Table 5.1. Timing and Mass of Glass Pours During the AP-107 Recycle CLSM Run

Pour Time (h)	Glass Weight (g)	Cumulative Glass Weight (g)
0.55	412.00	412.00
0.97	378.57	790.57
1.43	312.91	1103.48
1.62	332.34	1435.82
1.90	351.35	1787.17
2.28	291.73	2078.90
2.67	311.14	2390.04
2.67	217.95	2607.99
3.06	383.66	2991.65
3.44	217.15	3208.80
3.74	267.19	3475.99
4.10	298.73	3774.72
4.50	1801.31	5576.03
8.89	331.86	5907.89
9.42	360.65	6268.54
9.89	255.44	6523.98
9.89	143.57	6667.55
10.29	425.86	7093.41
10.62	373.21	7466.62
10.93	1772.38	9239.00

5.2 Production Results

The production results from the AP-107 recycle CLSM run are given in Table 5.2, which include the total values of the feeding time (and low flow duration within the feeding time), operational downtime, mass of glass produced, mass of melter feed consumed, and average values for the glass production rate, feeding rate, bubbling flux rate (bubbling rate in L min^{-1}, scaled by the glass surface area of the CLSM vessel), glass temperature, and plenum temperature.

Table 5.2. CLSM Production Results During the AP-107 Recycle CLSM Run

Parameter	AP-107 Recycle CLSM Run
Test Date	February 25, 2020
Feeding Duration, h	8.87
Low Flow Duration, h	0.00
Downtime, h	1.93
Glass Produced, kg	7.27
Melter Feed Consumed, kg	16.57
Average Glass Production Rate, kg m^{-2} d^{-1}	1739
Average Feeding Rate, kg h^{-1}	1.87
Average Bubbling Flux Rate, L m^{-2} min^{-1}	70 (a)
Average Glass Temperature, °C	1138 (a)
Average Plenum Temperature, °C	700 (a)

(a) Values reported only during the times when the CLSM was at full glass inventory.

The processing values recorded during the AP-107 recycle CLSM run are displayed in Figure 5.1. These results include the glass and plenum temperatures, the effective glass production rate (based on the average glass production rates and the variable changes in the feeding pump rates), 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 5.1. Individual figures of each processing value can be found in Appendix A along with the temperatures at the start of the offgas system, at the sampling valve, and of the primary SBS (measurement locations shown on Figure 4.4); the operation rate of the feed pump, in revolutions per minute [RPM]; and the pressure of the feeding line, in pounds per square inch absolute [psia], at the inlet of the pump.

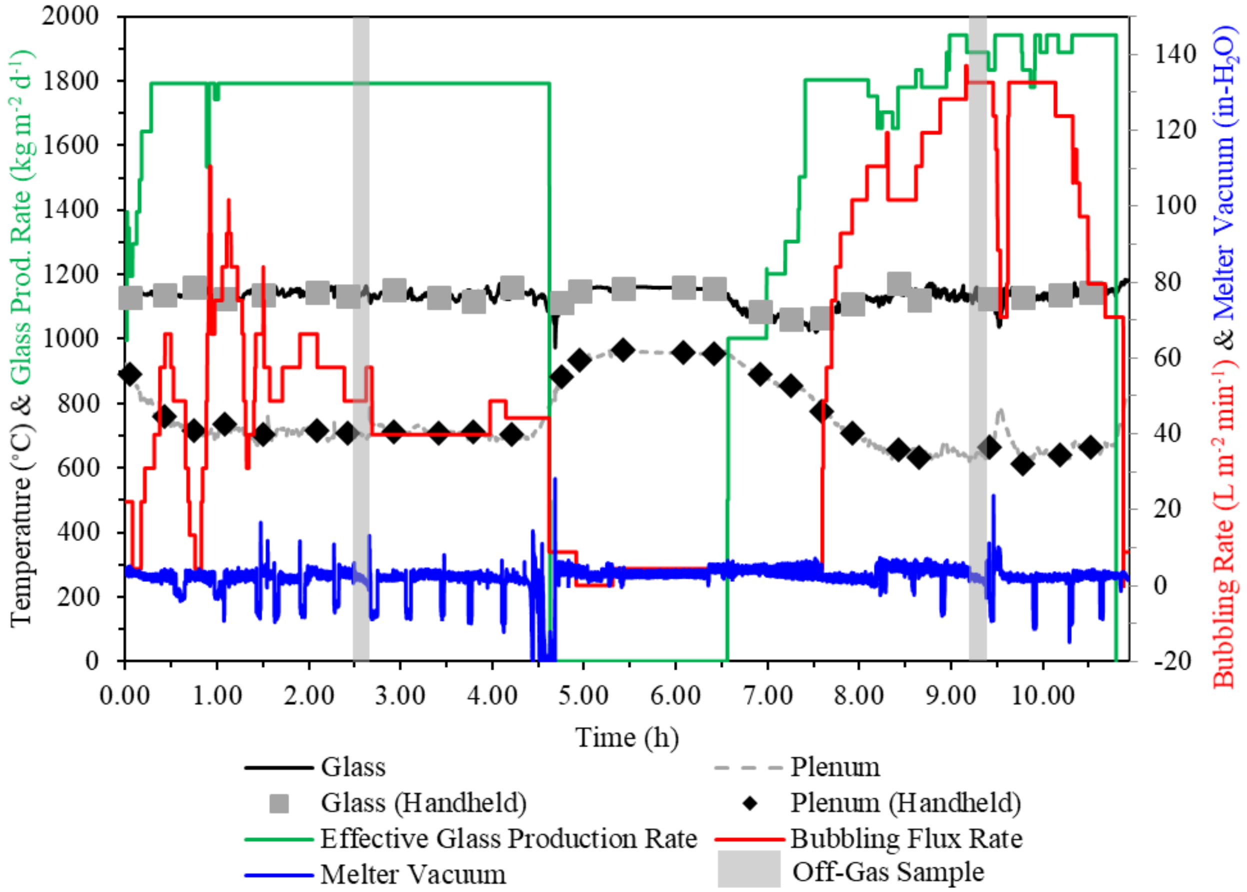

Figure 5.1. Processing values (glass and plenum temperatures, effective glass production rate, bubbling flux rate, and melter vacuum measurements) and offgas sample timing recorded during the AP-107 recycle CLSM run.

Two offgas samples were collected during the AP-107 recycle CLSM run. The timing and duration of the samplings are listed in Table 5.3 and the occurrence of each offgas sample in the timeline of the run is shown in relation to the processing values in Figure 5.1.

Table 5.3. Timing of Offgas Samples During the AP-107 Recycle CLSM Run

Sample Number	Offgas Sample Start on Test Run Timeline (h)	Offgas Sample End on Test Run Timeline (h)	Total Sampling Duration (min)
1	2.487	2.659	10.32
2	9.199	9.385	11.16

5.3 Sample Chemical Analysis

The samples selected for chemical analysis from the AP-107 recycle 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 four portions: 1 – all condensate produced prior to the idling period at hour 4.63; 2 – all condensate produced during the melter inventory buildup from hour 6.56 to 8.66; 3 – all condensate produced from hour 8.66 to the end of melter feed charging; and 4 – all condensate produced during the cold-cap burn off and flushing of the feeding system. Similarly, the liquid that accumulated in the demister was combined into two portions: 1 – all liquid produced prior to the idling period at hour 4.63; and 2 – all liquid produced after the idling period. The seal pot was tripped during the vacuum event at hour 4.50, so the liquid from the seal pot was collected. Liquid accumulated in the pre-filter housing during the run, so it was drained from the housing and collected. The sumps from both the primary SBS and sampling SBS were drained after the run. Aliquots of all the liquid portions described were sent for chemical analysis. For the collection of each offgas sample, two sampling HEPA filters were used and were thus combined and analyzed together.

6.0 Discussion

This section discusses the insights gained from the AP-107 recycle CLSM run and compares them to the previous AP-107 CLSM run (Dixon et al. 2019) in the RPL at PNNL.

6.1 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 produced from the CLSM and the offgas 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. (6.1):

$$\mathrm{DF}_i = \frac{\dot{m}_{i,feed}}{\dot{m}_{i,feed} - \dot{m}_{i,glass}} \tag{6.1}$$

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. (6.2):

$$R_i = \frac{\dot{m}_{i,glass}}{\dot{m}_{i,feed}} \tag{6.2}$$

The R_i value can be reported as a fraction or percentage (if Eq. (6.2) 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. (6.3):

$$\mathrm{Rec}_i = \frac{\dot{m}_{i,glass} + \dot{m}_{i,offgas}}{\dot{m}_{i,feed}} \tag{6.3}$$

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. (6.1), Eq. (6.2), and Eq. (6.3), 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.

6.2 Glass Composition

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

6.2.1 Primary Glass Components

Table 6.1 compares the average glass composition (determined by converting the analyzed component concentrations in each glass sample listed in Table A.1 in Appendix A to their associated oxides and averaging based on the mass of glass poured with each composition) from the AP-107 recycle CLSM run with the target AP-107-1R glass composition shown in Table 4.4. The analytical method used to detect the Cl concentrations in the glass and melter feed samples was insufficient to register concentrations above the detection limits. The Cl concentration values in the AP-107 recycle melter feed and AP-107 simulant recycle melter feed were estimated based on the analytical result of the Cl in the AP-107 waste recycle composition (see Table 4.2). The Cl concentration values in the glass product from the AP-107 recycle CLSM run were then estimated based on the concentration in the melter feeds and the average single-pass retention of Cl in the DM10 melter vessel calculated by VSL for a variety of LAW melter feed simulants (Matlack et al. 2012), an average of ~55 %. This Cl concentration estimation will be investigated, based on the amount of Cl in the analyzed offgas products, in Section 6.4. The percent difference between the component oxides in the glass composition from each run and those component oxides in the target glass composition are reported for all the primary glass components, those greater than 1.00 wt%.

Compositional trends for each component oxide in the analyzed glass product from the AP-107 recycle CLSM run, labeled as ‘Measured’, with respect to the amount of glass discharged are shown in Figure A.7 through Figure A.24 in Appendix A. Each figure displays the AP-107-1R glass target composition for the particular component oxide, labeled as ‘Glass Target’, with a grey block illustrating a ± 10 % window around the target composition and the actual composition for the component oxide expected based on the analyzed melter feed samples, labeled as ‘Feed Actual’.

The composition of all other primary glass components held relatively constant over the course of the AP-107 recycle CLSM run, fluctuating within the ± 10% glass target window based on minimal variation in the melter feed composition. The same consistency of primary glass components in the glass product was calculated in the previous AP-107 run in the CLSM (Dixon et al. 2019 and 2020b). Since a new CLSM vessel was fabricated for the AP-107 recycle CLSM run, there were no traces of glass in the vessel from previous runs and slight impurities were not observed in the initial glass composition as has been discussed from previous CLSM runs (Dixon et al. 2020a).

Table 6.1. Comparison of the AP-107 Recycle CLSM Run Glass Product with the Target AP-107-1R Glass Composition

Component	Target AP-107-1R Composition (wt%)	Measured AP-107-1R Composition (wt%)	% Difference Between Target and Measured (wt%)
Al_2O_3	6.13	5.99	-2.3
B_2O_3	9.95	9.28	-6.7
CaO	4.53	4.45	-1.8
Cl	0.18	0.18	--
Cr_2O_3	0.07	0.11	--
F	0.03	0.05	--
Fe_2O_3	5.52	5.45	-1.3
K_2O	0.36	0.39	--
Li_2O	1.52	1.43	-5.9
MgO	1.49	1.46	-2.0
Na_2O	16.34	17.14	4.9
NiO	0.01	0.02	--
P_2O_5	0.19	0.17	--
SO_3	0.39	0.43	--
SiO_2	45.36	45.65	0.6
TiO_2	1.40	1.42	1.4
ZnO	3.51	3.60	2.6
ZrO_2	3.02	2.79	-7.6
Sum	100.00	100.01	

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

6.2.2 Minor Glass Components

The measured Cr_2O_3 and NiO content in the AP-107 recycle CLSM run glass product (Figure A.22 and Figure A.24, respectively), as well as their glass target values and content measured in the melter feeds, are displayed together in Figure 6.1. A spike in the Cr_2O_3 and NiO content above the glass target and melter feed levels was observed at the start of the run, and after the ~2-hour idling period between the 3.85 and 4.19 kg glass discharged, 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, 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[1], an alloy with a relative composition of a minimum of 58.0 % Ni, a range of Cr content from 27.0 to 31.0 %, and 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 at high Cr levels and heated by electrodes that have high Ni levels, after idling periods (Matlack et al. 2010, 2011, and 2018).

Other minor glass components present in the AP-107-1R composition (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 or irregular volatility from the glass melt or in the cold cap. In particular, the Cl content in the AP-107-1R glass composition was estimated, as described in Section 6.2.1, and these assumptions may not fully capture the behavior of Cl. The behavior of S, K, P, Cl, and F in the CLSM offgas system will be discussed further in Section 6.4.

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

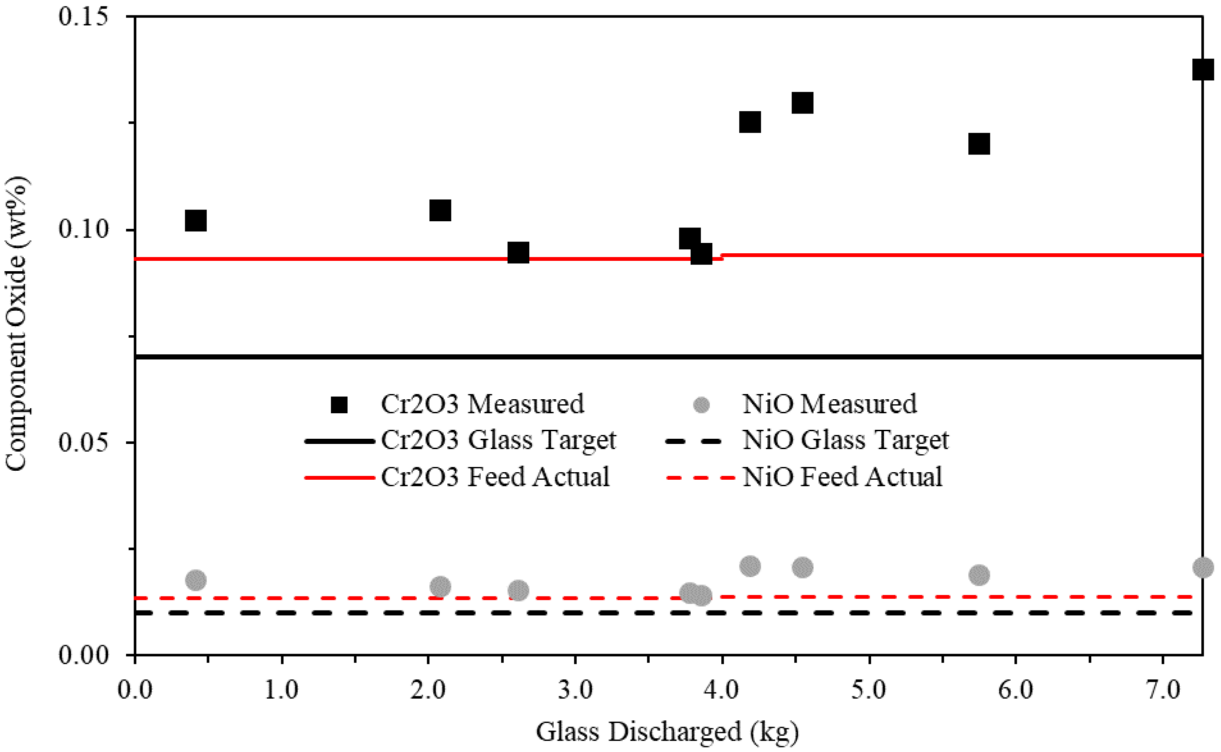

Figure 6.1. Cr_2O_3 and NiO content in the AP-107 recycle CLSM run glass product.

6.2.3 Minor Impurity Components

Several components were present as minor (300 ppm or less) impurities in the AP-107 recycle CLSM run melter feeds and glass product. The measured concentration of each component in the glass product, with respect to the mass of glass discharged, and their measured concentrations in the melter feed (if above the analytical detection limit) marked by the inset line, are shown in Figure 6.2 and Figure 6.3. 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 target values, based on their measured concentrations in the melter feeds, and then their concentrations plateaued at their target values. For these components, their concentrations reached their target 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 target value after 2 turnovers of the glass inventory.

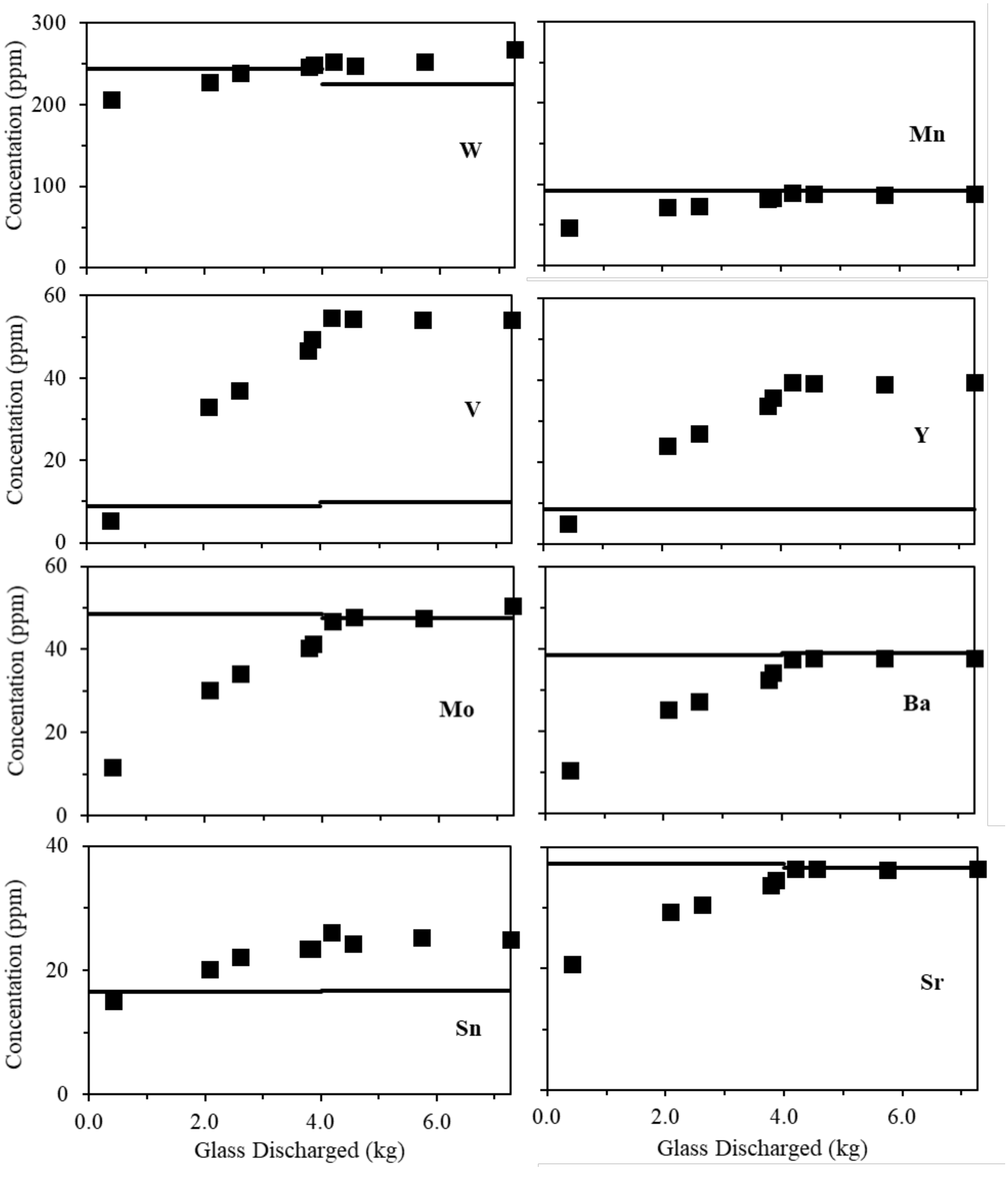

Figure 6.2. Concentrations of minor impurities (W, Mn, V, Y, Mo, Ba, Sn, and Sr) in the glass product from the AP-107 recycle CLSM run.

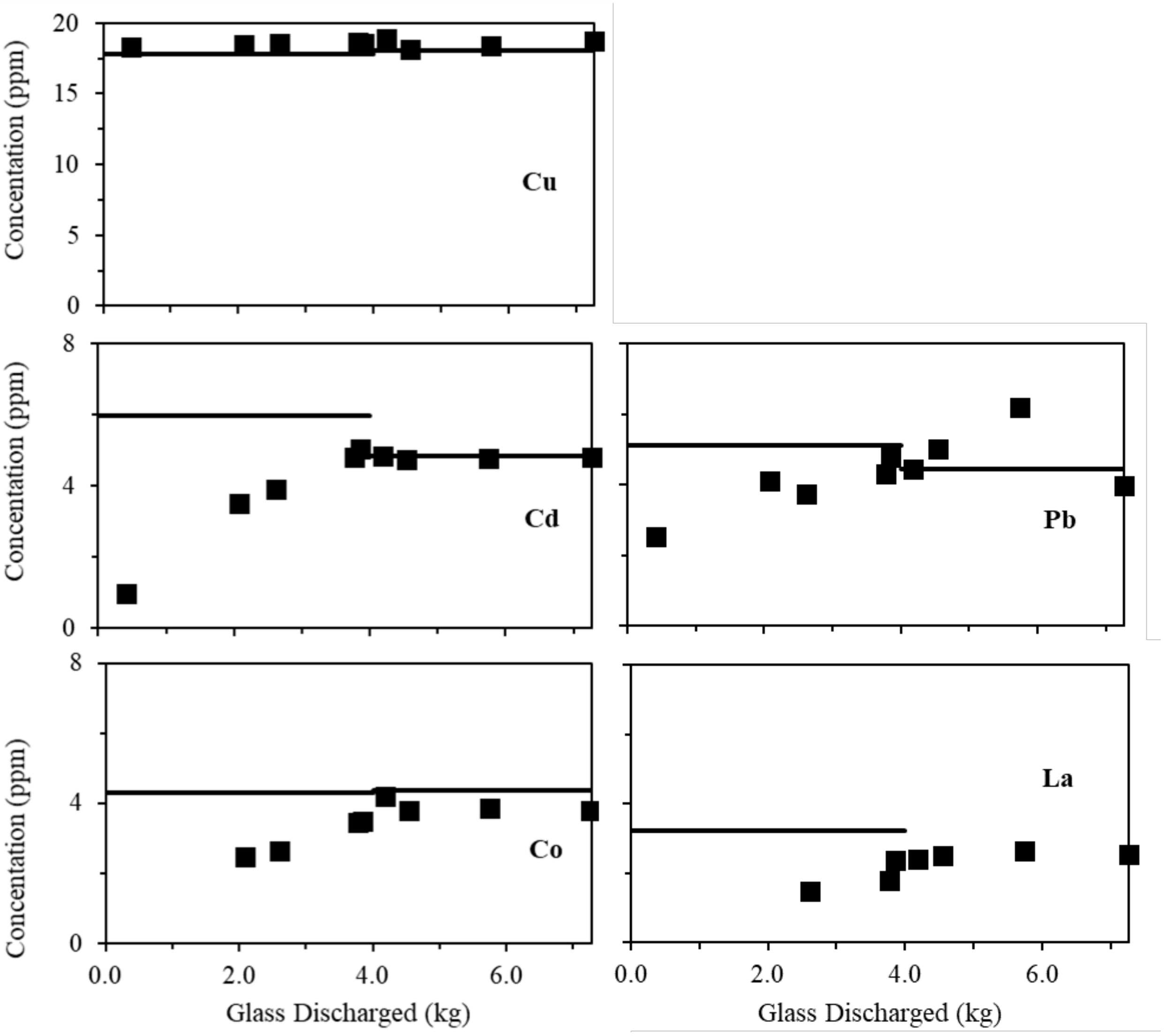

Figure 6.3. Concentrations of minor impurities (Cu, Cd, Pb, Co, and La) in the glass product from the AP-107 recycle CLSM run.

Several components were exceptions to the general impurity trend. The concentration of Cu in the initial glass inventory was equal to the target in the melter feed, so it did not increase, but remained at that level throughout the AP-107 recycle 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 target values in the melter feeds. These plateau values (~55 ppm for V, ~40 ppm for Y, and ~15 ppm for Sn), were similar to the concentrations of the components analyzed in the glass product from the previous AP-107 CLSM run (Dixon et al. 2019) and in the processing of AN-105 melter feed simulant in the CLSM (Dixon et al. 2020a). 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 in regard to the Cr_2O_3 and NiO content in the glass product in Section 6.2.2.

The measured activity of each analyzed radionuclide in the glass product, with respect to the mass of glass discharged, and their measured activity in the melter feed marked by the inset line, are shown in

Figure 6.4. 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.

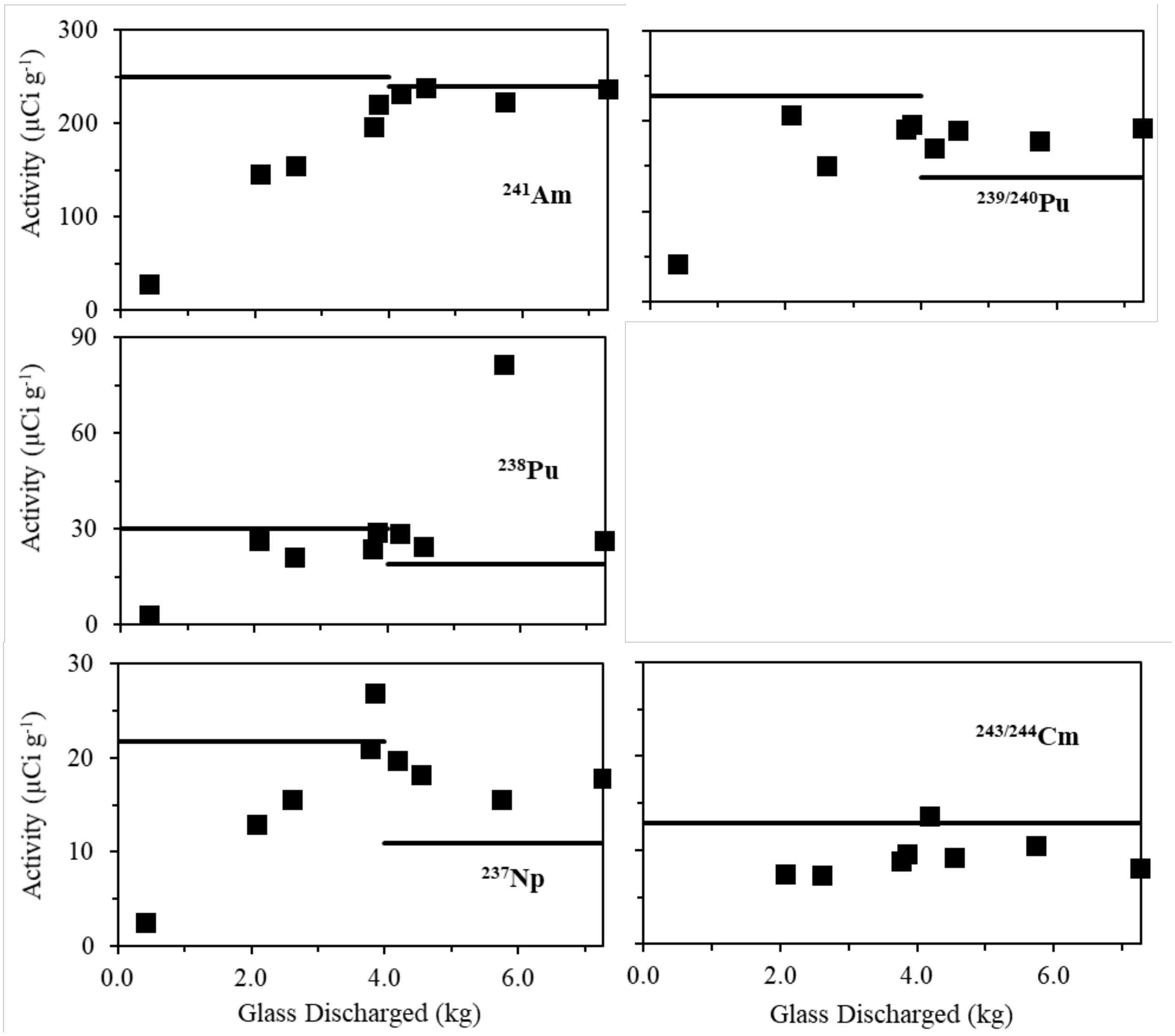

Figure 6.4. Activity of radionuclides (^{241}Am, $^{239/240}$Pu, ^{238}Pu, ^{237}Np, and $^{243/244}$Cm) in the glass product from the AP-107 recycle CLSM run.

6.3 DF, R, and Rec Calculations

For each component of the AP-107-1R glass composition captured in the CLSM glass product (Table 6.1), and the additives ^{99}Tc and Cs 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 fed of 16.57 kg) by the total runtime of 8.87 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 7.27 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 5.51 kg, demister liquid of 1.67 kg, primary SBS sump of 1.19 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 AP-107 recycle CLSM run are given in Table A.2 in Appendix A denoted in the 'Sample Duration' row as 'Run'. In addition, the mass flow tables contain the components' mass flow rates calculated exclusively during each offgas sample time period, denoted in the 'Sample Duration' row as '1' or '2' as related to the sample number, while the summation of each component's mass flow rates is listed as the 'Total'.

The total mass flow rate in the glass during the sampling time periods compared with the total mass flow rate in the glass during the total runtime can give an indication of the difference between the glass production rate when the system was believed to be at a steady cold-cap size and coverage and when the cold cap behavior included unsteady characteristics. During the first sample time period, the percent difference relative to the total mass flow rate in the glass at the total runtime was +2.8 %, and during the second sample time period, the percent difference was +8.0%. Both values were expectedly greater during the sampling time periods compared to the total runtime because the sampling time periods did not include the lower feeding rates while the cold cap was being built up.

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 (6.1), (6.2), and (6.3), respectively. The values were calculated for the entire runtime (denoted in the 'Sample Duration' row as 'Run') and exclusively during each offgas sample time period (denoted in the 'Sample Duration' row as '1' or '2') for the AP-107 recycle CLSM run in Table 6.2. In addition, the average (and standard deviation) recoveries of each component during the sampling times were calculated along with the percent difference between the sampling time average and the total runtime recoveries.

Rec_i values of ~90 – 100 %, as previously reported in the CLSM (Dixon et al. 2020a) and typically observed in scaled melter systems (Matlack et al. 2012 and 2018), are reported for all the primary glass components. Several minor components' Rec_i values are reported to be outside of the 90 – 100 % range. The recoveries of Cr and Ni were greater than 100 %, due to leaching from of the CLSM vessel, as discussed in Section 6.2.2, while the recoveries of the halides (Cl and F) and ^{99}Tc were below 90 % due to their volatility. The R_{Cl} was set to 55 % of the amount in the melter feed, as explained in Section 6.2.1, while the measured R_F = 68 %, which aligned with the average single pass retention of F (~65 %) in the DM10 melter vessel calculated by VSL for a variety of LAW melter feed simulants (Matlack et al. 2012). The behavior of Cl, F, and ^{99}Tc in the offgas system is discussed in Section 6.4, with additional detail about the performance of ^{99}Tc examined further in Section 6.5.

Table 6.2. Component DF, Percent Emitted, Retention, and Recovery During the AP-107 Recycle CLSM Run

Component	Melter DF	Melter DF	Melter DF	% Emitted	% Emitted	% Emitted	*R*	*R*	*R*	Rec	Rec	Rec	Rec Avg.	Rec St. Dev.	% Diff
Sample Duration:	Run	1	2	Run	1	2	Run	1	2	Run	1	2	1&2	1&2	
Units				%	%	%	%	%	%	%	%	%	%	%	%
^{99}Tc	1.5	1.3	1.5	68	78	68	32	22	32	81	52	72	62	14	-24
Total Cs	3.1	2.5	2.1	32	40	47	68	60	53	104	251	60	156	135	50
Al	18	15	107	5	6	1	95	94	99	95	94	99	96	4	2
B	13	28	34	8	4	3	92	96	97	93	97	98	97	0	4
Ca	27	22	98	4	4	1	96	96	99	97	96	99	97	2	1
Cl	2.2	--	--	45	--	--	55	--	--	76	--	--	--	--	--
Cr	--	19	--	--	5	--	115	95	134	127	97	136	116	27	-9
F	3.1	2.4	2.8	32	42	36	68	58	64	77	61	64	62	2	-19
Fe	22	23	21	5	4	5	95	96	95	96	96	95	96	0	0
K	15	11	15	7	9	7	93	91	93	97	93	95	94	2	-3
Li	10	19	7	10	5	15	90	95	85	90	95	85	90	7	-1
Mg	17	11	35	6	9	3	94	91	97	94	91	97	94	4	0
Na	28	16	36	4	6	3	96	94	97	98	94	98	96	2	-2
Ni	--	--	--	--	--	--	120	108	146	121	108	146	127	26	5
P	--	--	6	--	--	16	102	125	84	102	126	84	105	29	2
S	10	10	7	10	10	15	90	90	85	96	92	87	89	4	-7
Si	16	13	32	6	8	3	94	92	97	94	93	97	95	3	1
Ti	18	11	--	6	9	--	94	91	102	95	91	102	96	8	2
Zn	15	11	273	7	9	0	93	91	100	94	91	100	95	6	2
Zr	17	11	--	6	9	--	94	91	102	94	91	102	96	8	2
Total	18	15	36	6	7	3	94	93	97	95	93	98	95	3	0

Values marked with ‘--’ were detected in greater concentration in the glass than the melter feed or were not calculated during sampling (Cl)

6.4 Offgas Analysis

Six components were discovered to have more than 1.5 % of their recovered inventories in the CLSM offgas system: ^{99}Tc, Cs, S, K, Cl, and F. The previous study of CLSM operation likewise registered S, K, Cl, F, and Re (a non-radioactive surrogate for ^{99}Tc) in the offgas system (Dixon et al. 2020a). ^{99m}Tc and S were similarly detected in appreciable quantities in the DM10 offgas system (Matlack et al. 2012) while K was present at higher than expected levels in the offgas system of the Large C melter (Zamecnik et al. 2002). In addition, iodine, which was not found above its ICP-MS analytical detection limit in the CLSM glass product or melter feed (~4.60 ppm), was also detected in significant quantities in the offgas system. The locations/units in the offgas system (see Figure 4.4 for unit arrangement) where these components were recovered were the sampling loop (including the sampling HEPA filters and sampling SBS sump), the primary SBS sump, the condensate (accumulated over the course of each run), the demister, and the final filters (including the pre-filter, the liquid that accumulated in the pre-filter housing, and the primary HEPA filters).

The amount and percent of each volatile component (^{99}Tc, Cs, I, S, K, Cl, and F) recovered in the glass versus the offgas system and the locations of those components in the offgas system units are listed in Table 6.3. The trends of the percent offgas recoveries for each component in the offgas units are shown in Figure 6.5. The recoveries for all components in the offgas units followed the same pattern, indicating a similar response for all components within the CLSM offgas system. Specifically, given that the halides followed the same pattern as the other volatile components, the credibility of the estimate for the Cl mass in the melter feed and recovered in the glass product, discussed in Section 6.2.1, is strengthened.

Table 6.3. Offgas Recovery During the AP-107 Recycle CLSM Run

	^{99}Tc		Cs		I	
Units	mg	%	mg	%	mg	%
Glass	18.2	39.1	15.8	65.7	0.0	0.0
Offgas System	28.4	60.9	8.2	34.3	45.7	100.0
Offgas Units						
Sampling	0.8	3.0	0.1	1.6	1.6	3.5
SBS Sump	2.9	10.4	0.7	7.9	9.4	20.9
Condensate	11.5	41.0	4.2	51.0	19.3	42.9
Demister	3.6	12.6	1.4	17.0	8.3	18.5
Filters	9.3	33.1	1.9	22.5	6.4	14.2

	S		K		Cl		F	
Units	mg	%	mg	%	mg	%	mg	%
Glass	12356	93.5	23364	96.4	13301	72.6	3510	87.8
Offgas System	858	6.5	874	3.6	5020	27.4	487	12.2
Offgas Units								
Sampling	10	1.2	31	3.6	17	0.3	9	1.9
SBS Sump	72	8.5	77	8.8	767	15.4	44	9.1
Condensate	376	44.0	336	38.6	2688	53.9	253	52.2
Demister	114	13.3	101	11.7	913	18.3	87	18.0
Filters	282	33.0	325	37.3	605	12.1	91	18.8

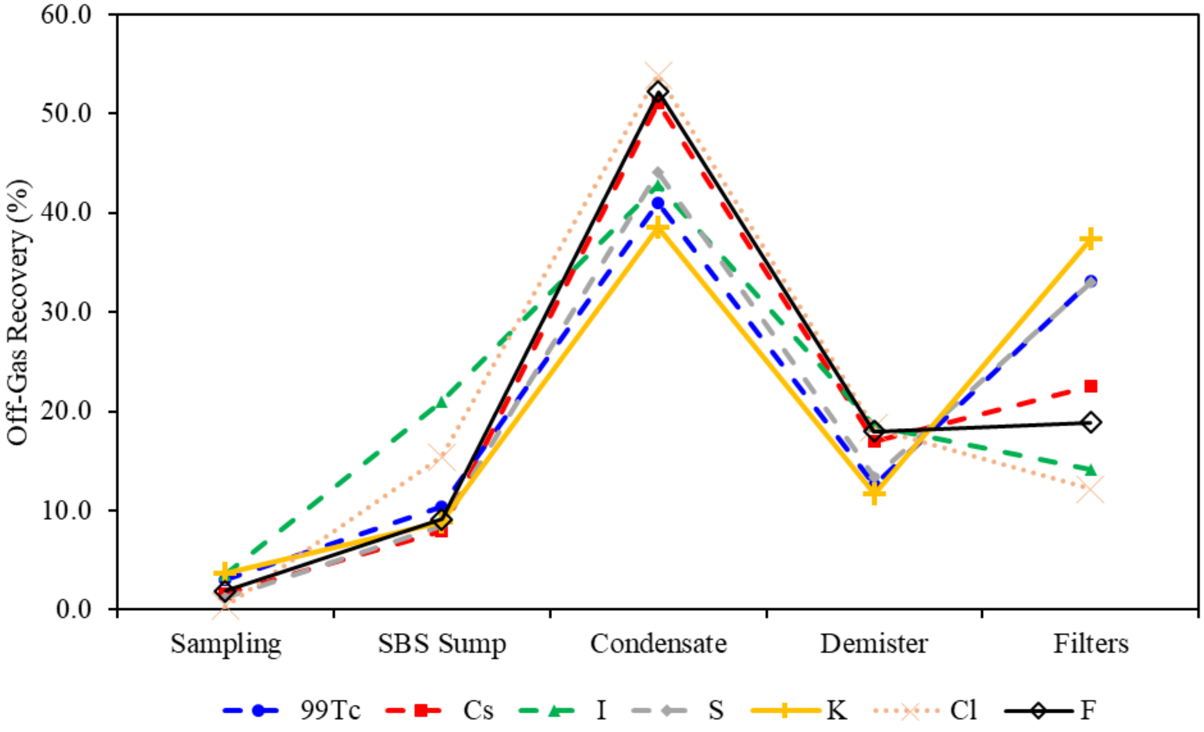

Figure 6.5. Percent of component mass recovered in each offgas unit during the AP-107 recycle CLSM run.

Compared to the previous AP-107 CLSM run (Dixon et al. 2019), a greater percentage of each component was recovered in the final filters, indicating that the updated offgas system design with the pre-filter improved component recovery. However, in the CLSM system analysis with AN-105 melter feed (Dixon et al. 2020a), 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 recovered in the offgas system (Re, S, K, Cl, and F) was discovered in the offgas piping wash. If this principle is applied for all of the components recovered in the offgas system during the AP-107 recycle CLSM run, their Rec values during the complete runtime, given in Table 6.2, increased to Rec_{99Tc} = 92 %, Rec_{Cs} = 112 %, Rec_{S} = 98 %, Rec_{K} = 98 %, Rec_{Cl} = 81 %, and Rec_{F} = 79 %. The Rec_{Cs} value being greater than 100 % may be a result of the previous AP-107 having a large Cs spike (~770 ppm) in the initial glass sample while the offgas system was not washed out between the runs and deposits in the offgas system made during the previous run may have been released during the AP-107 recycle CLSM run.

6.5 ^{99}Tc and Cs Retention and Analysis

The measured concentration of ^{99}Tc and Cs in the glass product, with respect to the mass of glass discharged, and their measured concentrations in the melter feed marked by the inset line, are shown in Figure 6.6 and their characteristic relationships are shown in Table 6.4 for both the AP-107 recycle CLSM run and the previous AP-107 non-recycle CLSM run (Dixon et al. 2019). The Cs source in the AP-107 non-recycle CLSM run was impurity in the source materials used to batch the initial glass for the CLSM vessel, so it was initially present in the glass product as a spike then decreased in each subsequent glass pour. As a result, while the total mass of Cs recovered in the CLSM offgas system during the AP-107 non-recycle CLSM run was greater than the mass recovered during the AP-107 recycle CLSM run, listed in Table 6.5, the percents recovered in each offgas unit were relatively equivalent during the two runs, shown in Figure 6.7.

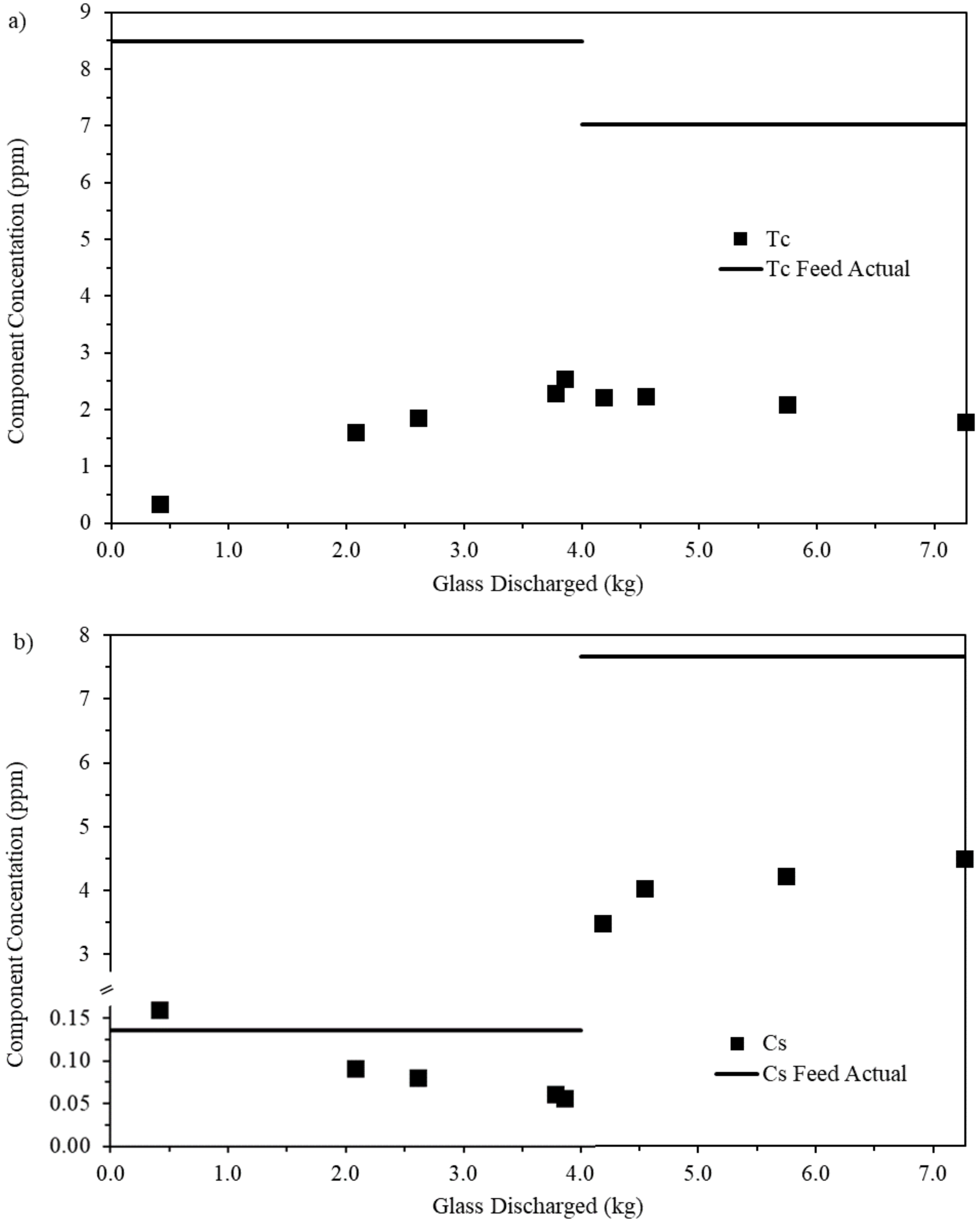

Figure 6.6. Measured a) ^{99}Tc and b) Cs content in the AP-107 recycle CLSM run glass product and melter feeds.

Table 6.4. ^{99}Tc and Cs Relationships During the AP-107 Recycle CLSM Run and AP-107 Non-Recycle CLSM Run

AP-107 Recycle

^{99}Tc Glass Target (ppm)	Cs Glass Target (ppm)	^{99}Tc/Cs Target Mass Ratio (mg mg^{-1})	Glass Pour Time (h)	Glass Discharged (kg)	^{99}Tc Glass Actual (ppm)	Cs Glass Actual (ppm)	^{99}Tc Retention (%)	^{99}Tc Recovery (%)	Cs Retention (%)	Cs Recovery (%)
8.49	0.135	62.8	0.55	0.412	0.341	0.160				
8.49	0.135	62.8	2.28	2.079	1.60	0.0908				
8.49	0.135	62.8	2.67	2.608	1.86	0.0809	22	52	60	251
8.49	0.135	62.8	4.10	3.775	2.28	0.0613				
8.49	0.135	62.8	4.50	3.853	2.55	0.0565				
7.02	7.66	0.9	8.89	4.185	2.21	3.48				
7.02	7.66	0.9	9.42	4.546	2.24	4.03	32	72	53	60
7.02	7.66	0.9	10.62	5.744	2.09	4.22				
7.02	7.66	0.9	10.93	7.266	1.78	4.50				
						Total	32	92(a)	68	112(a)

AP-107 Non-Recycle(b)

^{99}Tc Glass Target (ppm)	Cs Glass Target (ppm)	^{99}Tc/Cs Target Mass Ratio (mg mg^{-1})	Glass Pour Time (h)	Glass Discharged (kg)	^{99}Tc Glass Actual (ppm)	Cs Glass Actual (ppm)	^{99}Tc Retention (%)	^{99}Tc Recovery (%)
5.73	< 0.07	--	0.33	0.240	0.499	771		
5.73	< 0.07	--	3.19	1.981	2.04	331	35	71
5.73	< 0.07	--	3.54	2.277	4.00	294		
5.73	< 0.07	--	5.20	3.537	2.31	170	40	108
5.73	< 0.07	--	6.04	4.244	2.53	131		
5.73	< 0.07	--	7.38	5.198	2.26	73.9	39	94
5.73	< 0.07	--	9.45	6.526	2.48	39.2		
5.73	< 0.07	--	10.07	7.009	2.53	109		
						Total	50	94(a)

(a) Values reported included the estimate for mass recovered in offgas piping wash, described in Section 6.4.

(b) Dixon et al. (2019)

Table 6.5. ^{99}Tc and Cs Offgas Recovery During the AP-107 Recycle CLSM Run and AP-107 Non-Recycle CLSM Run

	AP-107 Recycle				AP-107 Non-Recycle[(a)]			
	^{99}Tc		Cs		^{99}Tc		Cs	
Units	mg	%	mg	%	mg	%	mg	%
Glass	18.2	39.1	15.8	65.7	20.3	56.7	--[(b)]	--
Offgas System	28.4	60.9	8.2	34.3	15.5	43.3	113.9[(b)]	--
				Offgas Units				
Sampling	0.8	3.0	0.1	1.6	1.6	10.4	9.7	8.5
SBS Sump	2.9	10.4	0.7	7.9	3.0	19.6	10.5	9.2
Condensate	11.5	41.0	4.2	51.0	8.8	56.7	78.4	68.9
Demister	3.6	12.6	1.4	17.0	0.9	6.0	6.5	5.7
Filters	9.3	33.1	1.9	22.5	1.1	7.2	8.8	7.7

(a) Dixon et al. (2019)

(b) The Cs in the glass was not from the melter feed, but a result of impurity in the initial glass, resulting in a spike in the offgas system.

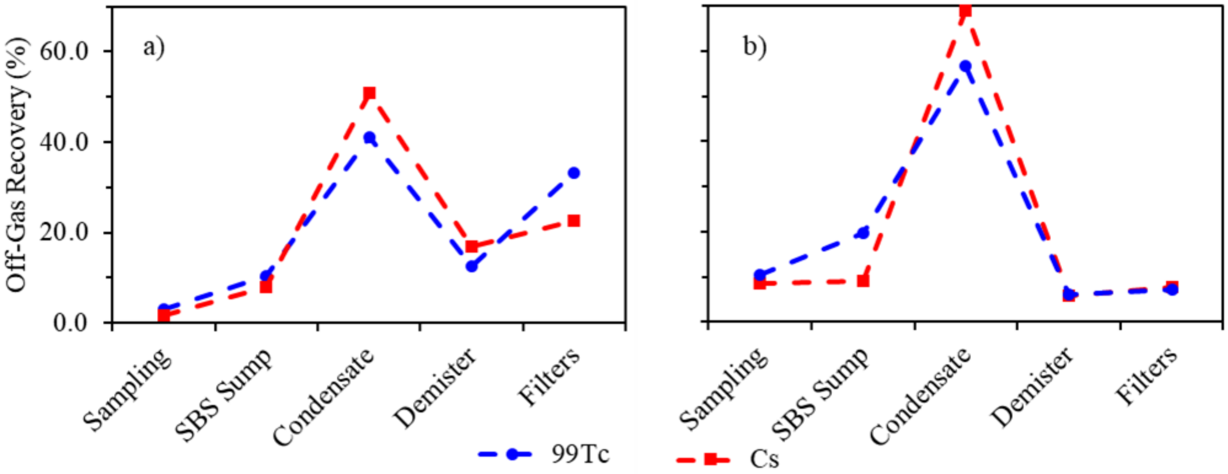

Figure 6.7. Percent of ^{99}Tc and Cs mass recovered in each offgas unit during the a) AP-107 recycle CLSM run and b) AP-107 non-recycle CLSM run (Dixon et al. 2019).

The ^{99}Tc/Cs mass ratio during the first sampling time period (see Table 6.4), in the midst of the AP-107 waste simulant recycle melter feed charging, was 62.8, while the R_{Cs} = 60 % and R_{99Tc} = 22 %. During this period, the cold-cap coverage was limited, as discussed in Section 5.1, while the trend of the ^{99}Tc content in the glass appeared to be increasing, as shown in Figure 6.6a, which helped explain why the R_{Cs} and R_{99Tc} during this period were both ~10% less than their values calculated for the total run.

During the second sampling time period, amid the AP-107 recycle melter feed charging, the ^{99}Tc/Cs mass ratio was 0.9, while the R_{Cs} = 53 % and R_{99Tc} = 32 %. It was expected that the retention values would increase during this period due to the steadier cold-cap coverage allowing less volatility, which was the case with the R_{99Tc} increasing by ~10 %. The R_{Cs} value decreased slightly (~7 %), though this relative change was within the variability expected for measurements of low concentrations of Cs in glass during short sampling times given the differences in retention calculations for other components in the glass product, shown in Table 6.2.

The retention of Cs for the total runtime during the AP-107 recycle CLSM run was R_{Cs} = 68 %, which spanned the ^{99}Tc/Cs mass ratio from 0.9 to 62.8. This value in the CLSM aligned with the value of R_{Cs} ≈ 60 %, calculated by Matlack et al. (2004) from a series of runs on the DM100 melter system with a LAW simulant of AN-105 waste at ^{99}Tc/Cs mass ratios varying from 1 to 100, and the value of R_{Cs} = 73.05 ± 10.97 %, calculated by Zamecnik et al. (2002) from a run on the Large C Melter system with pretreated Hanford tank 241-AN-102 waste and a ^{99}Tc/Cs mass ratio of ~0.8. The similarity across different scaled melter systems and melter feed compositions indicated the consistency of Cs performance during LAW vitrification in the described ^{99}Tc/Cs mass ratio range. In addition, the AP-107 recycle CLSM run and the work by Zamecnik et al. (2002) were performed without ^{99}Tc or Cs spikes, thus affirming Cs behavior at the concentrations of those components expected during DFLAW operations. The Cs retention results from the AP-107 recycle CLSM run do not address the Cs behavior at much greater ^{99}Tc/Cs mass ratios (>100), also performed by Matlack et al. (2004), or the concerns raised by Kim and Kruger (2018) about the mass of the ^{99}Tc spike (or its non-radioactive surrogate, Re) required to perform work at such high ^{99}Tc/Cs mass ratios that would not be present during the DFLAW program.

The retention of ^{99}Tc during the second sampling time period of the AP-107 recycle CLSM run, when the cold-cap coverage was believed to be steady, was R_{99Tc} = 32 %. During the second and third sampling time periods of the AP-107 non-recycle CLSM run, when the cold-coverage was believed to be steady as discussed in Dixon et al. (2019), the R_{99Tc} = 40 ± 1 %, which was ~8 % greater than during the AP-107 recycle CLSM run under similar cold-cap conditions. The primary reason for the difference in ^{99}Tc retention between the two runs is believed to be the pouring of a majority of the glass inventory during the midst of the AP-107 recycle CLSM run, which resulted in an idling period when ^{99}Tc volatilized from the remaining glass. However, the R_{99Tc} value during the AP-107 recycle CLSM run was within the range achieved by Zamecnik et al. (2002) of R_{99Tc} = 30.85 ± 4.08 %.

The action of evaporating the condensate from the AP-107 non-recycle CLSM run and adding it to the AP-107 waste effectively mimicked the recycle action as demonstrated by the increased levels of ^{99}Tc and Cs in the melter feed during the AP-107 recycle run (see ^{99}Tc and Cs ‘Glass Target’ columns in Table 6.4). While the Cs spike in the initial glass during the AP-107 non-recycle CLSM run and the cold-cap characteristics during the first half of the AP-107 recycle run make the efficacy of the recycle action difficult to observe from the R_{99Tc} and R_{Cs} values, the recovery of ^{99}Tc and Cs in their stated quantities and locations in the CLSM offgas system (see Table 6.5 and Figure 6.7) points to the appropriate recycle behavior if consistent cold-cap conditions are maintained during future CLSM runs. In addition, the increased content of other volatile components, like S, Cl, and F, in the target recycle AP-107-1R glass composition, compared to the target non-recycle AP-107 glass composition (see Table 4.4), and the retention of those components in the measured AP-107 recycle CLSM run glass product (see Table 6.1), indicate that the recycle action did not negatively affect the AP-107 waste vitrification.

6.6 Production Comparison with Previous AP-107 CLSM Run

A comparison of the production results in both the AP-107 recycle CLSM run and the AP-107 non-recycle CLSM run is shown in Table 6.6. A furnace with greater power capacity was installed between the two CLSM runs and as a result, the average glass temperature during the AP-107 recycle CLSM run remained in the target range (1150 ± 30 °C). The average glass production rate during the AP-107 recycle CLSM run was greater than during the AP-107 non-recycle CLSM run, but the difference in production rate between the two runs was within the variable range for production of the same melter feed in the CLSM system of ~300 kg m^2 d^{-1} established previously (Dixon et al. 2020a). The differences in the bubbling flux rate and the average plenum temperature between the two runs are a result of the production time during the AP-107 recycle CLSM run when the bubble placement through the cold cap was directly below the charging melter feed and the desired cold-cap coverage could not be maintained.

Table 6.6. Production Results During the AP-107 Recycle CLSM Run and the AP-107 Non-Recycle CLSM Run

Parameter	AP-107 Recycle	AP-107 Non-Recycle[(a)]
Test Date	February 25, 2020	August 8, 2018
Feeding Duration, h	8.87	10.07
Low Flow Duration, h	0.00	1.71
Downtime, h	1.93	0.00
Glass Produced, kg	7.27	7.01
Melter Feed Consumed, kg	16.57	15.0
Average Glass Production Rate, kg m^{-2} d^{-1}	1739	1477
Average Feeding Rate, kg h^{-1}	1.87	1.49
Average Bubbling Flux Rate, L m^{-2} min^{-1}	70	149
Average Glass Temperature, °C	1138	1115
Average Plenum Temperature, °C	700	648

(a) Dixon et al. (2019)

7.0 Conclusions

During the previous processing of AP-107 waste in the CLSM, 3.4 L of offgas condensate were collected during vitrification. A test apparatus was designed to operate like the EMF evaporator and used to successfully concentrate the AP-107 condensate by a factor of 10 while retaining over 90 % of the ^{99}Tc, Cs, and I inventory. A second portion 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 the recycling action to be performed at the WTP. The remainder of the AP-107 waste was combined with a simulant of the AP-107 condensate concentrate. A glass composition, AP-107-1R, was calculated from the Kim et al. (2012) glass models based on the composition of the combined AP-107 and condensate concentrate. GFCs were added to the two portions of AP-107 waste plus condensate concentrate (or simulant) and the resultant melter feeds were processed in the CLSM, during which two offgas samples were collected.

Over 8.87 hours of processing, 7.27 kg of AP-107-1R were produced for an average glass production rate of 1739 kg m^2 d^{-1}. This average glass production rate was greater than the processing rate of the AP-107 waste without the recycle, but within the potential range when processing melter feeds with similar composition. Other conclusions from the conversion of the AP-107 waste with recycled condensate concentrate include:

- All the primary components in the glass product from the CLSM were within 10 % of their targets based on the AP-107-1R glass composition.
- Cr_2O_3 and NiO content in the glass product from the CLSM were greater than their targets based on the AP-107-1R glass composition due to leaching from the CLSM vessel.
- The levels of minor impurity components (< 300 ppm) in the glass product from the CLSM did not change after the glass inventory had been turned over twice.
- The activity of radionuclides in the melter feeds (^{241}Am, $^{239/240}$Pu, ^{238}Pu, ^{237}Np, and $^{243/244}$Cm) were retained in the glass product from the CLSM.
- 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.
- While the cold cap covered ~80 % of the glass melt surface, 22% of the ^{99}Tc and 60% of the Cs were retained in the glass product.
- While the cold cap covered ~95 % of the glass melt surface, 32% of the ^{99}Tc and 53% of the Cs were retained in the glass product.
- Cs results indicated that Cs retention in glass varied little due to the ^{99}Tc/Cs relationship in the ^{99}Tc/Cs mass ratio range from 1 to 100, when the Cs content in the glass was at the levels expected given a condensate recycle with real Hanford tank waste.
- ^{99}Tc results indicated that a pouring event, when a majority of the glass inventory was poured from the CLSM, and the subsequent idling period together affected the total ^{99}Tc retention and recovery, allowing it to volatilize from the glass melt.

Ultimately, the condensate recycle action and ^{99}Tc/Cs relationship results have demonstrated the ability of the CLSM to support future WTP programmatic needs regarding cold-cap behavior, glass processing operations, and an understanding of component volatility into the offgas.

8.0 References

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Appendix A – CLSM Run Results Data

The figures and tables in this section display the various processing values (temperatures, effective glass production rates, bubbling flux rates, and melter vacuum measurements) collected during the AP-107 recycle CLSM run, the complete chemical analytical results, the compositional trends for each component oxide in the analyzed glass product, and component mass flow rates.

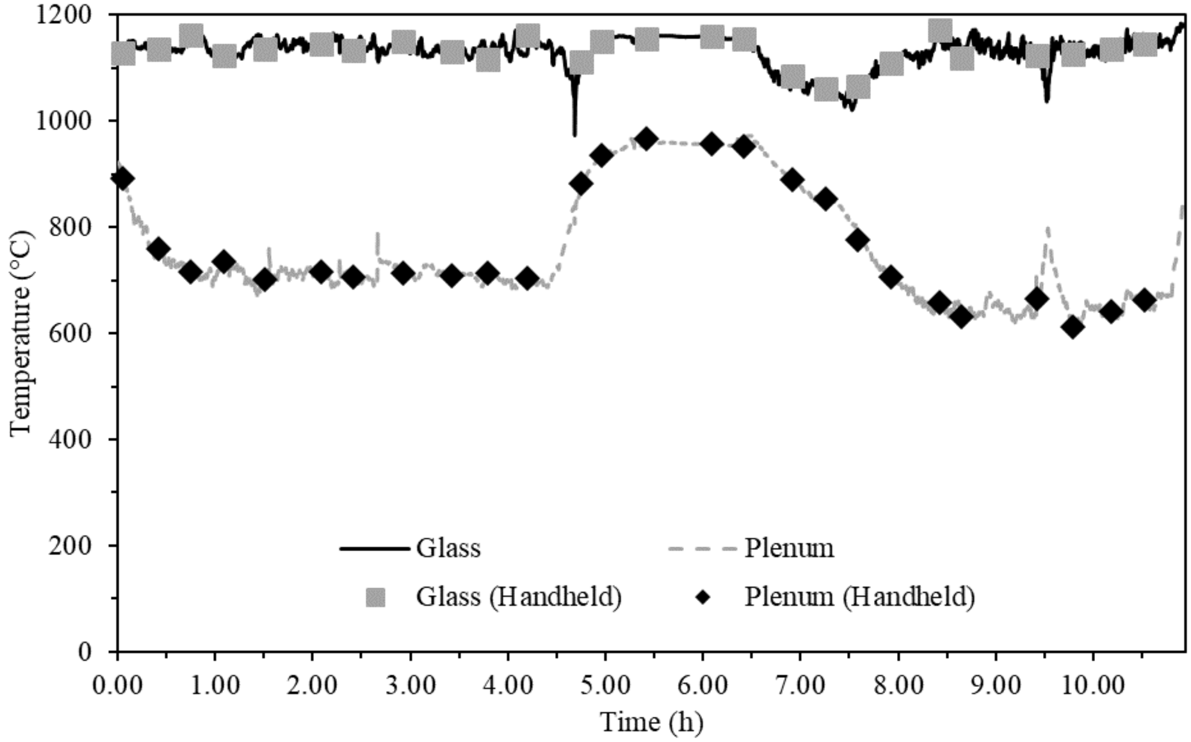

Figure A.1. Glass and plenum temperatures recorded by the CLSM data acquisition system and a calibrated handheld device during the AP-107 recycle CLSM run.

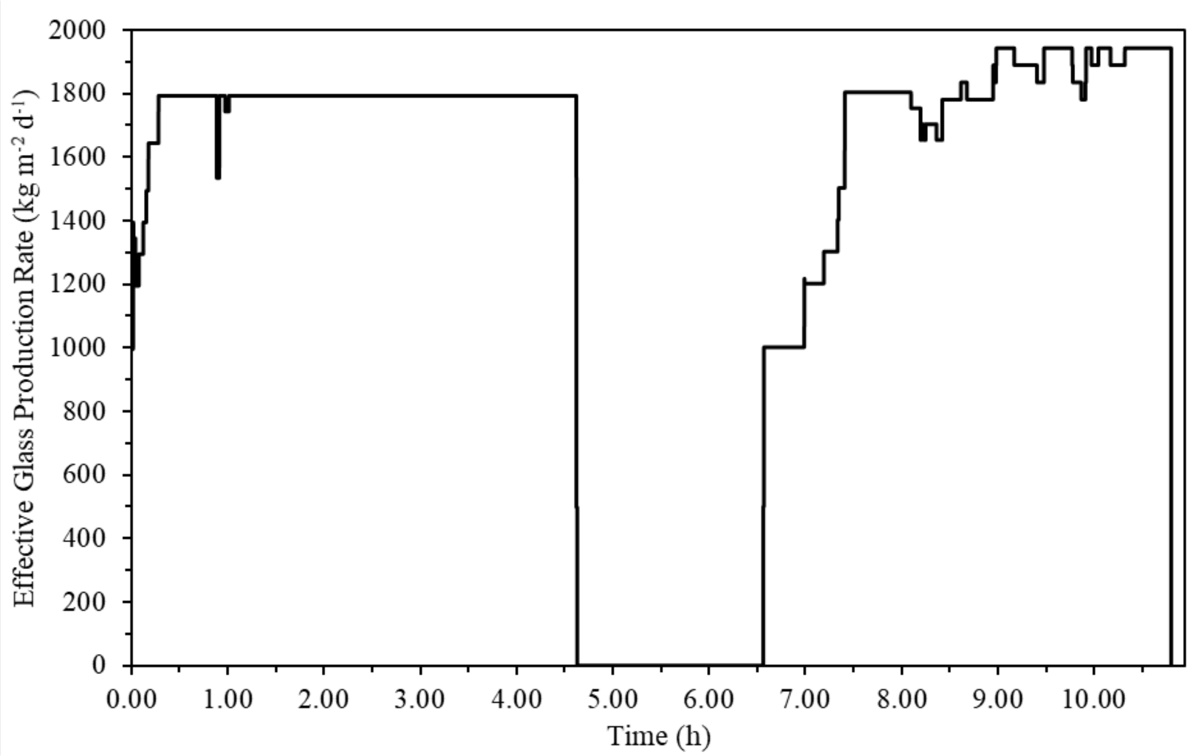

Figure A.2. Effective glass production rate during the AP-107 recycle CLSM run.

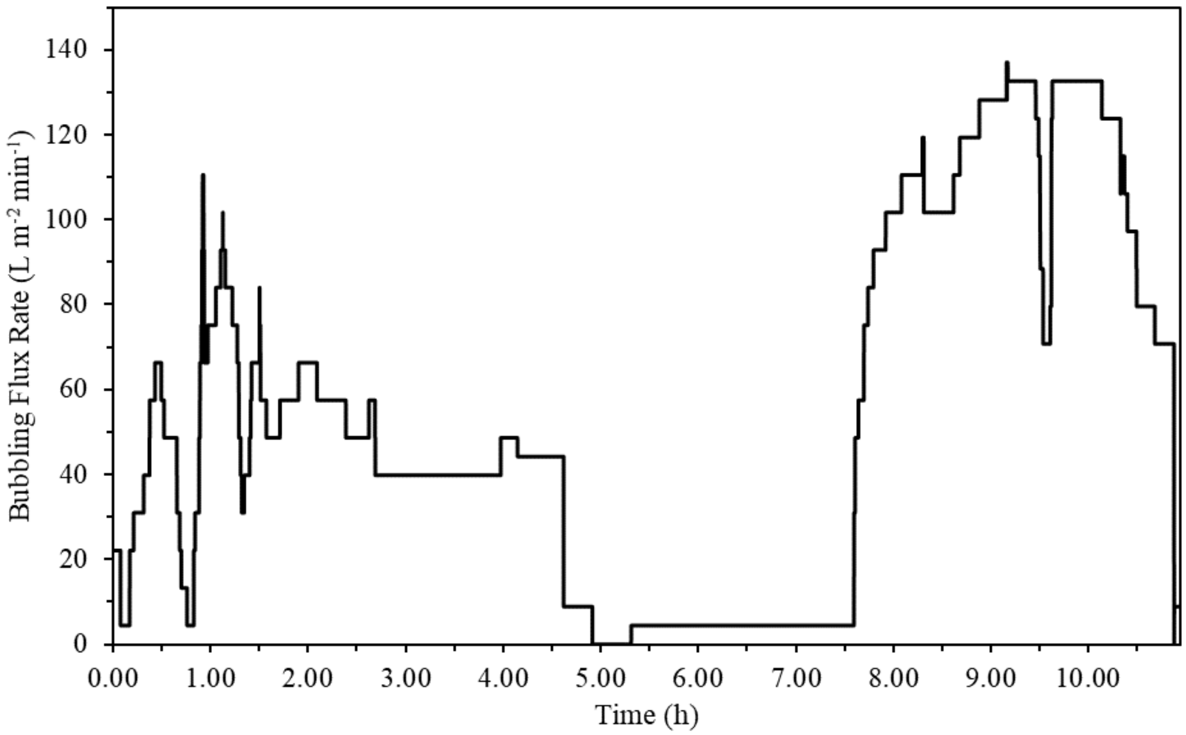

Figure A.3. Bubbling flux rate during the AP-107 recycle CLSM run.

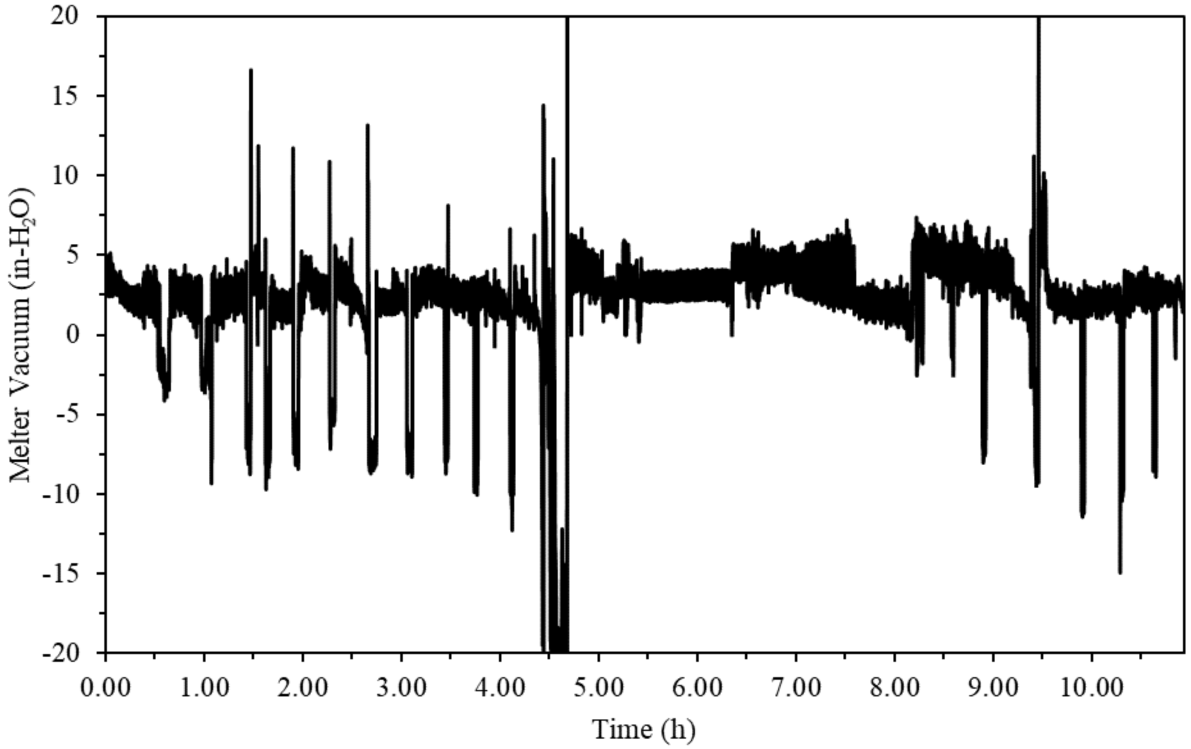

Figure A.4. Melter vacuum during the AP-107 recycle CLSM run.

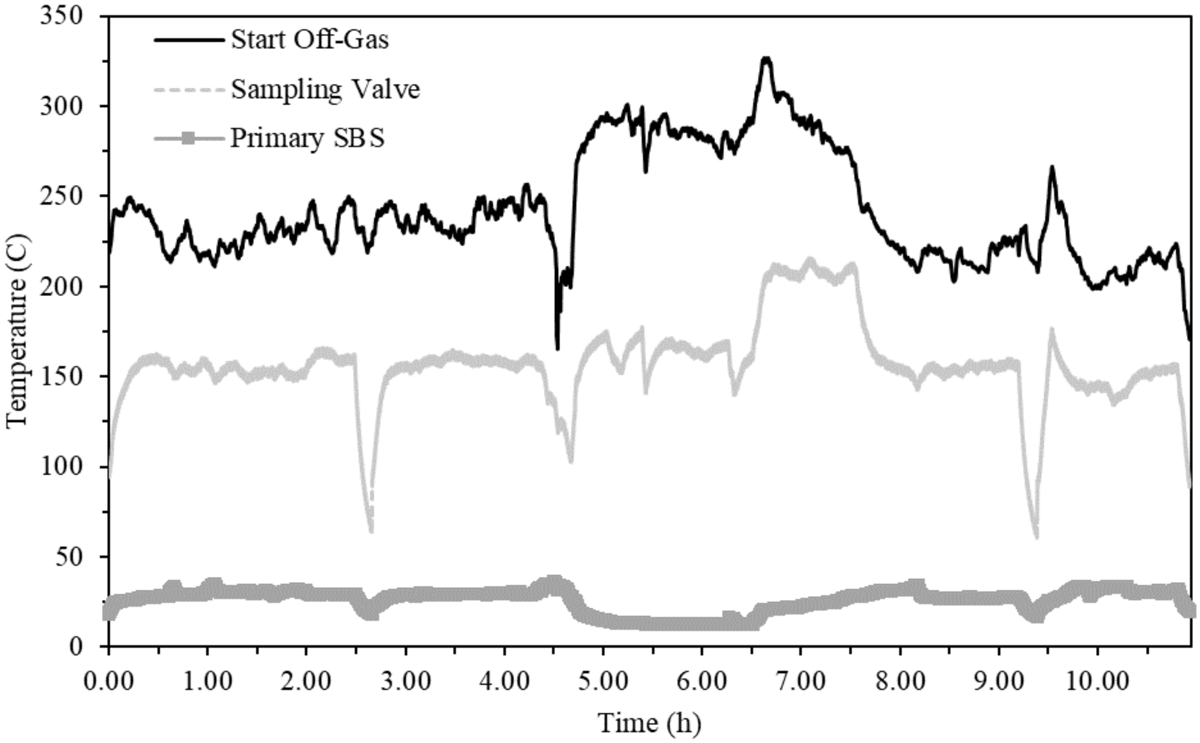

Figure A.5. Start of offgas system, sampling valve, and primary SBS temperatures during the AP-107 recycle CLSM run.

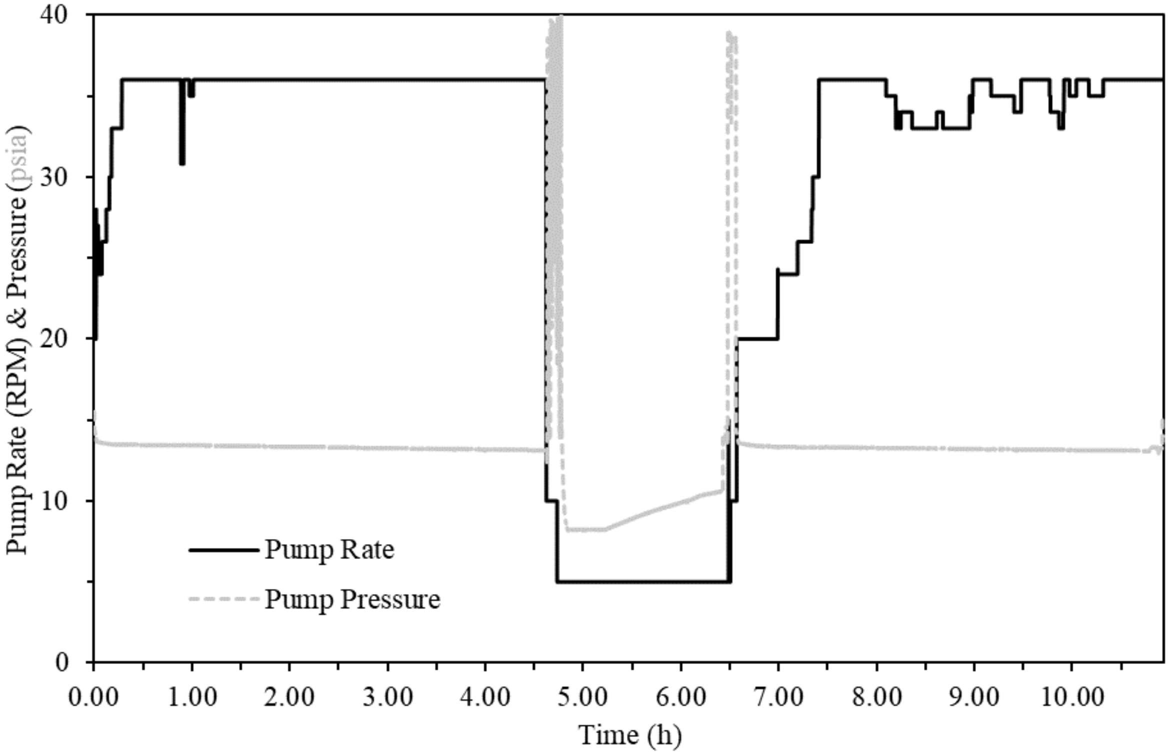

Figure A.6. Progressive cavity pump rate and pressure during the AP-107 recycle CLSM run.

Table A.1. Chemical Analysis of Selected Samples Produced During the AP-107 Recycle CLSM Run

Sample Name	Sample Type	Sample Mass (g)	Component Concentration (mg kg^{-1})								
			^{99}Tc	Total Cs	Total I	Al	Ba	B	Cd	Ca	Cr
Glass Pour 0.55	Glass	412.00	0.341	0.160	--	32500	10.6	30600	0.965	32300	694
Glass Pour 2.28	Glass	1666.90	1.60	0.0908	--	32100	25.2	30000	3.52	31900	711
Glass Pour 2.67	Glass	529.09	1.86	0.0809	--	32100	27.4	29800	3.92	31800	646
Glass Pour 4.10	Glass	1166.73	2.28	0.0613	--	31400	32.7	28800	4.80	31400	661
Glass Pour 4.50	Glass	1801.31	2.55	0.0565	14.4	31900	34.4	26100	5.05	32000	645
Glass Pour 8.89	Glass	331.86	2.21	3.48	--	31700	37.5	28600	4.83	32100	865
Glass Pour 9.42	Glass	360.65	2.24	4.03	--	31400	37.7	30100	4.75	31600	885
Glass Pour 10.62	Glass	1198.08	2.09	4.22	--	31700	37.7	29800	4.76	31500	818
Glass Pour 10.93	Glass	1772.38	1.78	4.50	--	31200	37.9	30100	4.82	31600	936
AP-107 Simulant Recycle Melter Feed	Melter Feed	9901.30	3.73	0.0594	--	15050	17.0	13550	2.62	14600	299
AP-107 Recycle Melter Feed	Melter Feed	6701.06	3.08	3.36	--	13900	17.1	13600	2.12	14000	290
Primary HEPA Filter A	HEPA Filter	18.21	0.553	0.0871	--	9910	14800	9340	--	3960	--
Primary HEPA Filter B	HEPA Filter	14.72	1.56	0.424	--	11800	17000	10700	--	4760	57.2
Sampling HEPA Filter 1	HEPA Filter	10.60	30.7	1.57	--	11600	16500	11000	--	4690	106
Sampling HEPA Filter 2	HEPA Filter	10.83	38.0	6.55	--	11000	16100	11600	--	4600	82.8
Sampling HEPA Filter 3	HEPA Filter	10.31	0.00734	0.0669	--	11900	16900	11400	--	4880	--
Pre-Filter	HEPA Filter	533.37	9.15	1.51	0.710	35.6	--	493	--	64.3	332
Condensate Combination 1	Aqueous	2692.93	1.19	0.638	2.79	19.5	--	133	--	27.2	15.0
Condensate Combination 2	Aqueous	830.32	3.53	1.19	4.50	66.0	--	407	--	111	59.9
Condensate Combination 3	Aqueous	971.16	2.84	0.84	3.11	45.4	--	324	--	65.6	61.2
Condensate Combination 4	Aqueous	1017.64	2.61	0.666	4.92	29.9	--	248	--	43.4	41.8
Demister Combination 1	Aqueous	1068.49	1.76	0.868	4.94	31.7	--	209	--	44.0	28.3
Demister Combination 2	Aqueous	605.91	2.76	0.775	5.04	41.6	--	303	--	63.0	55.6
Seal Pot Liquid	Aqueous	402.21	0.476	--	1.77	--	--	19.3	--	--	4.11
Sampling SBS Sump	Aqueous	1260.71	0.0809	0.0349	1.26	--	--	16.3	--	--	1.57
Primary SBS Sump	Aqueous	1192.84	2.45	0.547	7.88	16.5	--	177	--	--	22.2
Pre-Filter Liquid	Aqueous	753.82	5.87	1.38	7.94	54.8	--	527	--	88.7	187

Values marked with ‘--’ denote that the component concentration was below the analysis detection limit.

Table A.1. Chemical Analysis of Selected Samples Produced During the AP-107 Recycle CLSM Run (cont.)

	Component Concentration (mg kg^{-1})											
Sample Name	Co	Cu	Fe	La	Pb	Li	Mg	Mn	Mo	Ni	P	K
Glass Pour 0.55	--	18.3	38700	--	2.53	6840	8630	46.1	11.6	139	915	3070
Glass Pour 2.28	2.48	18.5	38300	--	4.09	6830	8800	71.0	30.3	128	556	3130
Glass Pour 2.67	2.64	18.6	38300	1.49	3.74	6700	8730	72.9	34.1	122	850	3180
Glass Pour 4.10	3.46	18.7	37850	1.80	4.30	6690	8720	81.1	40.3	116	839	3170
Glass Pour 4.50	3.50	18.6	38500	2.36	4.77	6700	8930	82.7	41.4	112	777	3210
Glass Pour 8.89	4.19	18.9	38200	2.41	4.45	6660	8810	89.6	46.9	167	844	3010
Glass Pour 9.42	3.80	18.1	37100	2.51	5.01	6520	8550	87.7	47.7	163	628	3100
Glass Pour 10.62	3.84	18.4	37600	2.64	6.18	6470	8450	87.2	47.5	150	820	3120
Glass Pour 10.93	3.79	18.7	37800	2.55	3.96	6530	8630	88.3	50.6	164	805	3340
AP-107 Simulant Recycle Melter Feed	1.89	7.81	17550	1.41	2.24	3105	4215	40.3	21.3	49.5	298	1540
AP-107 Recycle Melter Feed	1.91	7.91	17100	--	1.95	3370	3860	40.4	20.8	49.1	329	1460
Primary HEPA Filter A	--	1.47	--	--	2.16	4.24	516	8.61	22.5	4.08	12.2	7350
Primary HEPA Filter B	--	2.02	--	--	2.34	8.15	586	14.3	27.5	6.77	12.0	9030
Sampling HEPA Filter 1	--	1.18	--	--	2.57	70.3	624	4.45	31.9	3.26	26.5	9205
Sampling HEPA Filter 2	--	1.05	--	--	3.34	68.8	675	4.65	30.6	2.87	19.1	9160
Sampling HEPA Filter 3	--	0.811	--	--	2.28	2.49	680	4.05	28.1	2.19	11.7	8850
Pre-Filter	--	0.925	399	--	--	39.1	14.0	1.87	2.11	2.93	--	296
Condensate Combination 1	--	--	40.5	--	--	7.68	--	--	0.763	--	--	34.4
Condensate Combination 2	--	--	122	--	--	26.8	3.59	--	2.11	--	9.40	106
Condensate Combination 3	--	--	87.8	--	--	19.8	2.49	--	1.72	--	--	82.8
Condensate Combination 4	--	--	57.1	--	--	15.2	4.44	--	1.38	--	--	73.3
Demister Combination 1	--	--	75.4	--	--	12.4	--	--	1.17	--	6.18	51.7
Demister Combination 2	--	--	95.1	--	--	17.9	3.27	--	1.61	--	--	76.3
Seal Pot Liquid	--	--	8.98	--	--	1.75	--	--	--	--	--	9.12
Sampling SBS Sump	--	--	7.78	--	--	0.554	--	0.651	--	--	--	--
Primary SBS Sump	--	--	30.4	--	--	10.7	4.43	--	1.06	--	--	64.3
Pre-Filter Liquid	--	0.821	148	--	--	34.0	4.66	2.19	3.46	6.45	9.78	188

Values marked with ‘--’ denote that the component concentration was below the analysis detection limit.

Table A.1. Chemical Analysis of Selected Samples Produced During the AP-107 Recycle CLSM Run (cont.)

	Component Concentration (mg kg^{-1})											
Sample Name	Si	Na	Sr	S	Sn	Ti	W	V	Y	Zn	Zr	TOC
Glass Pour 0.55	211000	119000	20.8	1590	15.0	8450	206	5.46	4.84	29000	20800	--
Glass Pour 2.28	210000	123000	29.4	1610	20.1	8430	227	32.9	24.0	29400	20400	--
Glass Pour 2.67	212000	124000	30.6	1720	22.2	8470	239	37.0	27.1	29400	20800	--
Glass Pour 4.10	209500	124000	33.8	1705	23.4	8410	247	46.8	33.8	29200	20550	--
Glass Pour 4.50	216000	125000	34.7	1740	23.5	8580	249	49.3	35.7	29100	21100	--
Glass Pour 8.89	219000	125000	36.5	1540	26.1	8500	252	54.6	39.4	29300	20600	--
Glass Pour 9.42	213000	125000	36.5	1570	24.3	8410	247	54.4	39.3	28400	20400	--
Glass Pour 10.62	213000	124000	36.3	1650	25.2	8380	253	54.2	39.1	28100	20400	--
Glass Pour 10.93	209000	129000	36.5	1730	25.0	8330	267	54.2	39.4	27800	20200	--
AP-107 Simulant Recycle Melter Feed	100650	58050	16.4	837	7.28	4095	107	3.92	3.72	14150	10035	--
AP-107 Recycle Melter Feed	96400	56400	16.1	812	7.29	3630	98.8	4.32	3.76	12500	8790	--
Primary HEPA Filter A	333000	20100	174	161	--	--	63.8	--	2.01	9920	--	--
Primary HEPA Filter B	391000	24800	206	178	1.86	--	82.4	--	2.41	12300	83.9	--
Sampling HEPA Filter 1	389500	30700	201	582	--	--	86.8	--	2.39	10900	--	--
Sampling HEPA Filter 2	393000	30900	200	639	2.67	--	86.2	--	2.29	10800	--	--
Sampling HEPA Filter 3	401000	24500	210	163	--	--	75.3	--	2.37	11700	--	--
Pre-Filter	14.4	3215	--	257	--	2.78	3.23	--	--	135	3.14	--
Condensate Combination 1	54.4	583	--	38.9	--	2.11	1.52	--	--	42.6	2.95	19.6
Condensate Combination 2	188	1780	--	121	--	6.45	3.29	--	--	141	8.98	21.7
Condensate Combination 3	123	1340	--	95.9	--	4.58	2.59	--	--	94.8	7.01	33.7
Condensate Combination 4	85.2	1080	--	76.3	--	3.98	2.37	--	--	67.0	5.01	49.3
Demister Combination 1	77.7	889	--	57.9	--	3.46	2.05	--	--	69.1	4.72	27.1
Demister Combination 2	108	1230	--	85.3	--	4.73	2.97	--	--	89.5	6.53	42.0
Seal Pot Liquid	6.57	129	--	12.5	--	--	--	--	--	5.02	--	9.92
Sampling SBS Sump	6.51	39.0	--	--	--	--	--	--	--	5.53	--	23.6
Primary SBS Sump	52.7	870	--	60.6	--	2.33	2.16	--	--	38.2	2.51	54.9
Pre-Filter Liquid	136	2490	--	191	--	6.75	3.33	--	--	118	7.90	39.3

Values marked with ‘--’ denote that the component concentration was below the analysis detection limit.

Table A.1. Chemical Analysis of Selected Samples Produced During the AP-107 Recycle CLSM Run (cont.)

Sample Name	Component Concentration (mg kg^{-1})				Component Concentration (µCi g^{-1})					
	Cl	F	N (Nitrate)	N (Nitrite)	^{237}Np	^{242}Cm	$^{243/244}$Cm	^{241}Am	^{238}Pu	$^{239/240}$Pu
Glass Pour 0.55	--	236	--	--	2.55E+00	--	--	2.77E+01	2.99E+00	4.16E+01
Glass Pour 2.28	--	395	--	--	1.29E+01	--	7.42E+00	1.46E+02	2.66E+01	2.07E+02
Glass Pour 2.67	--	429	--	--	1.56E+01	--	7.41E+00	1.55E+02	2.13E+01	1.51E+02
Glass Pour 4.10	382	445	--	--	2.10E+01	1.88E+00	8.86E+00	1.96E+02	2.37E+01	1.91E+02
Glass Pour 4.50	398	492	--	--	2.69E+01	--	9.64E+00	2.20E+02	2.92E+01	1.97E+02
Glass Pour 8.89	--	317	--	--	1.98E+01	--	1.37E+01	2.32E+02	2.85E+01	1.70E+02
Glass Pour 9.42	--	434	--	--	1.82E+01	9.42E+00	9.21E+00	2.38E+02	2.46E+01	1.90E+02
Glass Pour 10.62	--	447	--	--	1.56E+01	--	1.05E+01	2.23E+02	8.16E+01	1.78E+02
Glass Pour 10.93	--	446	--	--	1.78E+01	--	8.09E+00	2.37E+02	2.66E+01	1.92E+02
AP-107 Simulant Recycle Melter Feed	--	322	--	--	9.54E+00	--	5.65E+00	1.10E+02	1.32E+01	9.99E+01
AP-107 Recycle Melter Feed	--	298	--	--	4.80E+00	--	5.66E+00	1.05E+02	8.30E+00	6.05E+01
Primary HEPA Filter A	--	--	--	--	--	--	--	2.63E+00	1.52E+01	--
Primary HEPA Filter B	--	--	--	--	--	--	--	--	1.45E+00	--
Sampling HEPA Filter 1	2345	221	--	--	--	--	--	2.12E+00	2.30E+01	4.35E+00
Sampling HEPA Filter 2	4500	--	--	--	--	--	--	--	--	5.29E+00
Sampling HEPA Filter 3	--	--	--	--	--	4.28E+00	--	3.62E+00	3.22E+00	--
Pre-Filter	301	52.6	18950	21.7	4.84E-02	4.84E+00	1.75E-01	8.68E+00	--	--
Condensate Combination 1	313	30.8	2730	6.97	--	--	--	--	--	5.27E-01
Condensate Combination 2	738	74.8	4370	34.5	--	--	--	9.50E-01	--	1.13E+00
Condensate Combination 3	626	60.1	4140	65.5	--	--	--	8.78E-01	--	7.31E-01
Condensate Combination 4	614	48.3	4770	120	--	--	--	5.25E-01	--	8.62E-01
Demister Combination 1	477	46.1	6660	299	--	--	--	4.63E-01	--	4.95E-01
Demister Combination 2	666	62.1	7050	263	--	--	--	8.40E-01	--	1.19E+00
Seal Pot Liquid	71.2	8.72	3320	132	--	--	--	--	--	4.79E-01
Sampling SBS Sump	21.3	5.51	596	--	--	--	--	--	--	--
Primary SBS Sump	643	36.9	5290	101	--	--	--	--	--	--
Pre-Filter Liquid	590	83.8	25900	33.5	--	--	--	7.07E-01	--	1.14E+00

Values marked with '--' denote that the component concentration was below the analysis detection limit.

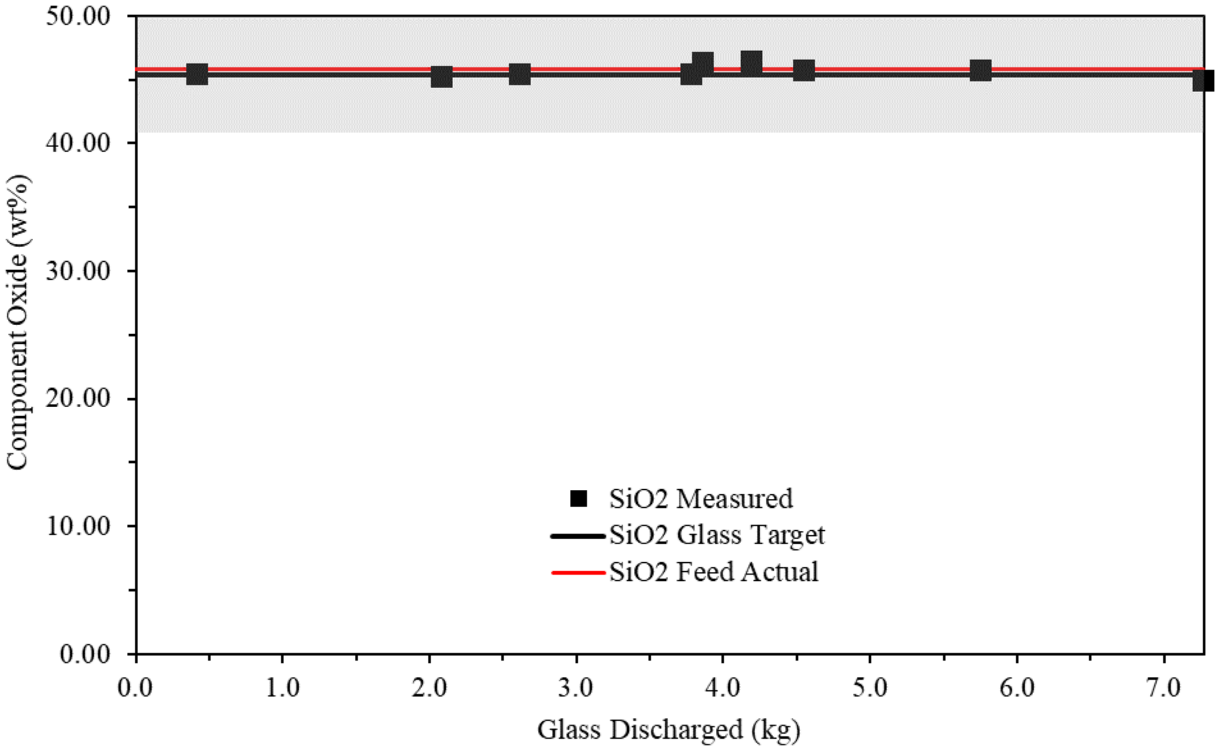

Figure A.7. Measured SiO_2 content in the AP-107 recycle CLSM run glass product and melter feeds along with the SiO_2 content target from the AP-107-1R glass composition.

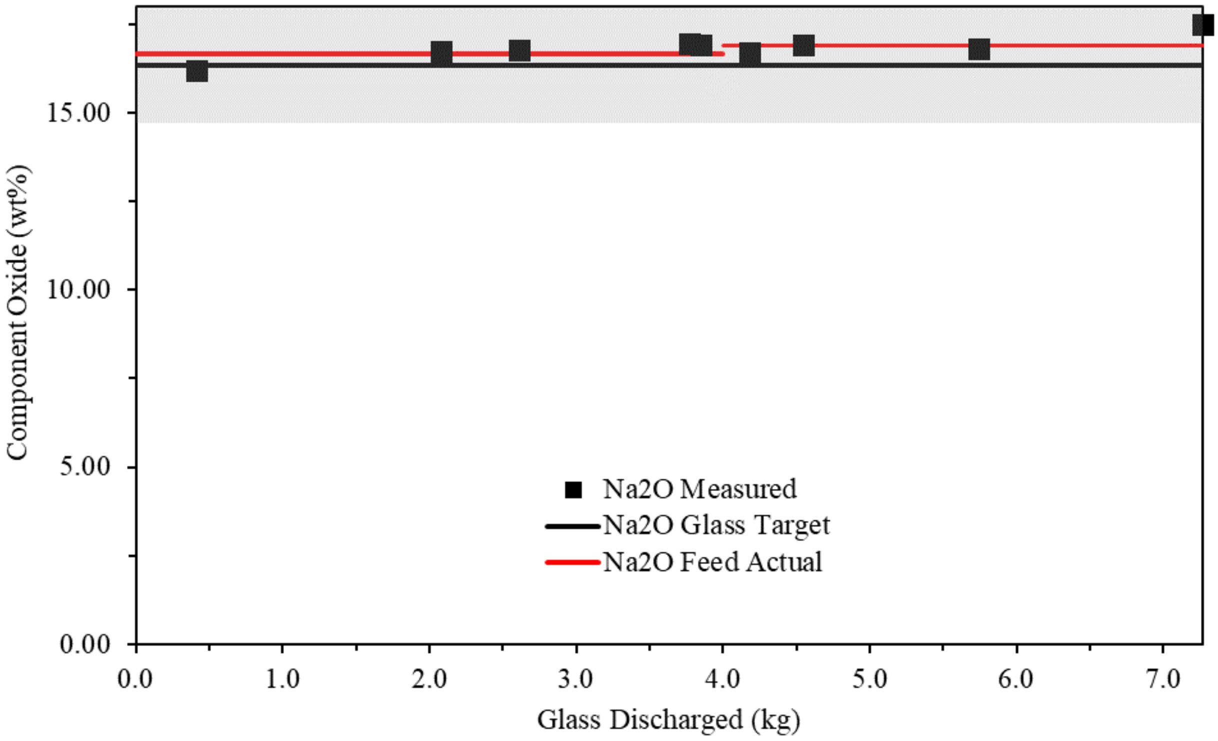

Figure A.8. Measured Na_2O content in the AP-107 recycle CLSM run glass product and melter feeds along with the Na_2O content target from the AP-107-1R glass composition.

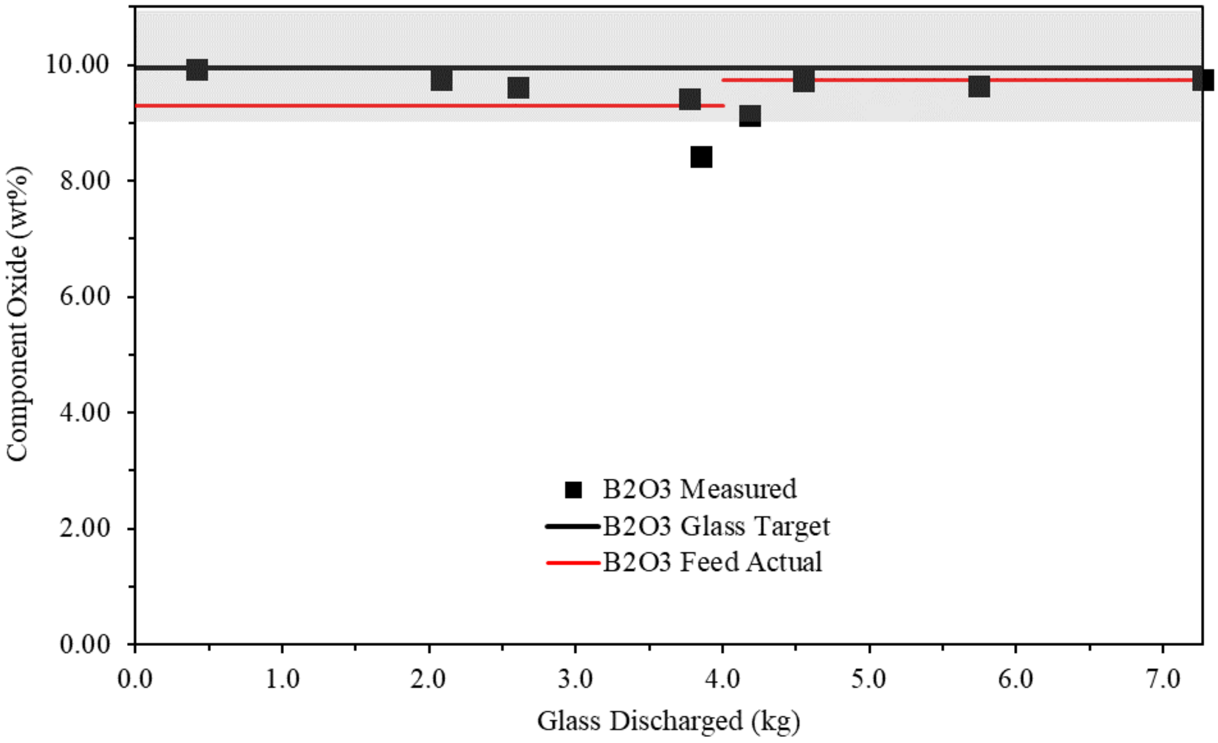

Figure A.9. Measured B_2O_3 content in the AP-107 recycle CLSM run glass product and melter feeds along with the B_2O_3 content target from the AP-107-1R glass composition.

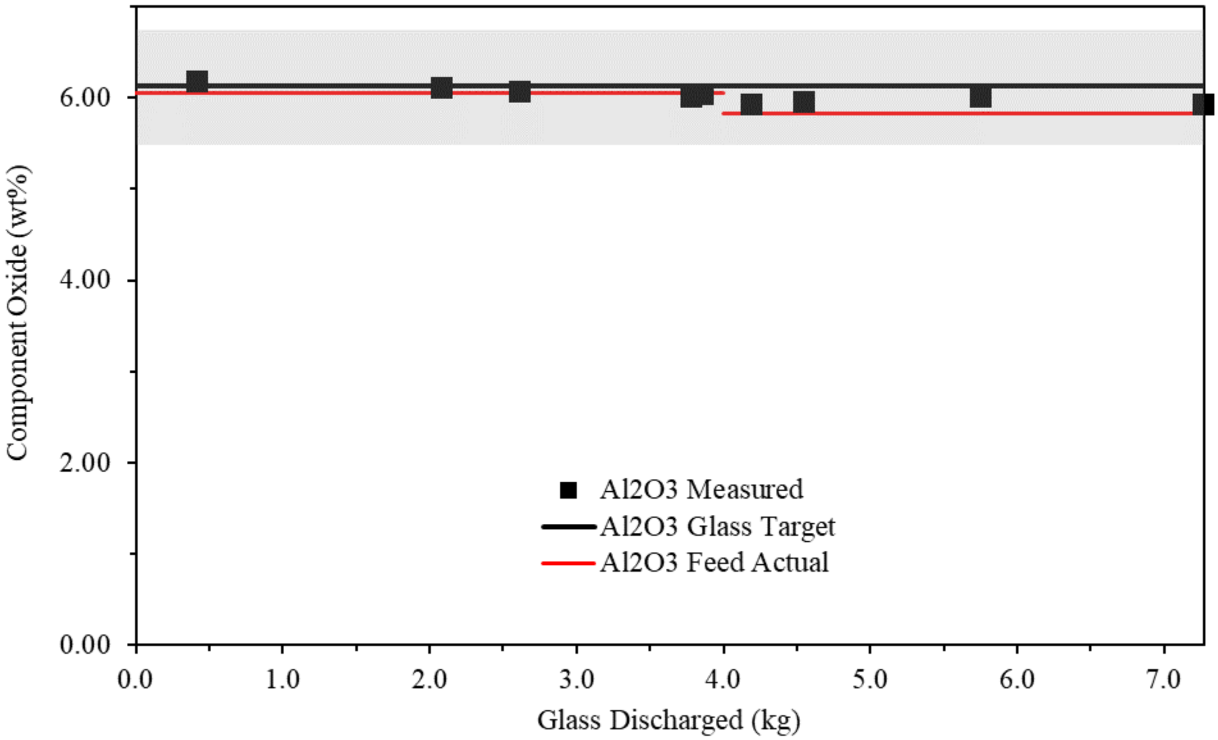

Figure A.10. Measured Al_2O_3 content in the AP-107 recycle CLSM run glass product and melter feeds along with the Al_2O_3 content target from the AP-107-1R glass composition.

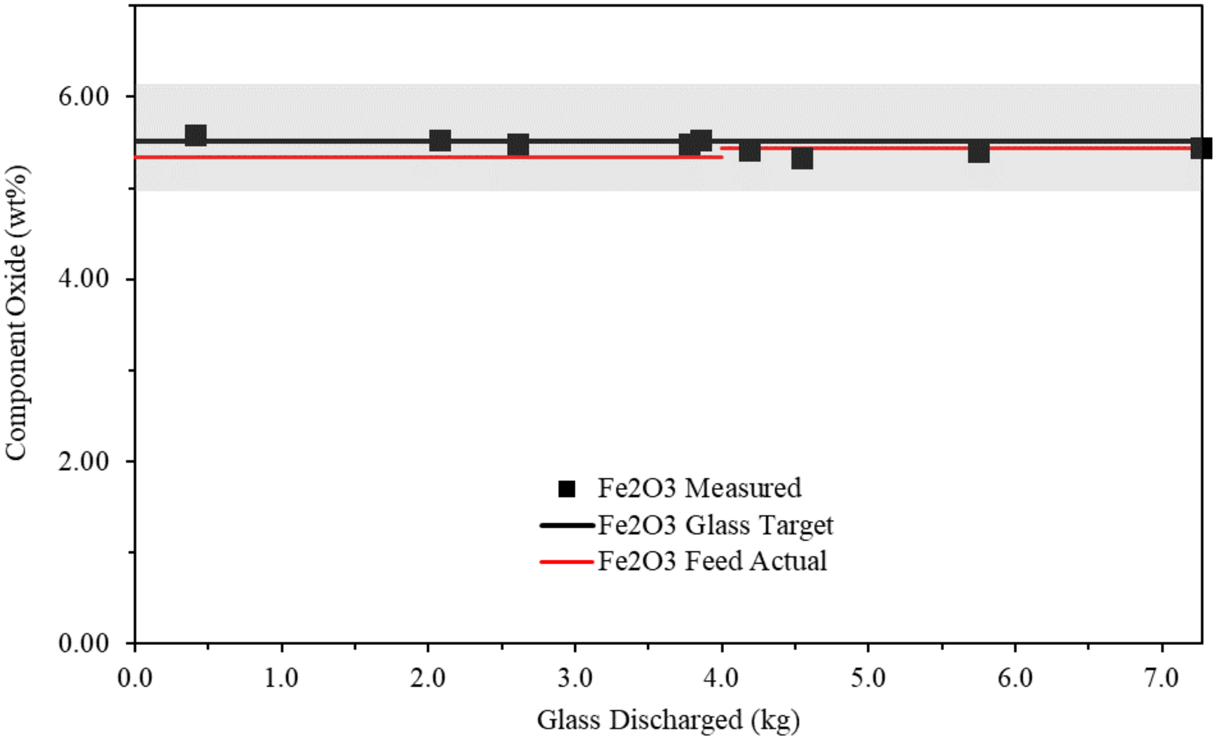

Figure A.11. Measured Fe_2O_3 content in the AP-107 recycle CLSM run glass product and melter feeds along with the Fe_2O_3 content target from the AP-107-1R glass composition.

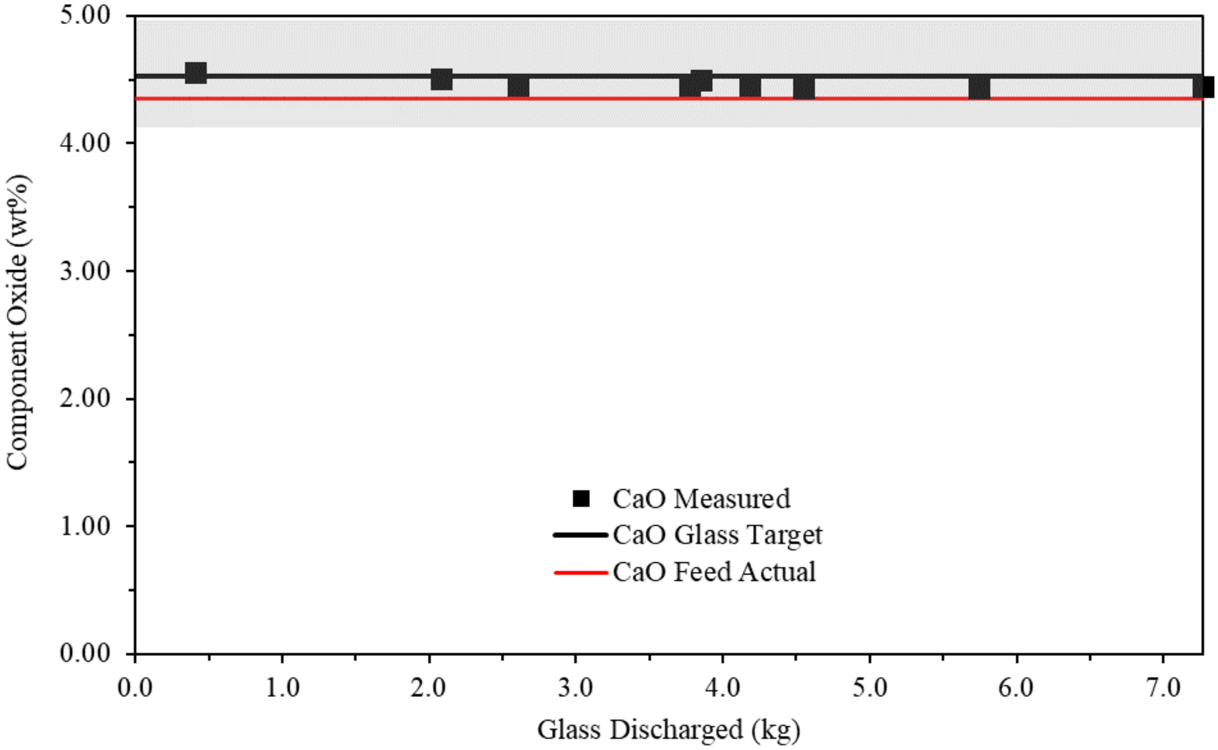

Figure A.12. Measured CaO content in the AP-107 recycle CLSM run glass product and melter feeds along with the CaO content target from the AP-107-1R glass composition.

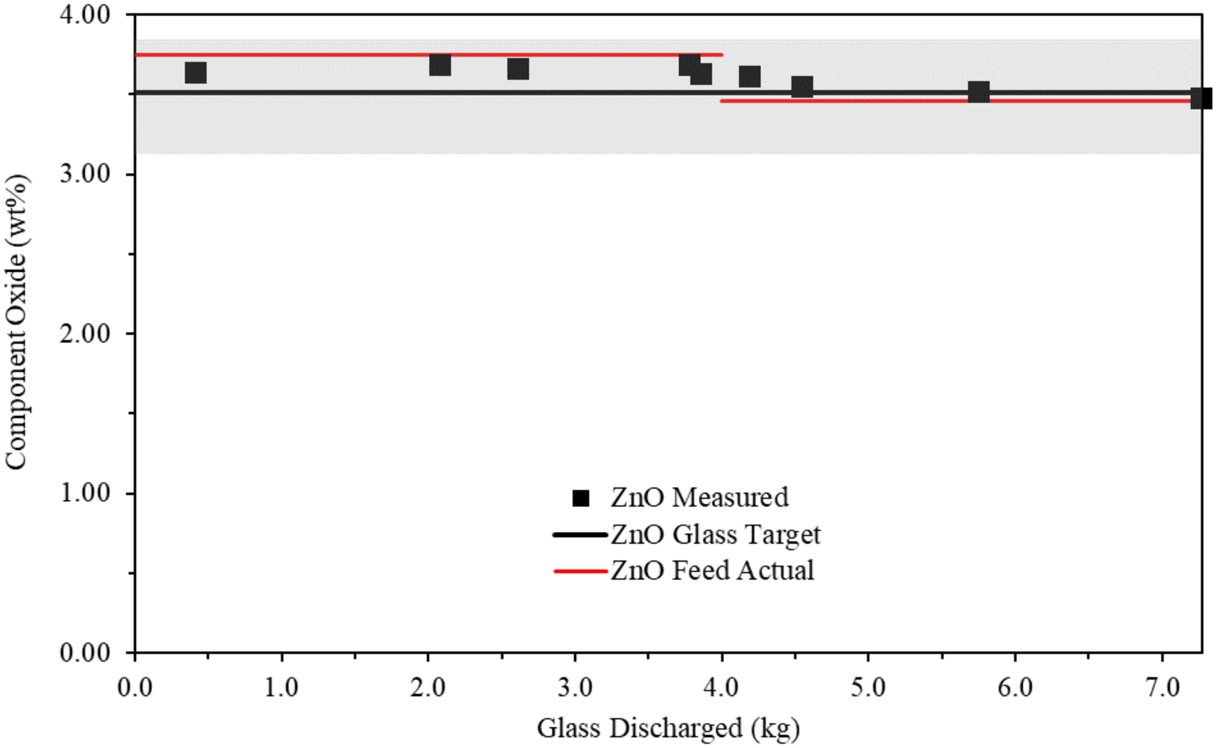

Figure A.13. Measured ZnO content in the AP-107 recycle CLSM run glass product and melter feeds along with the ZnO content target from the AP-107-1R glass composition.

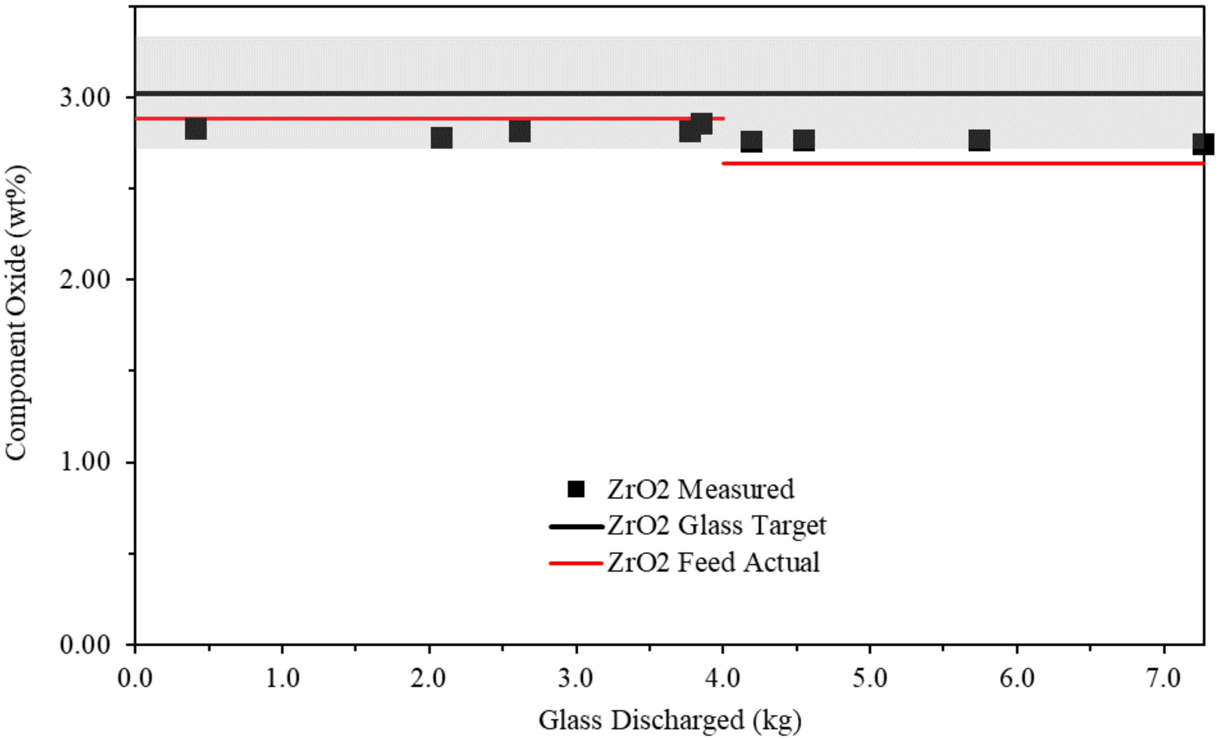

Figure A.14. Measured ZrO_2 content in the AP-107 recycle CLSM run glass product and melter feeds along with the ZrO_2 content target from the AP-107-1R glass composition.

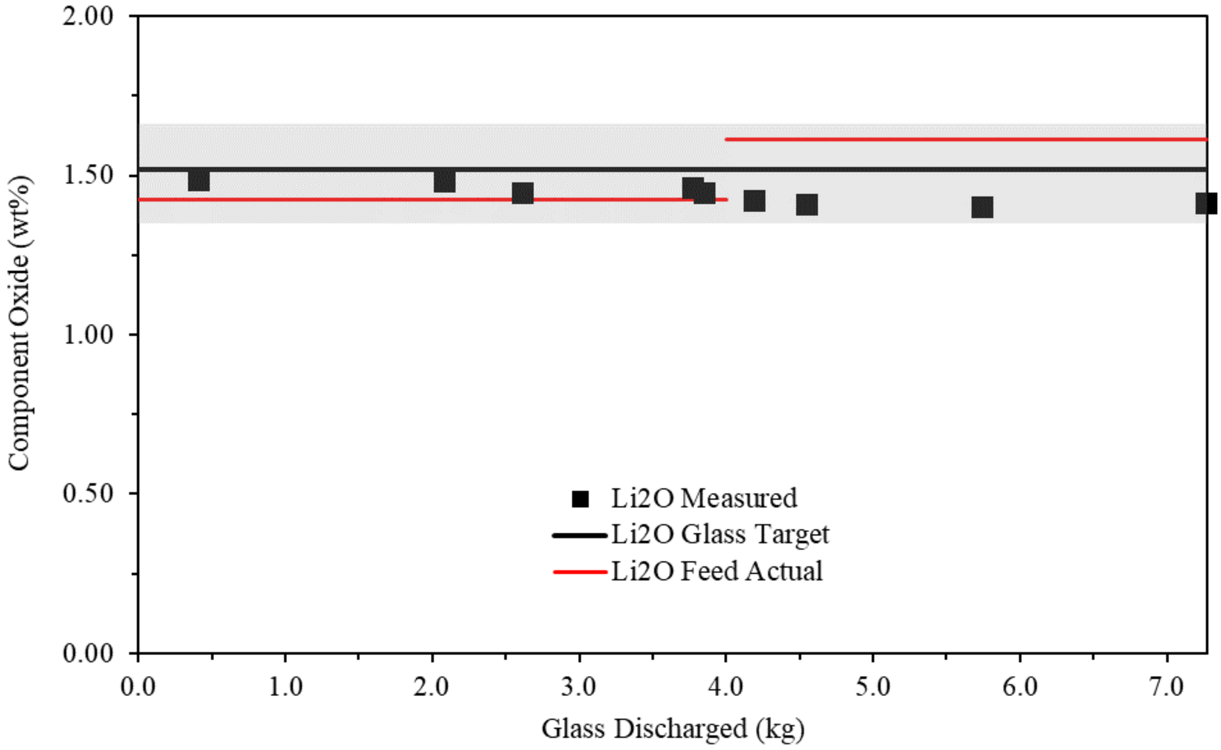

Figure A.15. Measured Li_2O content in the AP-107 recycle CLSM run glass product and melter feeds along with the Li_2O content target from the AP-107-1R glass composition.

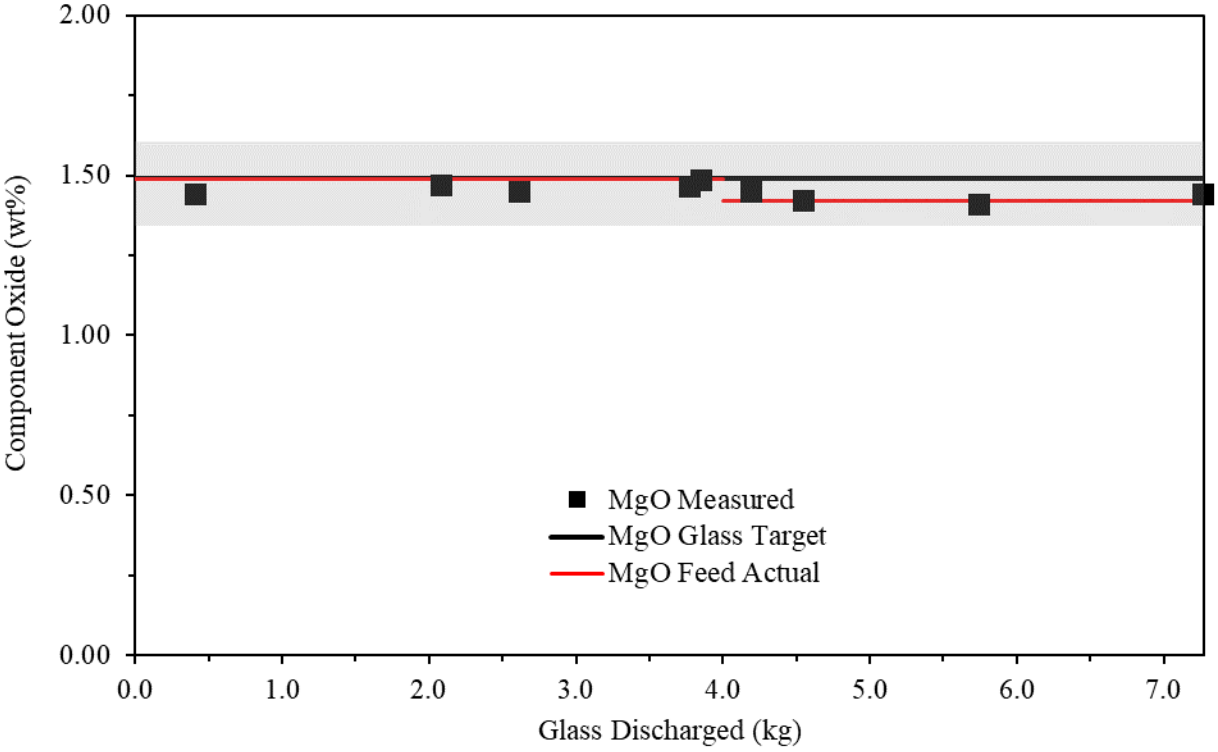

Figure A.16. Measured MgO content in the AP-107 recycle CLSM run glass product and melter feeds along with the MgO content target from the AP-107-1R glass composition.

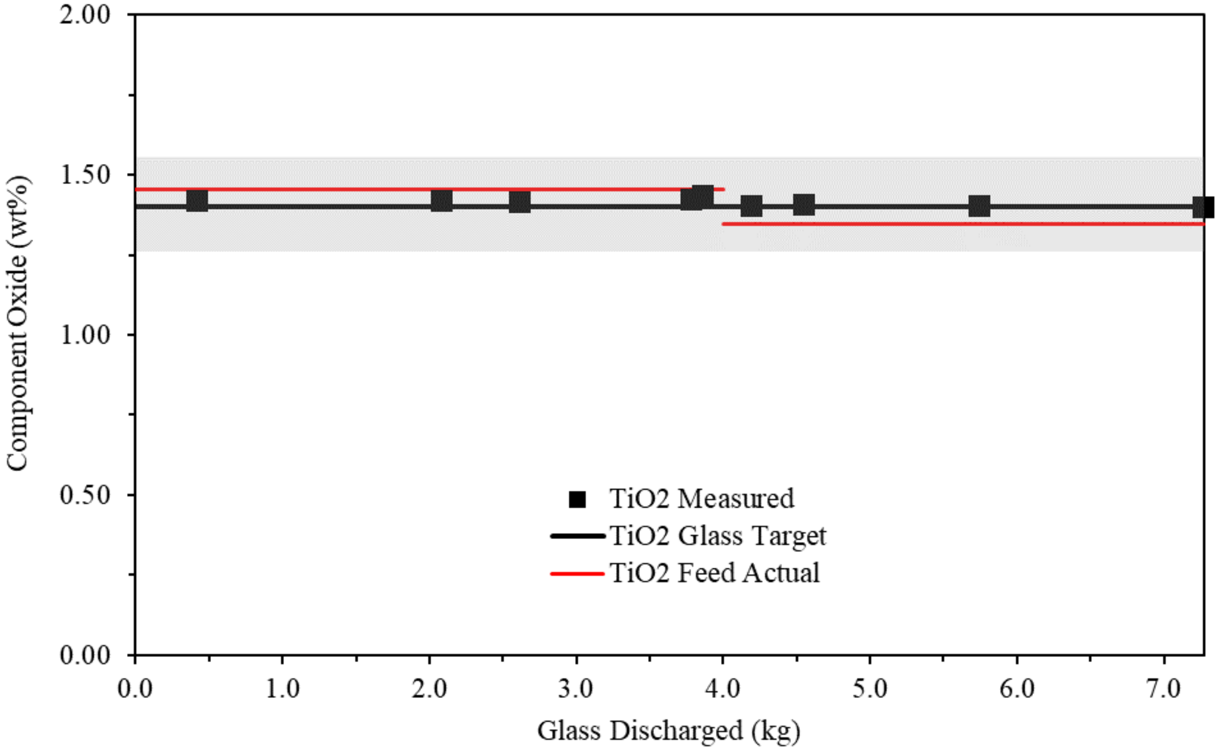

Figure A.17. Measured TiO_2 content in the AP-107 recycle CLSM run glass product and melter feeds along with the TiO_2 content target from the AP-107-1R glass composition.

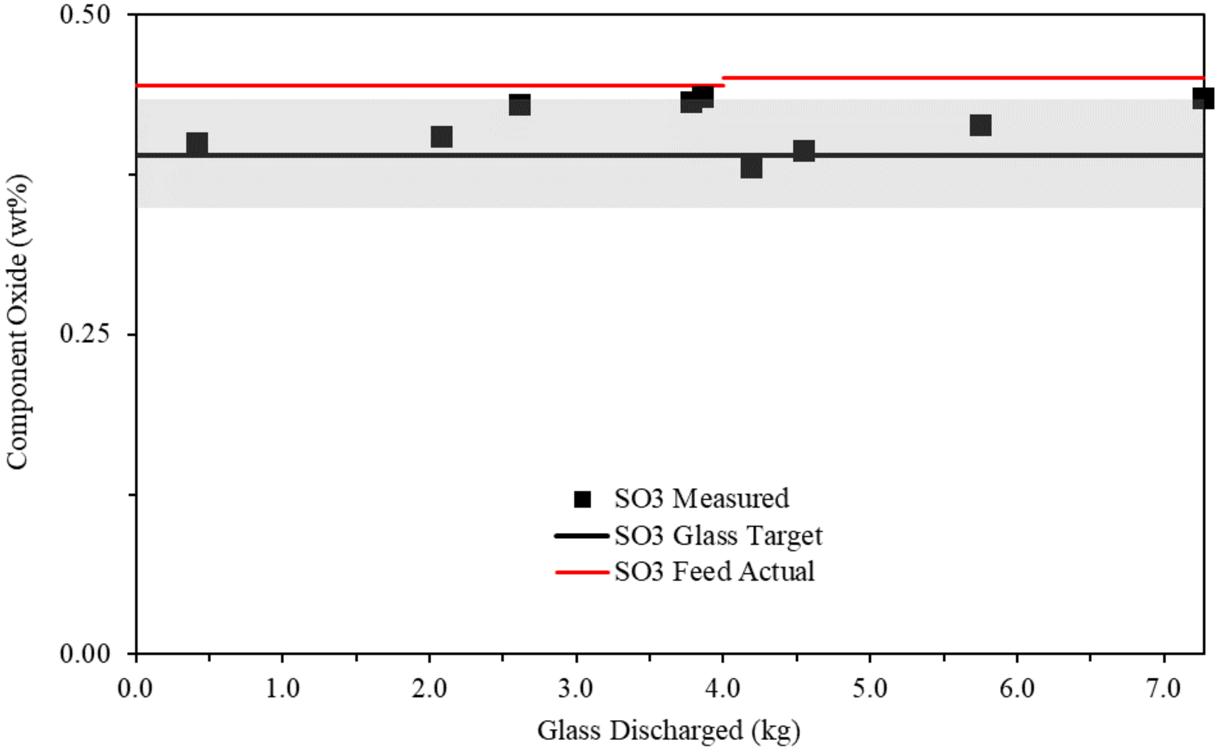

Figure A.18. Measured SO_3 content in the AP-107 recycle CLSM run glass product and melter feeds along with the SO_3 content target from the AP-107-1R glass composition.

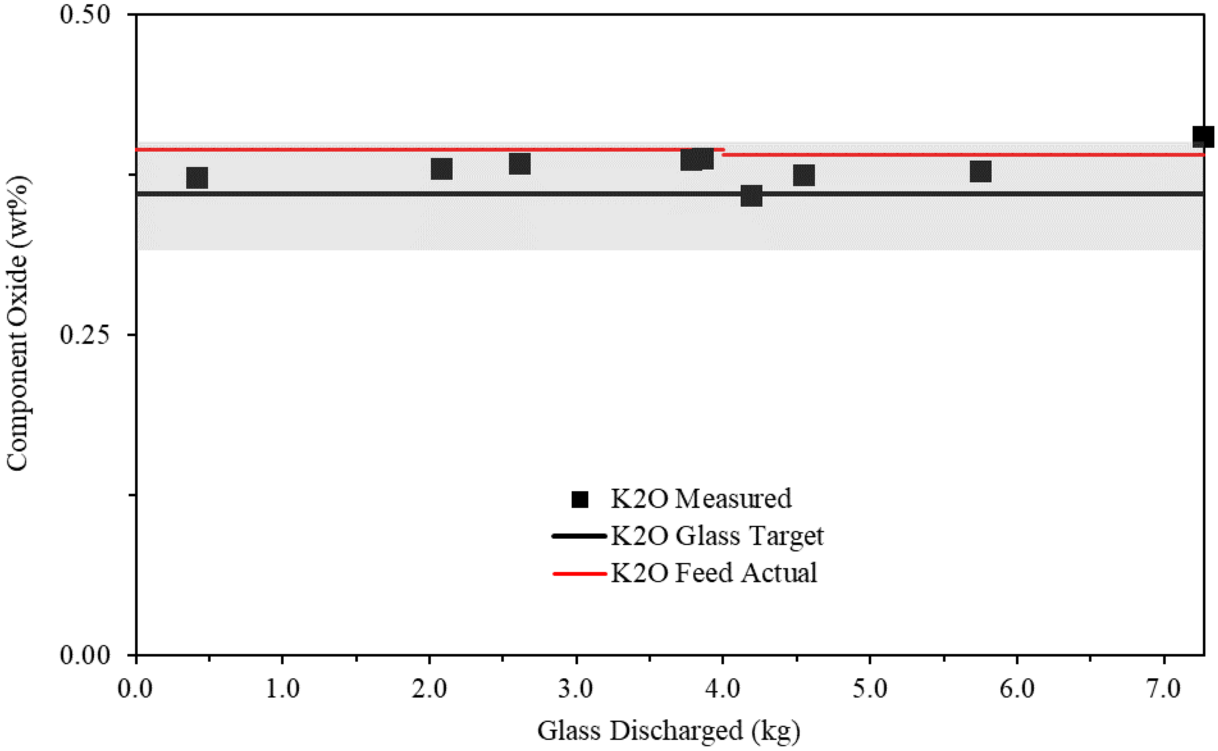

Figure A.19. Measured K_2O content in the AP-107 recycle CLSM run glass product and melter feeds along with the K_2O content target from the AP-107-1R glass composition.

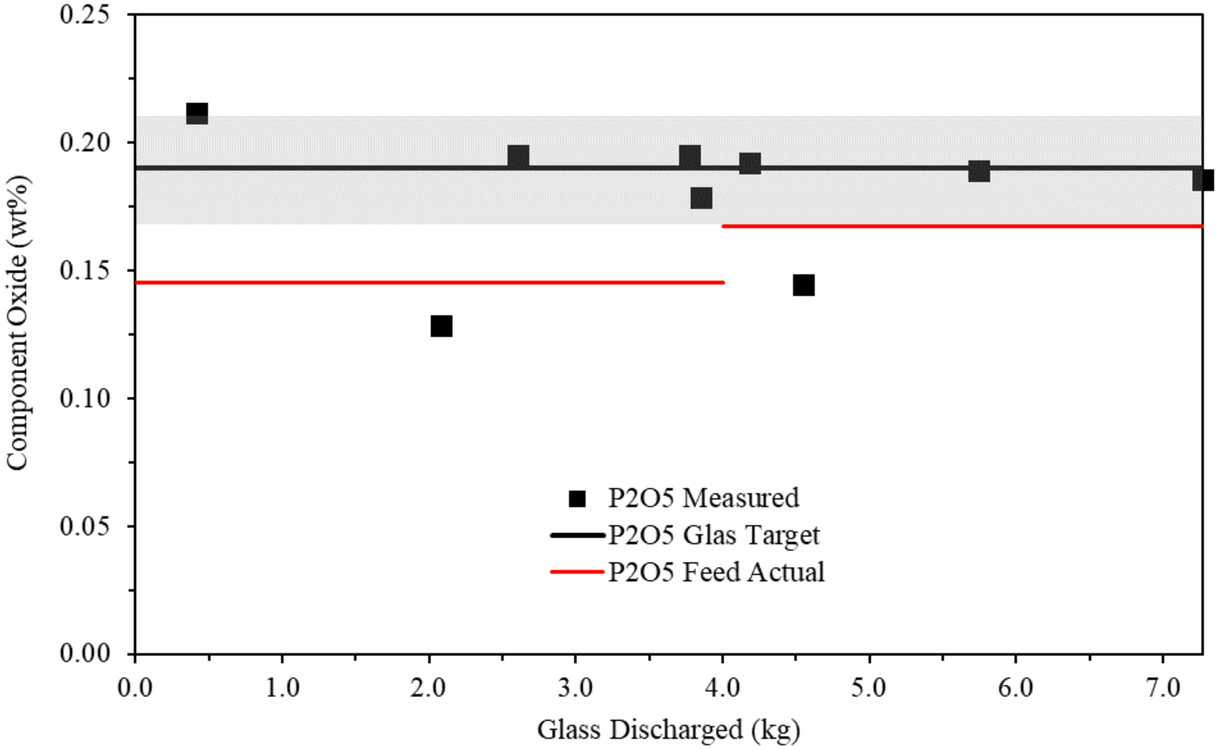

Figure A.20. Measured P_2O_5 content in the AP-107 recycle CLSM run glass product and melter feeds along with the P_2O_5 content target from the AP-107-1R glass composition.

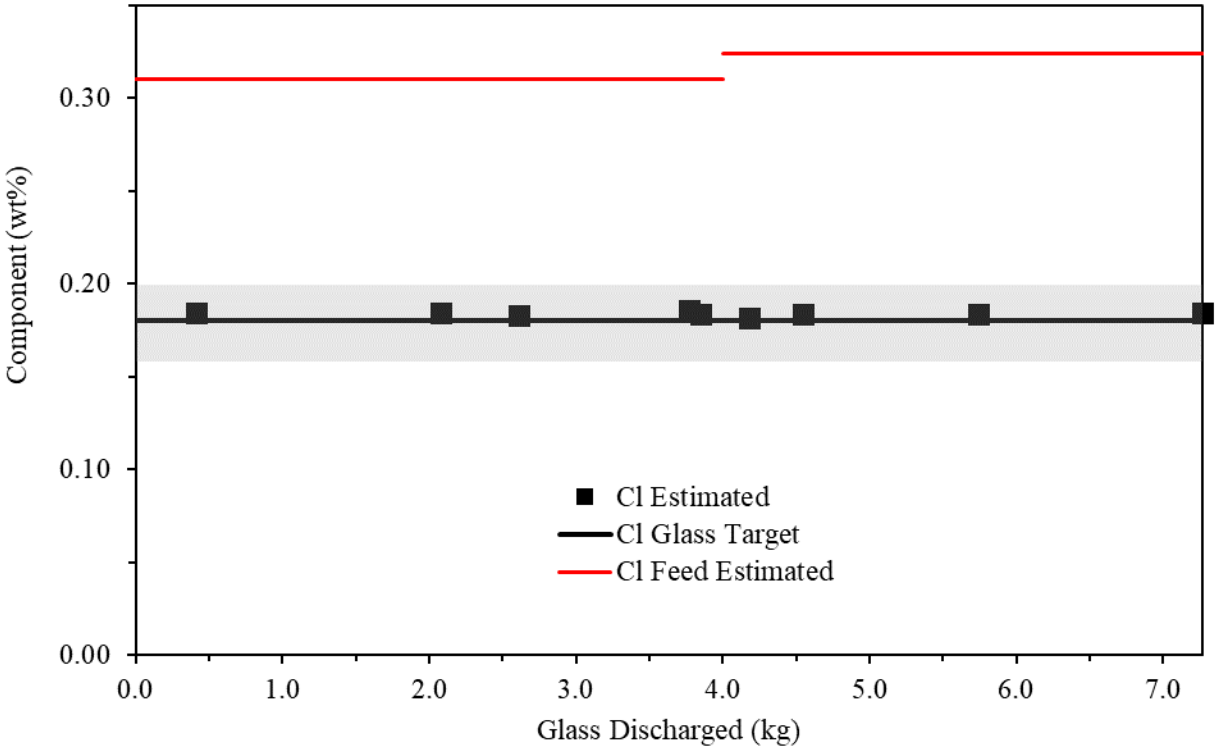

Figure A.21. Estimated Cl content in the AP-107 recycle CLSM run glass product and melter feeds (as described in Section 6.2.1) along with the Cl content target from the AP-107-1R glass composition.

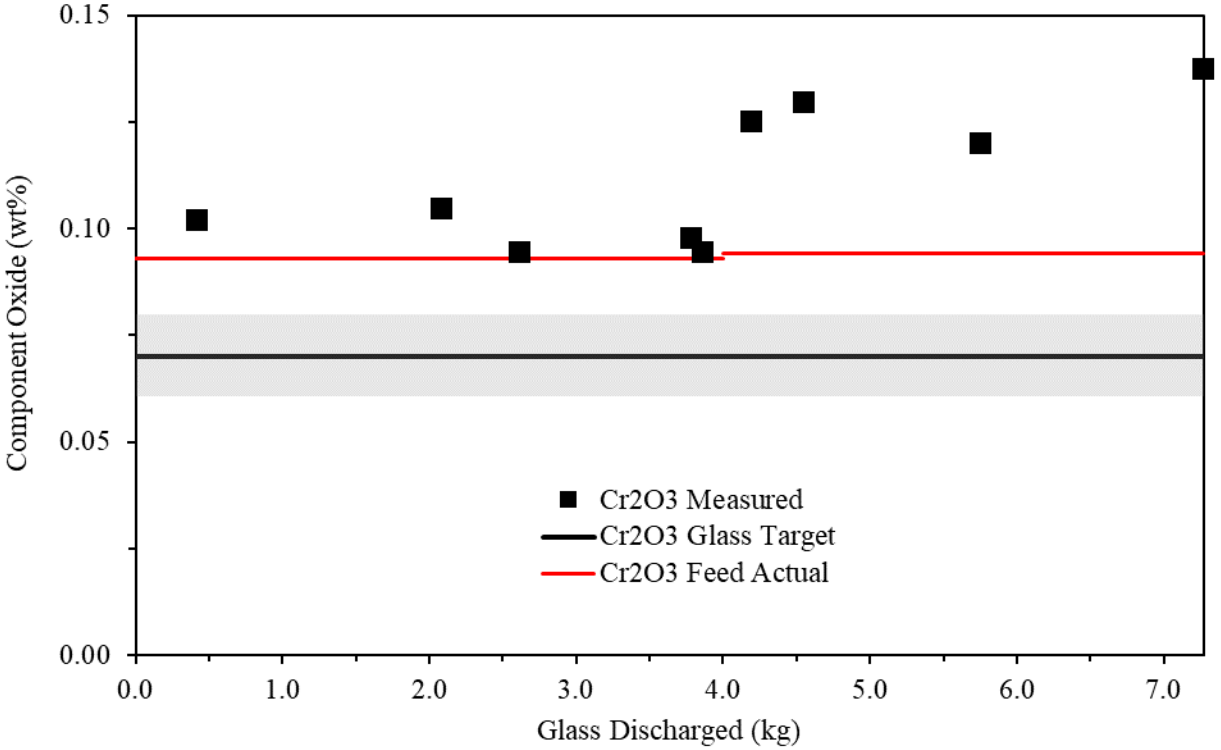

Figure A.22. Measured Cr_2O_3 content in the AP-107 recycle CLSM run glass product and melter feeds along with the Cr_2O_3 content target from the AP-107-1R glass composition.

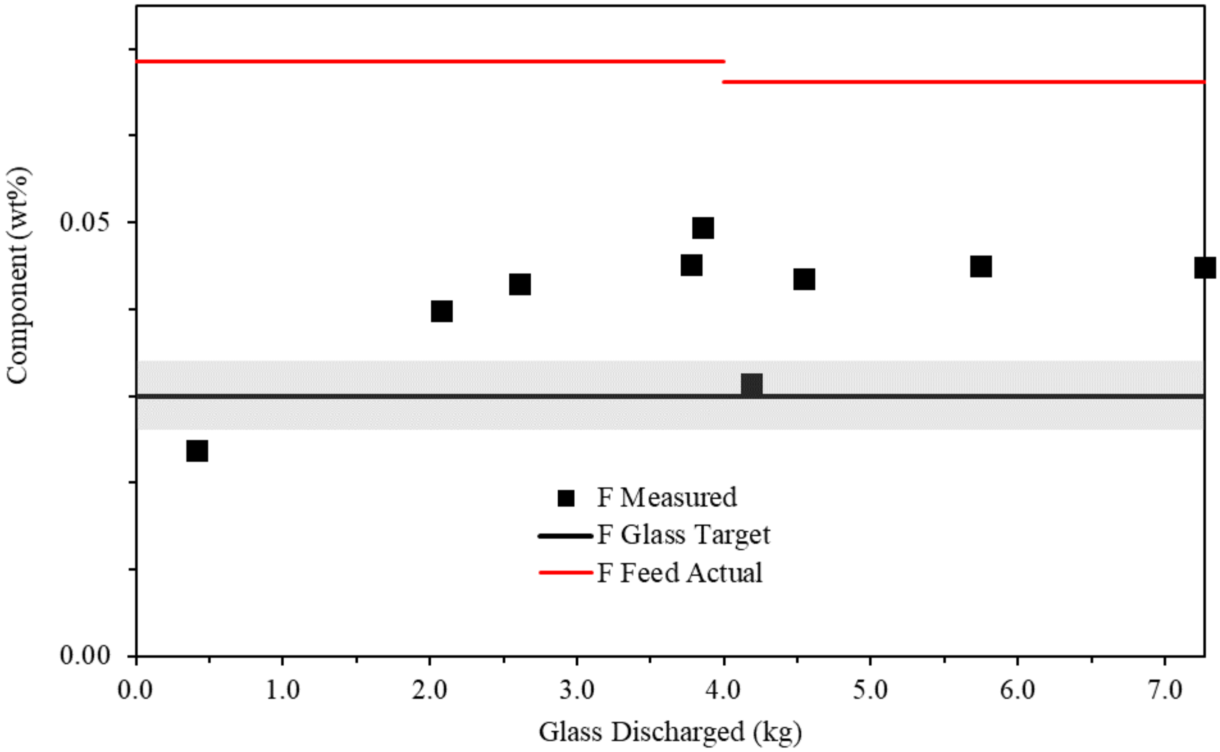

Figure A.23. Measured F content in the AP-107 recycle CLSM run glass product and melter feeds along with the F content target from the AP-107-1R glass composition.

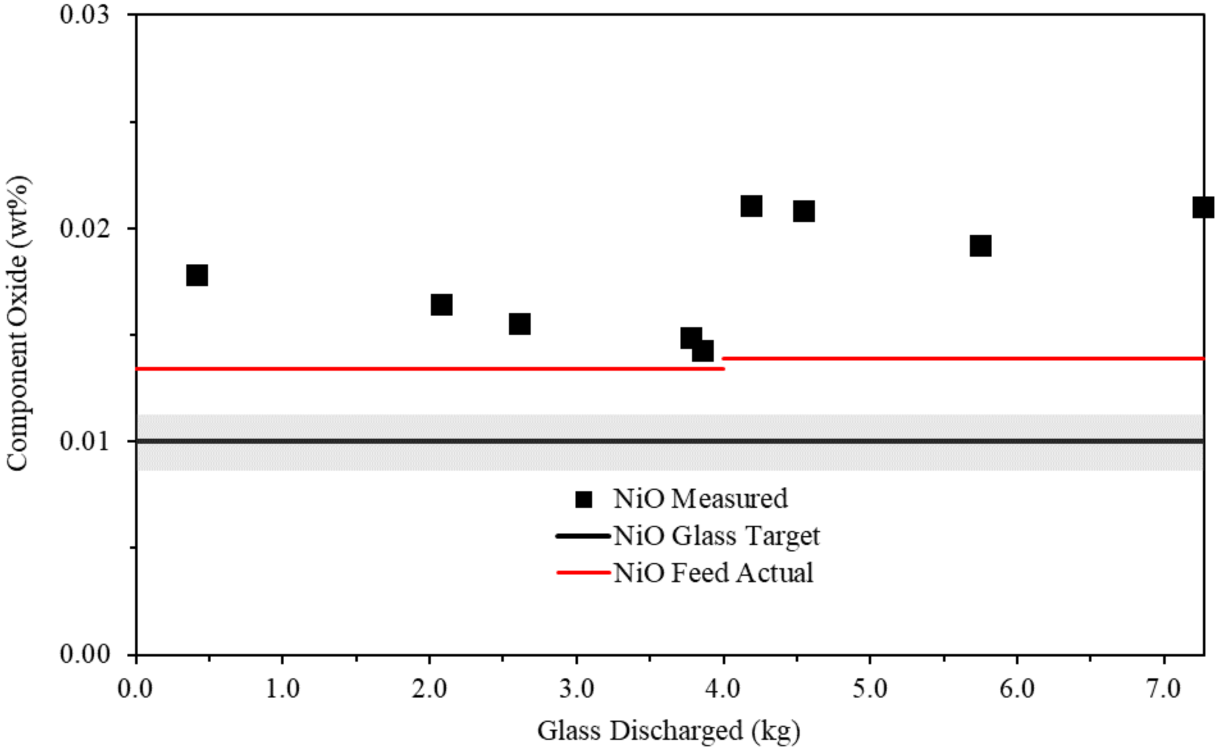

Figure A.24. Measured NiO content in the AP-107 recycle CLSM run glass product and melter feeds along with the NiO content target from the AP-107-1R glass composition.

Table A.2. Component Mass Flow Rates During the AP-107 Recycle CLSM Run

Component	$\dot{m}_{i,feed}$	$\dot{m}_{i,feed}$	$\dot{m}_{i,feed}$	$\dot{m}_{i,glass}$	$\dot{m}_{i,glass}$	$\dot{m}_{i,glass}$	$\dot{m}_{i,offgas}$	$\dot{m}_{i,offgas}$	$\dot{m}_{i,offgas}$
Sample Duration	Run	1	2	Run	1	2	Run	1	2
Units	mg min^{-1}	mg min^{-1}	mg min^{-1}	mg min^{-1}	mg min^{-1}	mg min^{-1}	mg min^{-1}	mg min^{-1}	mg min^{-1}
^{99}Tc	0.11	0.12	0.10	0.03	0.03	0.03	0.05	0.04	0.04
Cs	0.04	0.00	0.11	0.03	0.00	0.06	0.02	0.00	0.01
Al	455	483	470	430	452	466	1	0	0
B	423	435	460	391	420	446	5	2	2
Ca	448	469	474	432	448	469	1	0	0
Cl	45	--	--	25	--	--	9	--	--
Cr	9	10	10	11	9	13	1	0	0
F	10	10	10	7	6	6	1	0	0
Fe	542	564	578	518	539	550	2	0	0
K	47	49	49	44	45	46	2	1	1
Li	100	100	114	90	94	97	0	0	0
Mg	127	135	131	119	123	127	0	0	0
Na	1791	1864	1908	1726	1746	1854	23	10	10
Ni	2	2	2	2	2	2	0	0	0
P	10	10	11	10	12	9	0	0	0
S	26	27	27	23	24	23	2	0	0
Si	3087	3232	3261	2898	2985	3159	2	6	7
Ti	122	131	123	115	119	125	0	0	0
Zn	421	454	423	392	414	421	1	0	0
Zr	297	322	297	280	293	303	0	0	0
Total	7963	8297	8348	7519	7731	8118	50	21	22

The Cl values were not estimated during the sampling time periods and are marked with '--'.

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