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RPT-DFTP-002, Rev 0

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Functions and Requirements of the Radioactive Waste Test Platform

March 2017

RA Peterson
SK Fiskum
WC Eaton
J Geeting

U.S. DEPARTMENT OF ENERGY

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

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Pacific Northwest National Laboratory
Richland, Washington 99352

Acronyms and Abbreviations

CLSM	continuous laboratory-scale melter
CUF	cells unit filter
DFLAW	Direct Feed Low-Activity Waste
HEPA	high-efficiency particulate air
LAW	low-activity waste
LAWPS	Low-Activity Waste Pretreatment System
LFJHCM	liquid-fed joule-heated ceramic melter
LSM	laboratory-scale melter
P&ID	piping and instrumentation diagram
PNNL	Pacific Northwest National Laboratory
RCLSM	radioactive continuous laboratory-scale melter
RSM	research-scale melter
sRF	spherical resorcinol-formaldehyde
VSL	Vitreous State Laboratory
WAC	waste acceptance criteria
WTP	Hanford Tank Waste Treatment and Immobilization Plant

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1.0 Introduction

The purpose of this document is to provide the function and requirements for the Radioactive Waste Test Platform (hereafter called “test platform”). Historically, testing has been performed at the bench scale with actual waste samples (Fiskum et al. 2006a, b, 2008, 2009; Lumetta et al. 2009; Edwards et al. 2009; Smith et al. 2000, 2001). These tests form the basis for waste treatment operations at the Hanford Site. However, since 2008, there has been limited capability to perform bench-scale tests with actual waste samples. Prior work has shown that simulants can provide useful information about the performance of unit operations; however, there have been examples in the past where simulants did not adequately capture the expected performance of the tank waste materials. Therefore, as the Hanford Site moves towards initiating treatment of tank waste, it is important to have a platform available that can evaluate process options utilizing tank waste samples. This platform can be utilized to evaluate processing options and to address troubleshooting for unexpected system performance. The initial operations for the test platform will be to contribute towards maturing the Direct Feed Low-Activity Waste (DFLAW) feed qualification program and to demonstrate an option for disposition of the melter offgas stream. This equipment will be the ideal platform to evaluate additional process options including filtration operating conditions associated with precipitation, methods to extend resin lifetime associated with multi-cycle degradation, and evaluation of other approaches to minimize the impact of recycle streams from the effluent management system (including Tc) on melter performance.

The key unit operations for this platform include filtration, ion exchange, and melter operations. Therefore, Pacific Northwest National Laboratory (PNNL) will be installing these process units for radiological operations in its Radiochemical Processing Laboratory. This document provides the requirements, scaling basis, description, and process flow associated with the platform unit operations as well as future capabilities.

2.0 Requirements

An overall requirement is that the test platform be able to handle radioactive waste samples. The advantage of the test platform is that it can treat actual waste. The primary difference between waste and simulant tests is that there may be unknown chemistry associated with the actual waste that is not incorporated into the design of simulants. Thus, anything learned from the testing on actual waste will likely provide information on the waste compositional (and associated physical) properties. Therefore, most of the performance goals for the test platform are derived from the need to understand the chemical behavior effects on the performance of the integrated DFLAW process.

This set of requirements is associated with the test platform itself, not the requirements for a specific test (e.g., FY17 or FY18 testing scope). Not all requirements in this document need to be included in the initial version of the test platform used in FY17 and FY18 testing. The test platform can be expanded to grow capabilities in the future (see Section 6.0, "Future Capabilities").

The performance requirements for the test platform are divided into three unit operations: (1) filtration; (2) ion exchange; and (3) melter, melter offgas, and offgas condensate.

Overall Requirements

The test platform should be able to handle a sodium molarity between 4 and 8.

Filtration

- The filtration system shall be capable of providing approximately the same or greater feed tank volume/filter area ratio as the Low-Activity Waste Pretreatment System (LAWPS) design.
- The filtration system shall use a Mott filter with the same internal diameter (0.5-inch), thickness (1/16-inch), and retention grade (0.1-µm sintered metal) as the LAWPS design.
- The filter material should be 316L stainless steel to match the LAWPS design.
- To be consistent with the LAWPS design, flow through the crossflow filter should be continuously recirculated into and out of the feed tank.
- The LAWPS feed is pumped through the full-scale crossflow filter line at a little over 900 gpm (918 gpm for the base case in the LAWPS Mass and Energy Balance). The filter loop is an 8-inch line, so the axial velocity through the pipe going into the filter loop is 6.44 ft/s. The flowrate through the test platform crossflow filter line should be scaled so that the axial velocity in the filter is approximately the same as in the full-scale system.
- The filtration system should be able to remove solids using the Mott filter described above.
- The LAWPS is designed for a flowrate of 4 to 17 gpm, with a nominal flowrate of 9.48 gpm. The LAWPS filter area is 534 ft^2. Consequently, the flux per filter area is (9.48/534) = 0.0177 gallon/ft^2. The filtration system should be able to determine the transmembrane pressure required to meet that throughput per filter area and monitor changes with time (the length of time being defined by the amount of sample material available for a given test).

- The maximum transmembrane pressure in the LAWPS facility is 100 psid, so the filtration system needs to be able to provide that maximum pressure across the membrane.
- The filtration system should be able to introduce cleaning solutions (see below), either through the filter feed tank or back through the shell side of the crossflow filter. When cleaning solution is introduced through the shell side of the filter, the filtration system should be able to be drained to another tank or jar other than the filter feed tank, to match the LAWPS design.
- The LAWPS facility has the ability to clean the filter with 0.45M HNO_3, 3M HNO_3, or saturated oxalic acid. The test platform filtration system should therefore have the ability to clean the filter with any of these reagents.
- The LAWPS facility currently has space set aside for a backpulse system, while the need for this system is under consideration. The filtration system should have the ability to add a backpulse system, if required, in the future.
- The LAWPS uses temperature to control precipitation and optimizes the temperature to maximize ion exchange efficiency. The optimal temperature to avoid precipitation is determined up front with a feed sample.
- The filtration system should be able to control the temperature of the feed tank to the target temperature determined above for each sample.
- The filtration system should be designed so that samples from both the filter feed tank and the filter permeate can be taken.
- The filter should be easily removable and replaceable so that a post-test analysis of the filter can be undertaken, if required by a given test objective.

Ion Exchange

- The ion exchange system should be able to control the temperature to the target temperature within 5 °F of the target feed temperature when waste feed is in the system. The ion exchange system should be able to control temperature to a target temperature when reagents are in the system so that the effect of temperature on reagent performance can be evaluated when necessary.
- The ion exchange system should be designed to minimize oxygen entrainment to prevent resin degradation.
- The ion exchange system shall remove cesium using spherical resorcinol-formaldehyde (sRF) resin.
- The ion exchange system should have two identical vertical ion exchange columns operated in series.
- The ion exchange system columns should be removable and replaceable so that different column sizes and pressure tolerance capabilities can be investigated if required by an individual test objective.
- Each ion exchange column used in the LAWPS design has a sRF resin bed diameter of approximately 3.5 feet, a bed length of 4.2 feet, and a volume of approximately 308 gallons (41.2 ft^3) in sodium form. The vessel has a straight side length of approximately 7 feet and a vessel volume of approximately 593 gallons. The total column height accounts for the resin expansion, the liquid distributor/collector, and allowance for head/tail space. The flowrate of the LAWPS facility ranges between 4 and 17 gpm, with a nominal flowrate of 8.7 gpm. In the ion exchange system, the waste should flow through the columns at a rate such that the waste is in contact with the resin for the same

period of time as in the full plant. The flowrate should be able to be varied so that the full range of expected flowrates seen in the plant can be evaluated (4 to 17 gpm).

- The ion exchange system should be able to monitor for cesium breakthrough in the columns. The target breakthrough endpoint is 10% of the waste acceptance criteria (WAC) for Hanford Tank Waste Treatment and Immobilization Plant (WTP) low-activity waste (LAW) at the end of the second column. The WAC is 3.18×10^{-05} Ci of Cs-137 per mole of sodium. The concentration can momentarily exceed 10% of that value coming out of the column for the purposes of identifying where the breakthrough point is, but the final treated feed passed to the test platform melter must be below the WTP-WAC value.
- The LAWPS uses backpressure on the column applied using a pressure control valve in order to control gas solubility. The initial construction of the test platform does not need to include this backpressure, but it does need to be designed so that capability can be easily added in the future.
- The ion exchange system should be designed so that a sample of the resin can be taken after any elution or regeneration step. Strategies that stop the test at the sample point and sacrifice the whole column are acceptable. It is anticipated that a sample of the spent resin will routinely be taken after resin regeneration in order to determine residual constituents left on the resin by the elution cycle.
- Table 1 shows the full-scale LAWPS resin regeneration and replacement steps. The ion exchange system needs to be able to perform each of those steps. The flowrate through the column needs to be determined so that each gram of resin comes in contact with the same mass of reagent as in the full scale for the same period of time.

Table 1. Full-Scale LAWPS Resin Elution and Regeneration Steps

Step	Action (Note all volumes are per column, so the volume needs to be doubled for two columns)	Upflow or Downflow?
1	Displacement of the filtrate present in the ion exchange column with 0.1M NaOH. This is performed with 1.5 column volumes of the NaOH at a rate of 3 bed volumes/hr.	Down
2	Pre-elution rinse with demineralized water to remove the 0.1M NaOH. This is achieved with 1 column volume at a flowrate of 3 bed volumes/hr.	Down
3	Elution of the sRF resin with 0.45M HNO_3. This is achieved with 15 bed volumes of acid through the bed. The flowrate will be at 1.4 bed volumes/hr.	Down
4	Post-elution water rinse to remove HNO_3. This is done with 1.3 column volumes of demineralized water at a flowrate of 1.4 bed volumes/hr.	Down
5	Regeneration of the resin with 1M NaOH. This will be achieved with 3 bed volumes of base at a velocity of 12.4 cm/min. This is followed with 0.3 bed volumes at a velocity of 1.96 cm/min. Thus, the total volume of regeneration solution through the bed is 3.3 bed volumes, though the actual volume used to drive the 3.3 bed volumes through the resin bed will be larger to account for the larger vessel volume and the connecting pipe.	Up

- In Table 1, Steps 1 and 2 are processed from lead column to lag column. This means that the reagent is introduced at the top of the lead column, and the effluent that comes out the bottom of the lead column is routed to the top of the lag column. Blending the effluent from Step 1 with the treated waste feed is anticipated; however, this is currently being evaluated. Thus, the test platform needs to be able to either blend this effluent with the treated feed or plan alternate disposal. The effluent from

Step 2 is not currently planned to be blended with the treated feed at LAWPS, so the capability of isolating this effluent from the treated feed in the test platform should be realized.

- In Table 1, Steps 3, 4, and 5 are processed from the lag column to the lead column. This means the reagent is introduced at the top of the lag column and the effluent from the bottom of the lag column is routed to the top of the lead column. The effluents from these processes should not be blended in with the treated feed in normal conditions.

Melter, Melter Offgas, and Offgas Condensate

The design input from this section came from an email[1] to the author that provides the relevant WTP design information for the Radioactive Waste Test Platform. That same memorandum provides information on potential testing scope, but that is not covered here because this performance requirements document only covers the requirements of the test platform itself.

- The melter needs to vitrify the waste feed sample treated by filtration and ion exchange into glass in a continuous manner.
- The offgas treatment system needs to quench the offgas and collect the condensate.
- The melter and offgas combination needs to be designed such that a reasonable estimate of the distribution of all major components between glass, offgas condensate, offgas particulates, and offgas vapor can be made.
- The glass pool temperature should be approximately 1150 °C (± 30 °C).
- The WTP melter plenum temperature is 450 °C, and the melter system should try to get as close to this temperature as reasonably possible. Plenum temperatures up to 650 °C are acceptable.
- While it is not anticipated that the condensate will be recycled back into the feed in the melter system for the initial uses, the design should allow for future modifications for this.
- The model used for formulating glass should be the 2009 LAW Glass Models (collectively known as LAW Glass Shell V2.0). The chemical composition inputs to this glass algorithm need to be collected on the feed someplace (potentially upstream).
- The test platform melter should be able to meet the WTP design rate of 1500 kg/m^2-day and the throughput rates observed at pilot testing, which are approximately 2200 kg/m^2-day. The exact throughput rate for a specific test will be test-objective-specific.
- The WTP melters are bubbled to promote mixing. Provisions to add bubblers should be included in the test platform melter design. The bubbling rate will be determined by the throughput rate.
- The test platform melter system should maintain the ability to sample glass, melter feed, offgas condensate, and offgas particulates, and have some method of determining any volatile components that were not collected in the condensate, for mass balance purposes.
- The melter system should have the ability to be idled so idling tests can be performed while still monitoring for the mass balance of volatile components.

[1] Benson P. 2016. Personal Communication: Email from P Benson (AECOM) to J Reynolds (WRPS) and W Eaton (PNNL), *WTP Plant Engineering Input to DFLAW Radioactive Test Platform - Melter Operating Parameters (CCN 293262)*, November 14, 2016. WRPS-1605347, Washington River Protection Solutions, Richland, Washington.

- The test platform should have the ability to determine if a salt gall layer has formed on the melt surface. Test strategies that quench the melt to look for this gall layer are acceptable.

3.0 Scaling Basis

The Radioactive Waste Test Platform at PNNL is designed to support startup and operation of the WTP, LAWPS, and other chemical/nuclear processing facilities as a test platform for processing actual tank waste as well as simulants containing radioactive materials. As such, the scale of the unit operations is selected so that the equipment can be adequately operated in either hot cells or fume hoods/enclosures within the Radiochemical Processing Laboratory and so that meaningful data and results can be obtained with amounts of tank waste or simulated radioactive material on the order of several liters.

3.1 Crossflow Filtration Scaling Basis

Full-scale filtration typically uses bundles of filters that are 8 to 10 feet long. The filter bundles contain many filter tubes—typically more than 100 per bundle. Each tube is characterized by the internal diameter, wall thickness, and pore size rating. To avoid issues associated with differences in tube dimensions, the prototypic tube dimensions are used in bench-scale testing. However, shorter, single tubes are used in bench-scale processes. Previous work with simulants in the Pretreatment Engineering Platform has demonstrated that the performance for full-length bundles is approximately the same as for single 2-foot filter elements when represented as a filter flux (permeate rate/surface area) (Daniel et al. 2010).

The cells unit filter (CUF) will match the following filter characteristics planned for LAWPS:

1. Filter media (0.1-μm sintered stainless steel, 1/8-inch-thick filter media) – Matches filtration efficacy and filter media hydraulic resistance
2. Filter internal diameter (0.5-inch) – Maintains the cross-sectional surface area and boundary layer filter cake effects at planned velocities
3. Crossflow velocity (14.7 ft/s)

Filter lengths are not scaled. Standard filters are available with 6- to 24-inch active lengths. The 6-inch length has been tentatively selected to better match the filter feed/filter area ratio, as shown in Table 2.

Table 2. Comparison of Feed Volumes and Filter Area at Different Scales

Test Component	LAWPS Baseline Sizing	1/9th Scale Test Configuration	CUF 6-inch Filter Configuration	CUF 2-foot Filter Configuration
Simulant tank (gal)	1,000,000	6500	NA	NA
Filter feed tank (gal)	6467	700	1.1	1.1
Filter area (ft^2)	532	62.8	1/16 (6-inch filter element)	1/4
Filter feed tank volume/filter area (cm^3/cm^2)	50	45	66	16
Prototypic filter flux (0.016 gpm/ft^2)	9 gpm	1 gpm	0.25 liter/hour[(a)]	1 liter/hour

(a) Note that the CUF filter flux with a 6-inch filter is expected to be an order of magnitude higher than the targeted feed rate to the in-cell ion exchange column of approximately18 mL/h. The filter flux could be throttled and controlled to a lower rate by installing a mass flow controller; however, this design capability has been deferred per direction from Washington River Protection Solutions.

One key aspect of scaling filtration behavior is to attempt to maintain a similar ratio of feed vessel size (volume) to filter area (assuming similar solids concentrations at the different scales). This is balanced by the need to have a sufficiently long filter tube to avoid entrance and exit effects on filtration performance.

As indicated in Table 2, the 6-inch-long filter element more closely matches the full-scale filter feed tank-volume-to-filter-area ratio (66 vs. 50 cm^3/cm^2), and given the low amount of solids expected in the AP-105 feed (first feed slated to be tested), the 6-inch filter is recommended. However, it is worth noting that the 2-foot-long filter elements have been demonstrated to provide sufficient length to generally avoid measurable entrance effects (Daniel et al. 2010). This demonstration has not been performed for the 6-inch filter.

The CUF is designed to provide flexibility, and filter lengths between 6 inches and 2 feet may be readily installed and tested. With an appropriate match of volume-to-surface-area ratio, the other important variables to match are the crossflow velocity and the transmembrane pressure. These will be test parameters for the test platform.

3.2 Ion Exchange Column Scaling Basis

For the ion exchange testing, the minimum scale of testing is defined by the resin bead diameter. The objective is to have a sufficient column diameter to ensure that there are minimal wall effects on column performance. The resin bed diameter will be at least 37 bead diameters (based on a 1.5-cm inside diameter column). This should limit the wall effects on column performance where a minimum of 30 bead diameters was recommended (Helfferich 1962). A larger column would provide a further reduction of wall effects, but would increase the volume of feed required beyond what is currently practical. Further, it is desirable to maintain an aspect ratio near that of the full-size column. (A 10-mL resin bed in a 1.5-cm-diameter column will provide an aspect ratio of 3.8.) Also, with approximately 4 L of feed, this will provide ~400 bed volumes of feed for the column. Based on the expected cesium, potassium, and sodium concentrations and previous experience with AN-102 tank waste (Fiskum et al.

2006b), it may be possible to process up to 5.0 L prior to achieving 50% breakthrough on the lead column.

3.3 Melter Scaling Basis

3.3.1 Melter Feed

The melter feed system for the test platform is designed to support laboratory-scale melter (LSM) testing and is not prototypic of the production melter feed system. In a fume hood in the Radiochemical Processing Laboratory, glass-forming and modifying additives will be added to the treated LAW waste that has been transported from the hot cell. Glass formulation will be in accordance with the specified WTP glass model. The solids will be kept in suspension using mechanical agitation. The melter feed will be fed to the melter from a 1- to 2-L agitated feed vessel using a progressive cavity pump. Although this pumping method is not prototypic of the air displacement slurry pump in the production plant, it has been demonstrated to drip feed at a scaled, controlled rate on the cold cap in an LSM, forming a liquid feed pool under the feed nozzle, which is very similar to what is seen in larger liquid-fed joule-heated ceramic melters (LFJHCMs).

3.3.2 Melter

Scaling the dynamic process of converting slurry feed into molten glass in a melter is generally done using the surface area of the glass melt.[1] While surface area provides a rough scaling factor, complicated factors that are much more difficult to scale influence the conversion process taking place at the glass/cold cap interface. Significant influence on the conversion process includes the rate and size of the bubbles that impinge on the bottom of the cold cap and the amount/velocity of hot molten glass that is transported from within the melt to the bottom of the cold cap due to the bubbling action and other convective processes.[2] Depending on design, wall effects can also significantly influence the dynamic process.

The design objective for the radioactive continuous laboratory-scale melter (RCLSM), the melter in the test platform, is to balance the need to obtain meaningful data from a relatively small amount of waste feed with the need to adequately represent the various process mechanisms that influence the rate and incorporation of the individual feed components into the glass.

Fortunately, the development of LFJHCM technology for vitrification of radioactive waste has been well documented over the past five decades. There have been many tests on multiple test melters from which information on scaling and the influence of various mechanisms that affect the conversion of liquid waste feed to glass can be used for design of test systems. It is also well established that valuable data can be obtained using test systems with three orders of magnitude smaller surface areas than the production melters they represent. While the more recent test systems, which have provided the bulk of the test data

[1] Matlack KS, WK Kot, F Perez-Cardenas, and IL Pegg. 2000. *Determination of the Processing Rate of RPP-WTP Simulants Using a DuraMelter 1000 Vitrification System*. VSL-00R2590-2, Vitreous State Laboratory at the Catholic University of America, Washington, DC.
[2] Chapman CC. 2004. *Investigation of Glass Bubbling and Increased Production Rate.* REP-RPP-069, Duratek, Inc. Columbia, Maryland.

to underpin the WTP vitrification process and design of the WTP melters, have been scaled-down LFJHCMs, there are limits to how small ceramic test melters can be designed to be representative of the production melters.

PNNL has successfully designed test melter systems that incorporate a melter vessel within a furnace so that prototypic ceramic melter wall designs, with their inherent "cold wall" effect, do not need to be used to contain the glass melt (Whittington et al. 1996). PNNL has used fused silica melter vessels (approximately 4- to 5-inch diameter) for short, dynamic tests in the LSM system, Inconel melter vessels (approximately 2.5-inch diameter) for small, longer tests in a hot cell, and K3 lined Inconel vessels (6- to 10-inch diameter) for larger, longer tests in the research-scale melter (RSM) system. Recent work in fused silica LSM vessels has established that a representative cold cap can be formed in approximately half an hour with liquid melter feed at prototypic processing rates; however, the limited duration due to the quartz melter vessel prevents reaching chemical steady state between the melt and the cold cap. Previous tests in LSMs indicate that the design processing rate of 1500 kg/m^2-day as well as more recent processing rates of 2200 kg/m^2-day are achievable (Dixon 2016).

The RCLSM in the test platform will be fabricated from Inconel 690, which allows for longer tests and glass discharge. The ability to process three or more melter volume turnovers allows chemical steady state to be established between the melt and the cold cap to provide accurate split factors for the dynamic melting process. Although the continuous laboratory-scale melter (CLSM) and the RCLSM are designed for continuous operation, they can be idled to mimic other plant operational modes. The approximately 5-inch-diameter melter vessel was selected based on the successful work studying cold cap dynamics mentioned above, and evaluation of the data available on the DM10 and DM100 melter test runs at the Vitreous State Laboratory (VSL) at the Catholic University of America as well as test runs of RSMs at PNNL. The approximately 2.5-inch melt pool depth was selected to provide enough depth to adequately represent the mechanisms that provide heat to the cold cap and help convert the cold cap to molten glass (bubbling and flow of hot molten glass to the cold cap interface via roll cells). Experience from test and production melters in addition to preliminary computational fluid dynamics models indicate that a 2.5-inch depth for a 5-inch-diameter melt pool is near to the smallest scale at which these mechanisms can be adequately represented.

3.3.3 Offgas

The test platform offgas system, while not prototypic of the production plant offgas system, is designed to support the testing objectives of the LSMs in both radioactive and non-radioactive environments. A small vacuum is pulled on the melter vessel by the offgas system, drawing off components that are partitioned to the offgas from the dynamic melting process. The scale of the LSM allows for the full offgas stream to be sampled for a short period (about 10 to 15 minutes) during steady-state melter operations. This sample train consists of heated high-efficiency particulate air (HEPA) filters followed by a submerged bed scrubber. Full offgas stream sampling provides for accurate analysis of the offgas from the melter. During periods when the offgas is not being sampled, it is drawn through a submerged bed scrubber, condenser, and a HEPA filter so that the treated offgas can be exhausted to the hood exhaust system. The condensate from the offgas system is captured, allowing for mass balance evaluation across the complete melter system over the entire test period. In the test platform, the offgas condensate is captured, evaporated, and solidified into an acceptable secondary waste product. Although

the test scope in FY17/18 does not include recycling of offgas condensate back to the melter feed system, the test platform could be configured to include this process step in the future.

3.3.4 Confirmation of Performance and Scaling

While design and scaling of the melter system within the test platform are based on experience, prior melter test data, and technical understanding of the influential mechanisms in the dynamic melting process, correlation of the LSM system performance is ongoing. Investigations of cold cap dynamics and partitioning of Tc (Re as a surrogate) have led to the need for continuous processing in an Inconel melter vessel (CLSM). To benchmark those investigations against available test data, correlation tests in the 5-inch-diameter CLSM are planned and are directly funded by the U.S. Department of Energy Office River Protection. Results from those tests will be correlated with test data reported for other test melter systems to confirm that the scaling and design decisions made for the CLSM and RCLSM were appropriate.

The final step in correlating the test data from the RCLSM would be to run correlation tests in the non-radioactive CLSM test system with simulated AP-105 waste. Running these correlation tests would allow for the results from the real AP-105 waste tests in the RCLSM planned in FY18 to be compared with the results from simulant test in the CLSM, providing a direct comparison of radioactive waste to simulant on the same melter platform. In addition, running simulant tests in the CLSM with the same AP-105 waste simulant planned for melter testing at VSL in FY17 would provide a direct comparison between the two melter platforms and provide an important link between radioactive testing and the larger-scale melter tests. This final step is not yet funded.

4.0 System Description

This section describes the equipment that will be built as part of the test platform. Included in this description is a summary of the primary design components and a piping and instrumentation diagram (P&ID) for each of the unit operations.

4.1 CUF System Description

The CUF system P&ID is provided in Figure 1. The system is similar to previous systems used to test tank waste (Shimskey et al. 2009; Geeting et al. 2003).

The system will meet the following criteria:

- Maximum process capacity of 4 L and minimum process capacity of 1 L.
- Ability to control axial velocity and transmembrane pressure independently up to 15 ft/sec (9 gpm) and 50 psig; 50 psig is the practical limit for this scale of equipment. Given the small scale of the equipment, higher operating pressures are not required to achieve nominal operating conditions. If higher pressure needs to be addressed in future testing, it should be addressed discretely.
- Accommodate 0.5-inch inside diameter × 2-foot-long crossflow filter.
- Ability to recycle or remove permeate during operation.
- Ability to maintain and control slurry temperature between 25 °C and 90 °C during operation.
- Slurry tank will be well-mixed over the range of possible volumes.
- The filter apparatus will be able to introduce cleaning solutions (see below), either through the filter feed tank or back through the shell side of the crossflow filter. When cleaning solutions are introduced through the shell side of the filter, the filter will be able to be drained to another tank or jar other than the filter feed tank, to match the LAWPS design.
- The filter apparatus will have chemical compatibility with 3M NaOH, 3M HNO_3, and 0.5M oxalic acid and an accumulated radiation dose of 10^6 rad.
- The filter apparatus will provide a backpulse system to conduct backpulses.
- The filter apparatus will be designed so that samples from both the filter feed tank and the filter permeate can be taken.
- The filter will be easily removable and replaceable so that a post-test analysis of the filter can be undertaken, if required by a given test objective.

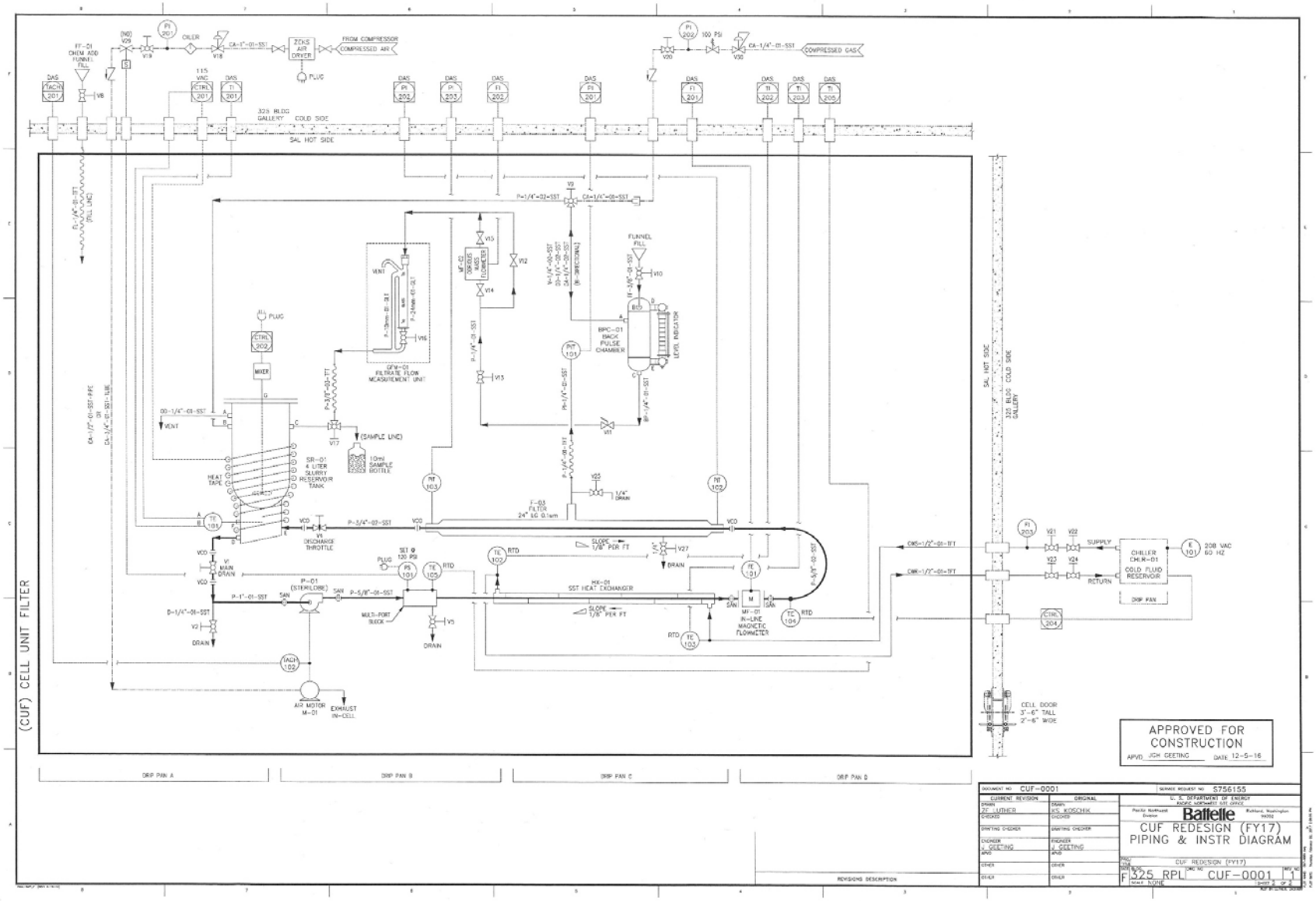

Figure 1. Cells Unit Filter P&ID

4.2 Cesium Ion Exchange System Description

The cesium ion exchange P&ID is provided in Figure 2 and Figure 3. This system design is nearly identical to a system design that was successfully used previously (Fiskum et al. 2006a, b). The system can be configured with either column in the lead position, and will meet the following criteria:

- Ability to process fluids through two ion exchange columns assembled in a lead/lag format.
- Ability to set the fluid-filled headspace above the resin bed.
- Ability to apply temperature control using jacketed columns.
- Ability to switch reagent flow direction through the columns (i.e., lead to lag and lag to lead).
- Ability to switch feeds, modify feed volumes, and modify feed flowrates from 0.2 to 10 mL per minute.
- Ability to collect samples directly from the lead column as well as from the lag column with concomitant ability to define the Cs load and elution curves.
- Ability to remove resin for analysis.

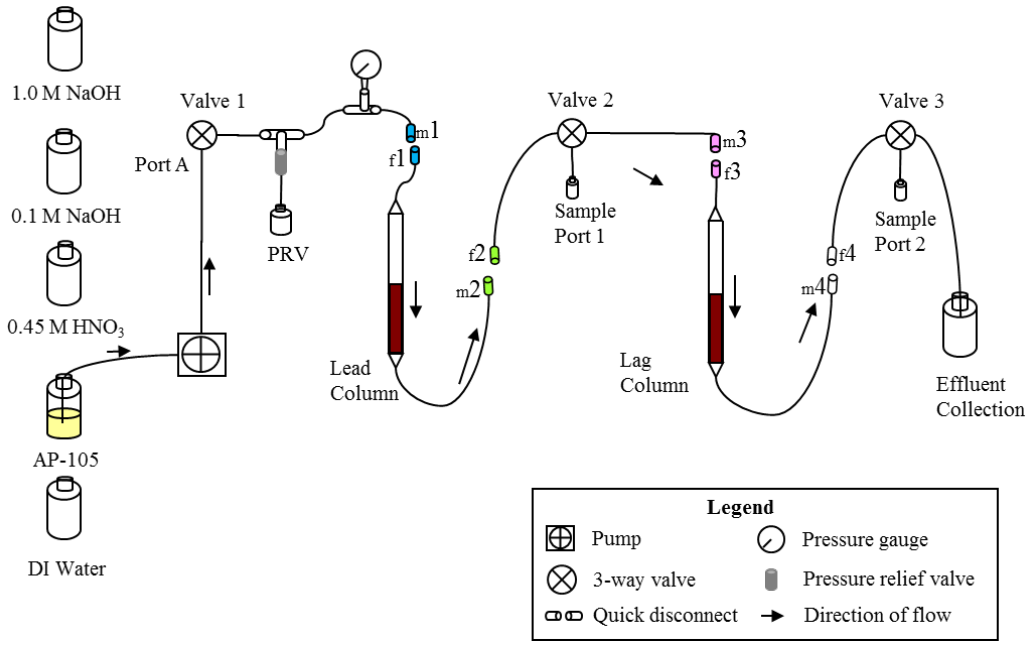

Figure 2. Cesium Ion Exchange P&ID for Downflow Lead-to-Lag Processing

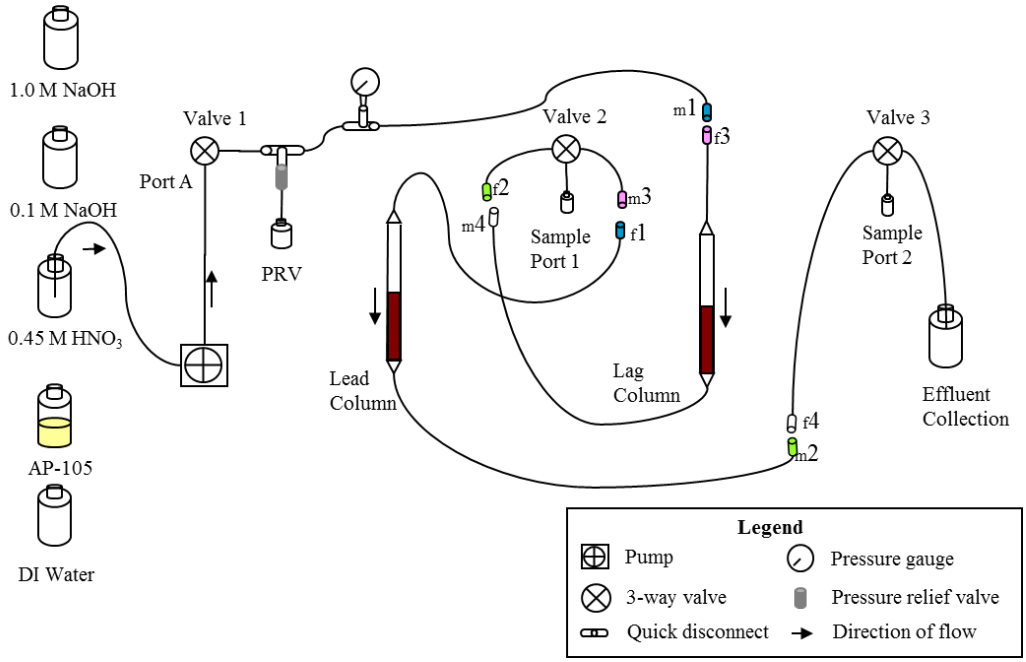

Figure 3. Cesium Ion Exchange P&ID for Downflow Lag-to-Lead Processing

4.3 Melter System Description

- Figure 4 provides the P&ID for the melter system.
- Melter feed will be kept under continuous agitation.
- Melter feed will be pumped to the melter continuously at a controlled rate through a water-cooled feed tube, with flushing capability.
- Melter vessel will have a 0.011-m^2 surface area (approximately 5-inch diameter).
- Melter will be heated by a furnace that surrounds the melter vessel up to the melt height.
- Melt pool will have bubbler agitation.
- Melt pool and plenum temperature will be monitored by thermocouples.
- Melt pool temperature will be maintained at approximately 1150 °C and plenum temperature targeted at 450 °C.
- Glass melt can be poured from the melter through an overflow discharge system.
- Poured glass can be weighed and samples taken for analysis.
- The offgas system and melter plenum will be kept at a negative pressure during processing.
- The offgas will be drawn through a scrubbing/quenching system and the offgas condensate collected.
- The full offgas stream will be sampled for a short duration during steady-state operation through heated HEPA filters and a simple scrubber to obtain chemical composition and quantities.

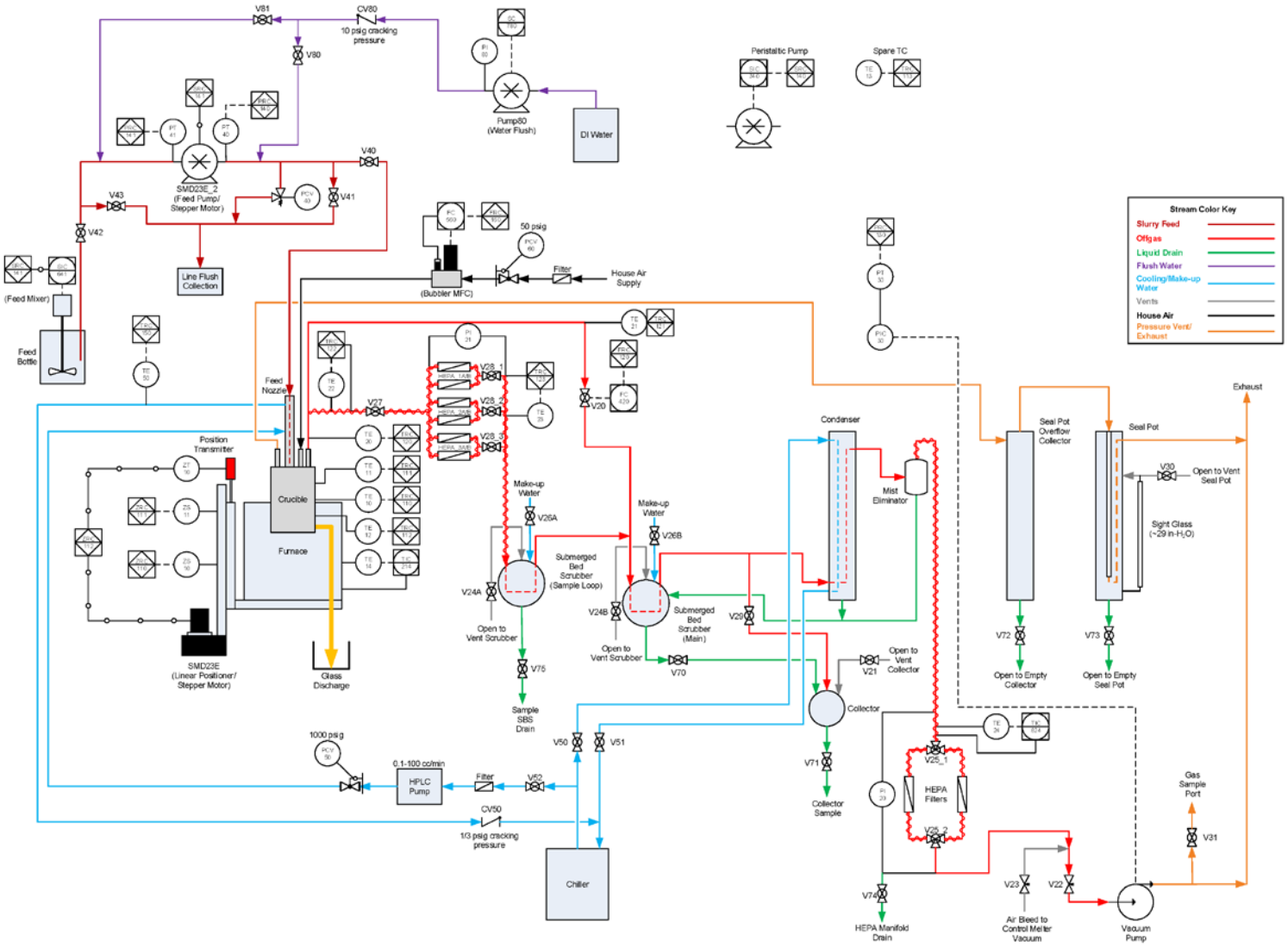

Figure 4. Continuous Laboratory-Scale Melter P&ID

5.0 Process Flow Diagram

The purpose of this document is to provide the initial basis for the Radioactive Waste Test Platform. Figure 5 summarizes the process flow for the test platform at PNNL for demonstration of DFLAW treatment with actual Hanford tank waste samples. Appendix A provides a more detailed diagram for the demonstration of these unit operations with waste that will be retrieved from tank AP-105 (estimated to be 8.5M Na) to produce 14 L of 5.6M Na feed. Note that this a general layout of how samples would be combined and would be adjusted as appropriate if the feed came from a different source with differing volumes and sodium molarities.

5.1 Charge the Vessel

The first step in the process will be to charge the CUF vessel. The total working volume of the CUF is 4 L; therefore, the quantity of tank waste will be added to produce 4 L at the target sodium molarity, and the balance will be made up with diluent (typically process water). This material will then be homogenized in the CUF vessel and samples will be collected as appropriate.

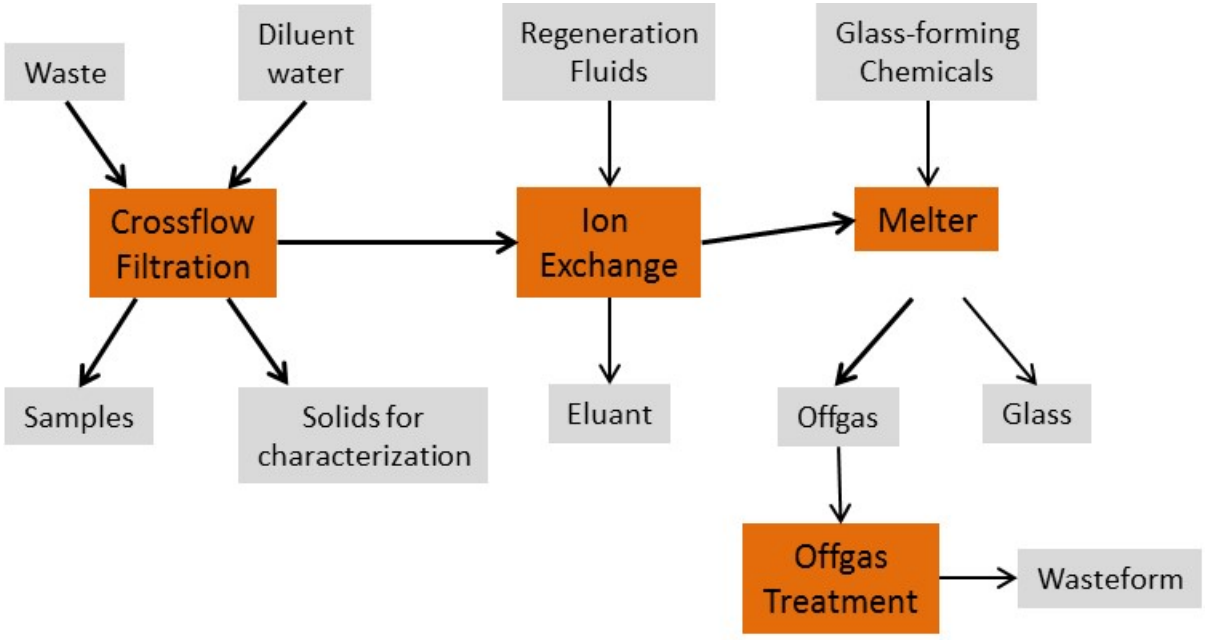

Figure 5. High-Level Process Flow Diagram for the Radioactive Waste Test Platform

5.2 Filtration

The next step in the process will be to filter the diluted feed material. The preliminary assumption is that this will be performed in a batch process. However, it could also be performed in a semi-batch process wherein 3000 mL aliquots of diluted feed will be added as each 3000 mL of clarified supernate is removed. Note that each added aliquot would need to be diluted to the target molarity.

5.3 Ion Exchange

The ion exchange column is sized to treat 3 to 5 L of clarified supernate, depending on the sodium, potassium, and cesium concentrations in the supernate. Each process run will be allowed to fully load the lead column and reach the target Cs breakthrough from the lag column. This will provide valuable information regarding the expected full column breakthrough for this feed material. Samples of the effluent will be collected from both columns. Effluent will be collected in stages such that the composite effluent Cs concentration will not exceed the vitrification processing criterion. The properly decontaminated lead and lag column samples will be combined with the composite effluent, as necessary. Cesium-contaminated samples from the initial processing runs may be combined with subsequent feed material and reprocessed to obtain the necessary decontamination factor.

5.4 Melter

The material from the ion exchange column runs will be loaded out of the hot cell to be used as LAW feed for the melter testing. Analysis of the ion exchange product will be used to determine the required glass-forming chemicals. Note that the quantity of glass-forming chemicals is not specified in the process flow diagram (Figure 5). It is planned to be determined from analytical data obtained from the ion exchange product. This material will be fed to the melter and converted to glass. Offgas condensate will be captured for further processing.

5.5 Evaporation and Disposition

The condensate from the melter offgas will be evaporated to an appropriate endpoint and then dispositioned into a final wasteform. The composition of this final wasteform is not yet determined and will depend on the composition of the condensate.

6.0 Future Capabilities

As described in Sections 3.0 and 4.0, the initial installation of the Radioactive Waste Test Platform will not have all the capabilities outlined in the requirements in Section 2.0. The majority of these additional capabilities were deferred during the initial construction of the platform to enable timely installation of the equipment. In addition, these capabilities were not required for the initial mission of the test platform, but would be deployed at a later time to meet specific process requirements. The following capabilities were deferred for future implementation.

- Constant Flowrate Filtration – The crossflow filtration system is currently designed to operate at fixed transmembrane pressure. Additional process control equipment would need to be installed to enable constant flux operations. At this time, the fluxes are expected to be relatively stable, and as such, the added complexity of constant flux operation is not warranted. However, future testing may explore the impact of constant flux operation.
- High-Level Waste Processing Including Leaching – The crossflow filtration system (including the feed vessel) is capable of handling the high solids loading associated with processing of high-level waste. However, the system does not have the ability to heat the feed vessel and hold at elevated temperature for an extended period of time. Heat tape could be applied to the feed vessel to enable temperature control for leaching capability. Note that the system does have temperature control for the filtration operation.
- Ion Exchange Temperature Control – Temperature control of the ion exchange columns would be deployed to address issues associated with the potential for precipitation of solids. It should be noted that the initial feeds are not expected to have a significant risk of precipitation, as there will be significant dilution of the sample prior to running, which should alleviate saturation of most (but not all) species of interest.
- Ion Exchange Media Sampling – At this time, it is not feasible to sub-sample the resin bed between column runs. For operations in the hot cell, this would require extensive effort. If future testing requires in-process sampling, the equipment configuration would need to be reevaluated.
- Melter Salt Gall Sampling – Sampling of a separated salt gall phase floating on the melt surface cannot be performed with the current configuration. In the future, a mechanism could be added to the lid of the melter for sampling such a phase. Alternately, as has been done with the fused silica LSM vessels in the past, a mechanism for removing the melter vessel from the furnace and quenching the melter to obtain a sample could be implemented.
- Melter Offgas Recycle – The current test plan for the vitrification system includes capturing the offgas condensate, but it will be evaporated and solidified, rather than being recycled back to the melter. A scheme for offgas recycle could be added to the system in the future.
- Flowsheet Changes – PNNL currently has an internal investment (Nuclear Process Science Initiative) that is aimed, in part, at identifying potential future flowsheet changes. These potential changes could be demonstrated in the test platform.
- Ion Exchange Backpressure – If testing is to evaluate ion exchange gas control, backpressure control on the ion exchange system would be added.

7.0 References

Daniel RC, JM Billing, JR Bontha, CF Brown, PW Eslinger, BD Hanson, JL Huckaby, NK Karri, ML Kimura, DE Kurath, and MJ Minette. 2010. *EFRT M-12 Issue Resolution: Comparison of Filter Performance at PEP and CUF Scale*. PNNL-18498, Rev. 1, Pacific Northwest National Laboratory, Richland, Washington.

Dixon DR. 2016. *Rhenium Mass Balance in a Laboratory-Scale Melter*. Presentation at the Glass and Optical Materials Division meeting. PNNL-SA- 118213, Pacific Northwest National Laboratory, Richland, Washington.

Edwards MK, JM Billing, DL Blanchard, EC Buck, AJ Casella, AM Casella, JV Crum, RC Daniel, KE Draper, SK Fiskum, LK Jagoda, ED Jenson, AE Kozelisky, PJ MacFarlan, RA Peterson, RW Shimskey, LA Snow, and RG Swoboda. 2009. *Characterization, Leaching, and Filtration Testing for Tributyl Phosphate (TBP, Group 7) Actual Waste Sample Composites*. PNNL-18119; WTP-RPT-169, Pacific Northwest National Laboratory, Richland, Washington.

Fiskum SK, EC Buck, RC Daniel, KE Draper, MK Edwards, TL Hubler, LK Jagoda, ED Jenson, AE Kozelisky, GJ Lumetta, PJ MacFarlan, K McNamara, RA Peterson, SI Sinkov, LA Snow, and RG Swoboda. 2008. *Characterization and Leach Testing for REDOX Sludge and S-Saltcake Actual Waste Sample Composites*. PNNL-17368; WTP-RPT-157, Pacific Northwest National Laboratory, Richland, Washington.

Fiskum SK, JM Billing, EC Buck, RC Daniel, KE Draper, MK Edwards, ED Jenson, AE Kozelisky, PJ MacFarlan, RA Peterson, RW Shimskey, and LA Snow. 2009. *Laboratory Demonstration of the Pretreatment Process with Caustic and Oxidative Leaching Using Actual Hanford Tank Waste*. PNNL-18007; WTP-RPT-171, Pacific Northwest National Laboratory, Richland, Washington.

Fiskum SK, ST Arm, MS Fountain, MJ Steele, and DL Blanchard, Jr. 2006a. *Spherical Resorcinol-Formaldehyde Resin Testing for ^{137}Cs Removal from Simulated and Actual Hanford Waste Tank 241-AP-101 Diluted Feed (Envelope A) Using Small Column Ion Exchange*. PNWD-3697; WTP-RPT-134, Rev. 0, Battelle—Pacific Northwest Division, Richland, Washington.

Fiskum SK, MJ Steele, and DL Blanchard, Jr. 2006b. S*mall Column Ion Exchange Testing of Spherical Resorcinol-Formaldehyde Resin for ^{137}Cs Removal from Pre-Treated Hanford Tank 241-AN-102 Waste (Envelope C)*. PNWD-3751; WTP-RPT-135, Rev. 1, Battelle—Pacific Northwest Division, Richland, Washington.

Geeting JGH, RT Hallen, LT Jagoda, AP Poloski, RD Scheele, and DR Weier 2003. *Filtration, Washing, and Caustic Leaching of Hanford Tank AZ-101 Sludge*. PNWD-3206, Rev. 1; WTP-RPT-043, Pacific Northwest National Laboratory, Richland, Washington.

Helfferich F. 1962. *Ion Exchange*. McGraw Hill Book Company, Inc., New York, New York.

Lumetta GJ, EC Buck, RC Daniel, K Draper, MK Edwards, SK Fiskum, RT Hallen, LK Jagoda, ED Jenson, AE Kozelisky, PJ MacFarlan, RA Peterson, RW Shimskey, SI Sinkov, and LA Snow. 2009. *Characterization, Leaching, and Filtration Testing for Bismuth Phosphate Sludge (Group 1) and Bismuth*

Phosphate Saltcake (Group 2) Actual Waste Sample Composites. PNNL-17992; WTP-RPT-166, Pacific Northwest National Laboratory, Richland, Washington.

Shimskey RW, JM Billing, EC Buck, AJ Casella, JV Crum, RD Daniel, K Draper, MK Edwards, RT Hallen, AE Kozelisky, PJ MacFarlan, RA Peterson, and RG Swoboda. 2009. *Filtration and Leach Testing for PUREX Cladding Sludge and REDOX Cladding Sludge Actual Waste Sample Composites*. PNNL-18048; WTP-RPT-181, Pacific Northwest National Laboratory, Richland, Washington.

Smith GL, DJ Bates, RW Goles, LR Greenwood, RC Lettau, GF Piepel, MJ Schweiger, HD Smith, MW Urie, and JJ Wagner. 2001. *Vitrification and Product Testing of C-104 and AZ-102 Pretreated Sludge Mixed with Flowsheet Quantities of Secondary Wastes*. PNNL-13452; WTP-RPT-006, Pacific Northwest National Laboratory, Richland, Washington.

Smith GL, LR Greenwood, GF Piepel, MJ Schweiger, HD Smith, MW Urie, and JJ Wagner. 2000. *Vitrification and Product Testing of AW-101 and AN-107 Pretreated Waste*. PNNL-13372; WTP-RPT-003, Pacific Northwest National Laboratory, Richland, Washington.

Whittington KF, DK Seiler, J Luey, JD Vienna, and WA Slinger. 1996. *Feed Process Studies – Research-Scale Melter*. PNNL-11333, Pacific Northwest National Laboratory, Richland, Washington.

Appendix A

Material Flow for LAW Actual Waste Test Platform

Attached is a material flow diagram that assumes approximately 9 L of waste from tank AP-105 is to be processed. Note that this material must be diluted and that it cannot all be accommodated in the cells unit filter feed vessel. Therefore, it would need to be staged in multiple filtration campaigns. Likewise, the filtrate produced would exceed the capacity of the ion exchange columns and would need to be processed in multiple column loadings. Also, samples would need to be diverted periodically to assess system performance. Finally, the decontaminated effluent would be composited for a single melter run.

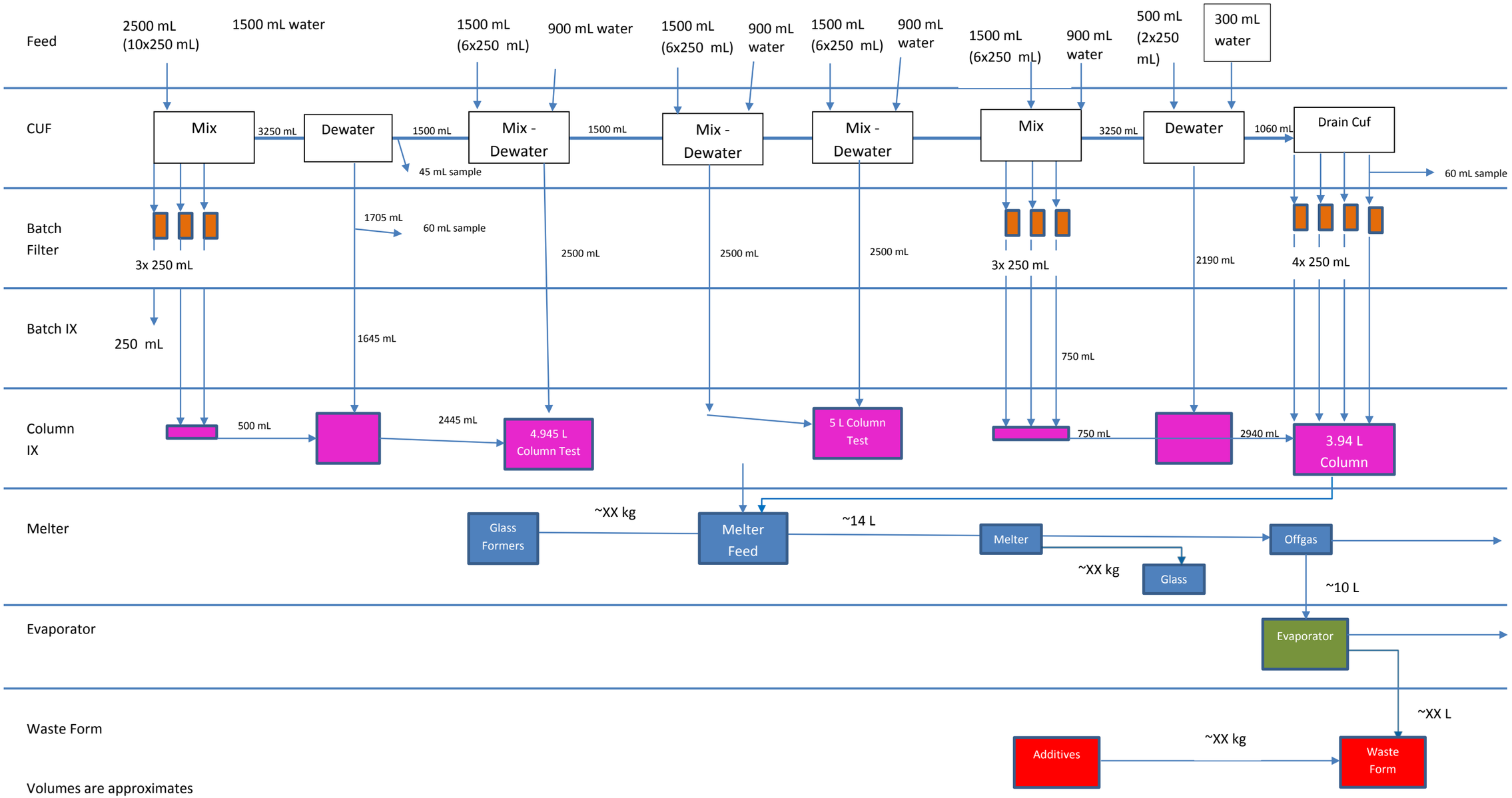

Figure A.1. Material Flow for LAW Actual Waste Test Platform

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