Direct-Feed Low-Activity Waste: First Feed Flowsheet – 17108

Laura H. Cree
Washington River Protection Solutions, LLC

ABSTRACT

In support of hot commissioning of the Low-Activity Waste Pretreatment System (LAWPS) and the Hanford Tank Waste Treatment and Immobilization Plant (WTP) LAW facility, the First Feed Flowsheet was initiated to project process stream characteristics and unit operation performance. The First Feed Flowsheet uses flowsheet models to simulate the preparation of the first feed campaign in Tank Farms and the subsequent feeding of the waste to the Low-Activity Waste Pretreatment System (LAWPS) and the WTP LAW facility. Three different models were used: a chemistry model of waste transfers developed in the Environmental Simulation Program[a] (ESP), Washington River Protection Solutions' (WRPS') dynamic gPROMS™[b] LAWPS flowsheet model, and WTP's Dynamic G2 flowsheet model. Information from all three models showed favorable comparison of the waste feeds and secondary liquid wastes against waste compatibility assessment and waste acceptance criteria at multiple stages in the flowsheet.

INTRODUCTION

As the Tank Farms and the WTP LAW facility transition to an operational phase, process flowsheets are being developed to define and support new processes. The establishment of process flowsheets is key to the direct-feed low-activity waste (DFLAW) program in providing assurance that facility waste acceptance criteria will be satisfied. Additionally, flowsheets provide the means by which processing opportunities can be evaluated, and potential process problems can be anticipated and resolved.

The First Feed Flowsheet represents the best available information regarding the chemical composition of the hot commissioning batch and first feed campaign that will be processed in the LAWPS and WTP LAW facilities. Flowsheet models are one of the best sources of information available for predicting feed chemistry and downstream impacts for facilities that are not yet operational. The use of the three models that were combined to create the First Feed Flowsheet (a chemistry model in ESP, WRPS' dynamic gPROMS LAWPS model, and WTP's Dynamic G2 flowsheet model) marks the first time that the three flowsheet models have been integrated to develop a complete picture of the downstream impacts of waste transfers and feed preparation, as well as the first use of the gPROMS LAWPS model.

---

[a] Environmental Simulation Program (ESP) is a product of OLI Systems, Inc., Cedar Knolls, New Jersey.

[b] gPROMS is a trademark of Process Systems Enterprise Limited, England, United Kingdom.

For modeling purposes, the scope of the First Feed Flowsheet was divided into three main processes: preparation of feed for delivery to LAWPS, processing in LAWPS, and immobilization in the WTP LAW facility.

PROCESS DESCRIPTIONS

Feed Preparation

Double-shell tank (DST) 241-AP-107[c] is the designated feed tank to LAWPS for all DFLAW feed campaigns. For most campaigns, the feed will be prepared and sampled in another DST and then moved to AP-107 for transfer to LAWPS. For the first feed campaign, however, feed preparation, mixing and sampling will all occur in AP-107 prior to hot commissioning. The waste that currently resides in AP-107 is considered undesirable as the first feed for DFLAW due to high concentrations of sulfur and halides, which negatively impact the amount of waste oxides that can be incorporated into LAW glass. Instead, the supernatant currently residing in AP-105 will be used to create the first feed.

Until the DFLAW program is started, and for some years afterward, space in Hanford's DST system will remain at a premium, providing several challenges in feed staging. It is desired to maintain the solids layer in AP-107 as low as possible in order to maximize feed campaign volume once LAWPS has begun. This prevents AP-107 from being used as an evaporator slurry receipt tank. In addition, to maximize usable DST space, dilution of the feed to a sodium molarity that can be accepted by LAWPS (5.6 moles per liter) will not occur until just prior to the startup of the facility, leaving enough time for sampling and waste feed qualification. For these reasons, the transfer of waste from AP-105 into AP-107 is desired as soon as possible, but enough supernatant will need to be transferred to fill the DST so that available space in AP-105 can be maximized. Some of that supernatant will then need to be removed from AP-107, prior to dilution of the feed, in order to prevent over-filling of the DST.

These restrictions require that (at least) four transfers are made to prepare the first feed for ultimate delivery to LAWPS. All four transfers, along with the final waste feed transfer, are shown in the simplified process flow diagram in Fig. 1. The first transfer will occur as soon as practicable and will involve moving as much of the supernatant out of AP-107 as possible without disturbing the solids layer. AP-107 will then be unavailable for use for a short time while infrastructure upgrades are installed. Once the new infrastructure is in place, the transfer of supernatant from AP-105 will occur, while again trying to prevent disturbing the saltcake layer in either DST. The waste in AP-107 will then sit undisturbed for several years until closer to the hot commissioning date of LAWPS. Leaving sufficient time for mixing, sampling, and waste feed qualification, approximately 40% of the supernatant will be transferred out of AP-107, followed closely by an inhibited water (0.01 M NaOH, 0.011 M $NaNO_3$) addition. The DST contents will then be recirculated to homogenize

---

[c] Hereinafter, the "241-" prefix in the official tank names for the DSTs will be omitted for readability purposes.

the supernatant prior to sampling. Following waste sampling, the diluted supernatant will remain undisturbed in AP-107 for approximately six months while the samples are processed and waste feed qualification is confirmed. The first feed batch will then be ready for delivery to LAWPS.

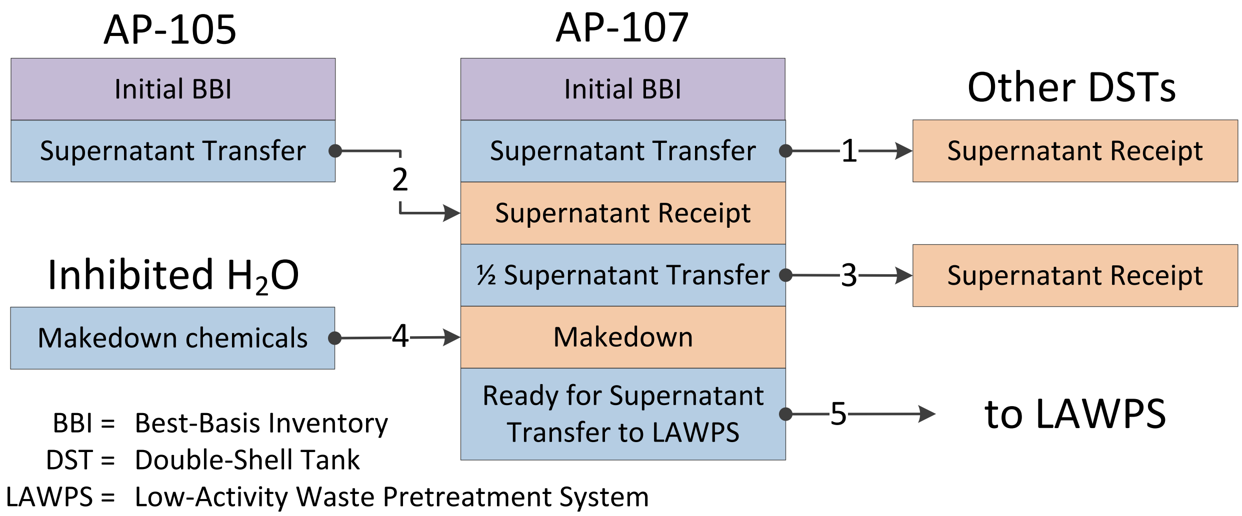

Fig. 1. Feed preparation process flow diagram

Processing in LAWPS

Qualified waste will be delivered to the filter feed tank in LAWPS using a continuous transfer loop. Supernatant feed will be pumped to the filter feed tank at a flow rate of 341 liters (90 gallons) per minute. The supernatant will then pass through a cross-flow filter where 23-38 liters (6-10 gallons) per minute of filtrate will continue through the LAWPS ion exchange system and the remaining 303-318 liters (80-84 gallons) per minute of supernatant and filtered solids will be returned to AP-107. This feed/return cycle will continue until a minimal amount of supernatant remains in AP-107, which will be determined by trying to minimize the disturbance and re-entrainment of solids in the DST.

The filtrate will be fed through two cesium ion exchange columns, which will operate in a “lead/lag” carousel fashion. The columns are expected to remove cesium at an overall removal efficiency of 99.99%. Treated supernate will then be collected in three lag storage tanks (LSTs). The LSTs will operate in parallel, with one vessel either empty or filling, one vessel sampled and awaiting analytical results, and one vessel approved and being transferred to the WTP LAW facility.

Immobilization in the WTP LAW facility

Treated supernate that is fed to the WTP LAW facility will enter one of two concentrate receipt vessels (CRVs). There it may be mixed with concentrate from the Effluent Management Facility (EMF) evaporator, sampled to determine glass formulation, and pumped to the melter feed preparation vessels. Glass-forming chemicals are loaded into glass former feed hoppers, mixed with demineralized water, and then discharged into the melter feed preparation vessels, where they are blended with the mixture from the CRVs. After adequate mixing occurs, the waste batch will be re-sampled and analyzed for waste compliance. Waste will then be transferred to the melter feed vessels and continuously fed to the LAW melters.

The LAW melters will operate by maintaining a large pool of molten glass at approximately 1,150 °C. This heat will decompose the waste into elemental oxides that dissolve into the glass. Bubblers will inject air that will agitate the glass pool, keeping a consistent temperature profile throughout. The molten glass will discharge into large cylindrical stainless steel containers, where it will cool and solidify.

Moist gases from the melters will be directed to the LAW primary offgas system where they will be cooled and treated in the submerged bed scrubbers and wet electrostatic precipitator. The treated gases are then sent to the secondary offgas treatment system and eventually released to the atmosphere. Condensate from the primary offgas system will be transferred to the EMF. The EMF evaporator will concentrate the secondary liquid effluent and recycle it back to the CRVs in the WTP LAW facility for mixing with the treated supernatant produced in LAWPS.

Secondary Waste Streams and Glass

Additionally, several secondary waste streams and the immobilized LAW glass will be generated during the DFLAW process:

- Solids slurry removed from the waste feed by the cross-flow filter will be returned to AP-107.
- Eluate from the LAWPS cesium ion exchange process will be temporarily stored in a holding tank in LAWPS, treated for corrosion control, and returned to DST AW-106 in the Tank Farms.
- Cesium ion exchange resin will be periodically replaced as degradation diminishes ion exchange capacity. The resin will be immobilized offsite and then returned to the Hanford Site for disposal at the Integrated Disposal Facility (IDF).
- Immobilized LAW glass containers will also be sent to IDF for disposal.
- Effluent from the caustic scrubber will be combined with offgas condensate from the EMF evaporator and sent to the Effluent Treatment Facility. The resulting powdered waste or brine will be transported to an offsite facility for stabilization and then returned to Hanford for disposal at IDF.
- During outages of the EMF, the effluent from the LAW offgas system will be treated for corrosion control and returned to DST AP-105 in the Tank Farms.

The processes described above are shown in a simplified process flow diagram in Fig. 2 and stream identification in Table I.

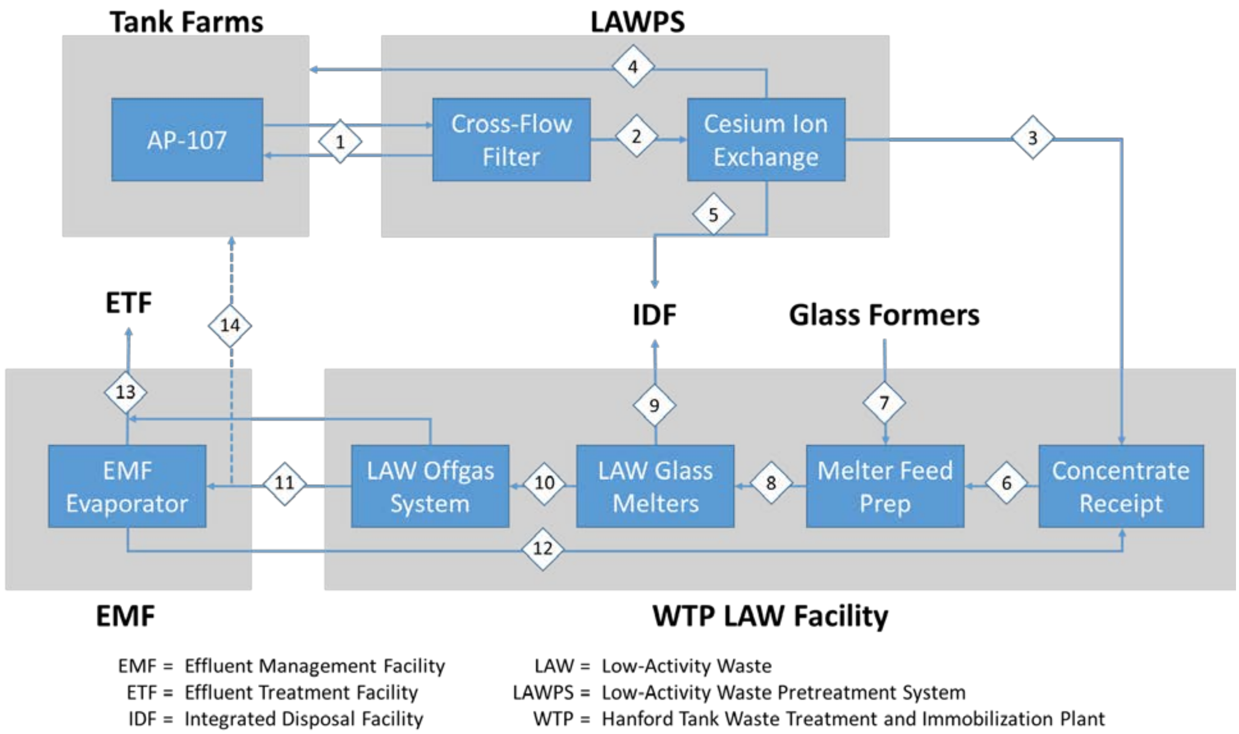

Fig. 2. Simplified process flowsheet

Table I. Stream identification for process flowsheet

Stream Number	Description	Stream Number	Description
1	Feed to LAWPS/Solids Return from Cross-Flow Filter	8	Prepared Melter Feed
2	LAW Filtrate	9	Immobilized LAW
3	Treated LAW	10	LAW Melter Offgas
4	Cesium Ion Exchange Eluate	11	LAW Offgas System Effluent
5	Cesium Ion Exchange Resin (Immobilized Off-site)	12	Concentrated EMF Evaporator Bottoms
6	Feed Ready for Glass Former Addition	13	EMF Offgas Condensate and Caustic Scrubber Effluent
7	Glass Former Addition	14	Off-normal Returns of Dilute EMF Feed to Tank Farms

MODELING

Each of the three models was operated as a stand-alone model, with the output of one becoming the input of another at a logical interface point. Fig. 3 shows a simplified modeling process breakdown with the arrows representing the interfaces between models.

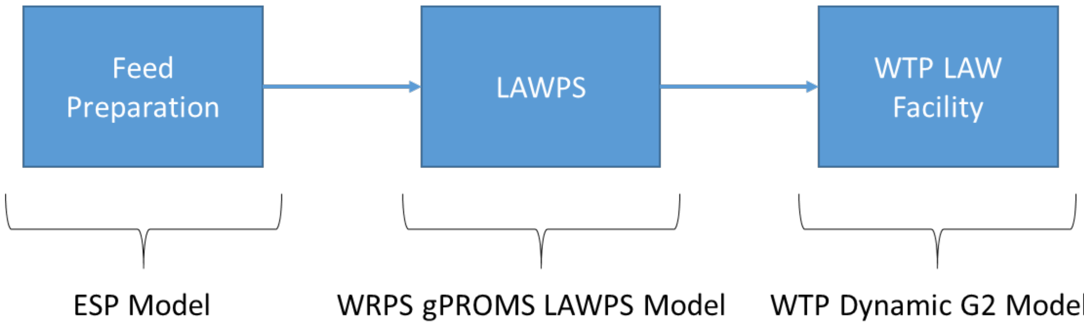

Fig. 3. First Feed Flowsheet process breakdown

For each model, the input (called a feed vector) was manipulated in order to be compatible with the model data feed configuration. Furthermore, specific assumptions were necessary for model functionality and to account for unknowns in the process.

ESP Model Input and Assumptions

Preparation of the feed vector for the ESP model [1] started by loading the BBI of the DSTs into OLI Studio Stream Analyzer[d]. In order to account for computational processing limitations, only components present in concentrations of greater than 100 parts per million were included in the model, with the exception of specifically requested radionuclides (Co-60, Tc-99, Cs-137, Eu-152, Eu-154, Eu-155, Pu-238, Pu-239, Pu 240, Pu-241, Pu-242, Am-241, Am-243, total strontium and total uranium).

Additionally, oxidation state and speciation assumptions were made in order to improve the charge balance: (1) Oxalate was subtracted from total organic carbon and the remaining carbon was attributed to formate. (2) Uranium was speciated as UO_2^{+2}. (3) Chromium was speciated as CrO_4^{-2}. (4) Aluminum was speciated as 100% $Al(OH)_4^-$ for tank AP-105, and 70% $Al(OH)_4^-$/30% Al^{+3} for AP-107. The Stream Analyzer calculated a charge-balanced equilibrium composition for each waste stream and these were used as inputs to ESP. Once in ESP, the Mixed Solvent Electrolyte chemistry allocated each component as necessary.

---

[d] Stream Analyzer is a product of OLI Systems, Inc., Cedar Knolls, New Jersey.

Further assumptions:

- It is assumed that chemical additions and waste transfers occur instantaneously and all supernatant is mixed homogenously using recirculation mixing.
- It is assumed that no appreciable evaporation occurs inside the DSTs, since the waste in question is not very thermally hot.
- No transfer line flushes are assumed necessary within the Tank Farms.
- It is assumed that during water/chemical additions and recirculation mixing no settled saltcake solids will dissolve.

WRPS gPROMS LAWPS Model Input and Assumptions

The final composition of AP-107 from the ESP model [1] was used as the input for the gPROMS LAWPS model. ESP tracks all OLI components and uses only mass values. gPROMS tracks a list of non-OLI as well as some OLI components, and tracks radioactive components according to their activity. Due to the different component lists for each model, the components were mapped in order to move from one platform to the other [2]. The isotopic abundance of the final waste stream in ESP was unknown, so it was assumed that the ratio of isotopes from the bulk of the waste in the AP-105 BBI remained the same.

Unlike the ESP model, which was designed specifically for the First Feed Flowsheet project, the gPROMS LAWPS model was created as a stand-alone model, which was modified to meet the requirements of this scenario. For this model run, the baseline model was updated to meet LAWPS facility requirements from the 30% design review [3]. It is assumed that at the time of the transfer of the first feed campaign, the facility and all interfaces will be complete and operational. Model-specific assumptions are documented in [4].

WTP Dynamic G2 Model Input and Assumptions

The gPROMS model recorded the final composition of each LST prior to the tank being emptied and refilled [1]. This file was used as the feed vector for the Dynamic G2 model. Due to the feed vector containing the entire tank volume for each LST, the heel volumes of each full LST 68,137 liters (18,000 gallons) were removed before the feed vector was transmitted to WTP for processing in the model. This prevented confusion with extra treated waste entering the WTP LAW facility in the model run. Additionally, similar to the ESP/gPROMS interface point, different component lists were used in the two models. The feed vector components were mapped to their appropriate counterparts in the Dynamic G2 model, which uses element-based compositions and tracks components in units of kilogram-moles.

The Dynamic G2 model is also a stand-alone model [5]. For the first feed scenario a previously run scenario was used as the starting point, and minor changes were made to accommodate the new feed vector [6]. Model-specific assumptions are documented in [7]. For this model run the 2009 LAW glass model (WTP baseline glass model) was used and 100% recycle of the EMF bottoms to the CRVs was assumed.

DISCUSSION

Modeling of the three processes separately went smoothly. Mapping of the feed vectors from one platform to another was expected and cannot be eliminated, as it is not possible to change the component lists for the various models.

One aspect that will be modified is the simulation timing of the gPROMS LAWPS model and the Dynamic G2 model. Both processes were run at 100% operating efficiency, however the gPROMS LAWPS model predicted that it would take 0.55 years to process the first feed campaign, and the Dynamic G2 model predicted it would take 0.37 years. Since the two processes were not directly coupled, this did not present an issue in evaluating model results and modeling was not changed. In future revisions, however, the gPROMS flow rate into the LSTs can be increased from the 15 liters (4 gallon) per minute set point used in these runs to an optimized flow rate that will feed the WTP LAW facility at the desired frequency and prevent downtime of the ion exchange process. This information will be useful for determining facility startup parameters in the future.

Technical bases for the assumptions used in the ESP model also need to be improved. Operating experience and process knowledge were relied on to develop the assumptions, however the experience came from single-shell tank retrievals and may differ from new processes. Computational fluid dynamics modeling is being used to investigate the assumption that recirculation mixing can homogenously combine two layers of significantly different densities. Laboratory analysis will be used to examine the assumption that no saltcake will dissolve while the diluted supernatant is mixed or fed to LAWPS.

Results

Overall the LAWPS model processed 4.13E+06 liters (1.09E+06 gallons) of waste and 4.9E+05 kilograms of sodium. The cesium ion exchange resin was eluted seven times, and 5.75E+05 liters (1.52E+05 gallons) of corrosion control treated cesium eluate was returned to the Tank Farms. A total of 4.8E+05 kilograms of waste sodium was turned into immobilized LAW glass, creating 639 glass packages. Glass production began at the maximum sodium loading allowed for the glass model of 21 wt%, however this was quickly reduced in order to lower the chlorine content to acceptable levels. The lowest sodium loading level was just over 15 wt%, and the average was about 18 wt%.

Outputs from all three models were used in calculations to compare the various waste streams against available transfer and waste acceptance requirements. The primary driver for creating the First Feed Flowsheet was to confirm that the chemistry of the first feed campaign will be able to meet waste acceptance criteria for both LAWPS and the WTP LAW facility.

Waste acceptance criteria and target concentrations for LAWPS [8] and the calculated values for the first feed campaign are outlined in Table II. Some criteria were not able to be evaluated from model results and are not presented here. None of the calculated values were outside the limits for LAWPS.

Table II. LAWPS Waste Acceptance Criteria Analysis

	Units	Limit	LAWPS Feed
Waste Acceptance Criteria			
Density	g/mL	< 1.35	1.25
Cs-137 concentration	Bq/L (Ci/L)	< 1.9E+10 (< 5.0E-01)	6.3E+09 (1.7E-01)
Cs-137:Total Cs ratio	unitless	< 0.24	0.18
Radiological source term – onsite liquids Unit Liter Dose	Sv/L	< 1.50E+02	8.55E+00
Radiological source term – onsite solids Unit Liter Dose	Sv/L	< 5.70E+04	3.27E-01
Radiological source term – offsite liquids Unit Liter Dose	Sv/L	< 1.09E+02	6.42E+00
Radiological source term – offsite solids Unit Liter Dose	Sv/L	< 8.50E+04	5.60E-01
Target Concentrations			
Sodium molarity	mol/L	5 < $[Na^+]$ < 6	5.6
Potassium concentration	mol/L	< 0.35	0.08
Slurry viscosity	Pa·s (cP)	0.015 (< 15)	0.0047 (4.7)

Treated waste fed to the WTP LAW facility is also required to meet acceptance criteria [9]. Since waste fed to LAWPS is treated only to remove solids and cesium, all other acceptance criteria are required to be met by the LAWPS feed as well. Criteria that were able to be calculated from model results are presented in Table III. None of the calculated values were outside of the limits for the WTP LAW facility.

Waste acceptance criteria and transfer requirements for secondary waste streams were also examined and found to be within known limits. Cesium eluate and secondary liquid effluent from the LAW offgas system that is returned to the Tank Farms will need to be treated for corrosion control prior to acceptance by the Tank Farms.

Table III. WTP LAW Facility Acceptance Criteria Analysis

	Units	Limit	LAWPS Feed	WTP LAW Feed
Feed pH	unitless	≥ 12	14.1	14.2
Feed viscosity	Pa·s (cP)	0.015 (≤ 15)	0.0047 (4.7)	0.0043 (4.3)
Sodium concentration	mol/L	$5 \leq [Na^+] \leq 8$	5.6	5.5
Chloride ratio	mol/mol sodium	< 3.7E-02	2.10E-02	2.10E-02
Fluoride ratio	mol/mol sodium	< 9.1 E-02	1.69E-04	1.73E-04
Sulfate ratio	mol/mol sodium	< 7.0E-02	6.47E-03	6.47E-03
Total organic carbon concentration	wt%	< 10	0.24	0.24
Cs-137 ratio	Bq/mol sodium (Ci/mol sodium)	1.18E+06 (< 3.18E-05)	[a]	5.25E+02 (1.42E-08)
Eu-154 concentration	Bq/L (Ci/L)	6.7E+05 (< 1.8E-05)	3.5E+05 (9.5E-06)	3.5E+05 (9.5E-06)
Sr-90 concentration	Bq/mol sodium (Ci/mol sodium)	4.40E+07 (< 1.19E-03)	3.12E+06 (8.44E-05)	1.67E+06 (4.50E-05)
Tc-99 concentration	Bq/L (Ci/L)	1.8E+07 (< 4.8E-04)	3.7E+06 (1.0E-04)	1.6E+06 (4.2E-05)
Pu-239 concentration	Bq/L (Ci/L)	1.1E+06 (< 3.0E-05)	1.1E+04 (3.0E-07)	1.1E+04 (3.1E-07)
U-233 concentration	Bq/L (Ci/L)	5.9E+03 (< 1.6E-07)	1.4E+02 (3.9E-09)	1.3E+02 (3.4E-09)
U-235 concentration	Bq/L (Ci/L)	6.3E+01 (< 1.7E-09)	2.1E+00 (5.7E-11)	2.1E+00 (5.6E-11)
TRU[b] ratio	Bq/mol sodium (Ci/mol sodium)	4.81E+05 (< 1.30E-05)	2.86E+03 (7.72E-08)	2.99E+03 (8.09E-08)
U fissile to U total	wt%	< 0.96	0.74	0.73

[a] This parameter is not required to be met for LAWPS feed as Cs-137 is removed in the LAWPS facility.
[b] For the purposes of this flowsheet, TRU is defined as alpha-emitting isotopes with an atomic number greater than 92 and half-lives greater than five years [10].

CONCLUSIONS

Modeling of the first feed campaign using three separate flowsheet modeling tools was successful. Results showed that the first feed campaign can be expected to meet waste acceptance criteria requirements for the LAWPS and WTP LAW facilities, as well as for secondary wastes and product to meet those of downstream facilities. Secondary liquid effluent and cesium eluate will need to be treated for corrosion control prior to being returned to the Tank Farms. No unexpected results were observed. The modeling process went well and can be repeated for future revisions to the flowsheet and future feed campaigns. Mapping of the feed vectors to each new modeling platform is a necessary process that can be streamlined and adjusting the model flow rates so that the gPROMS LAWPS model and Dynamic G2 model show the facilities working in concert is a planned activity. Technical bases are being improved for assumptions used in the modeling process.

First feed flowsheet modeling efforts will continue as the startup of the LAWPS and WTP LAW facilities draws near. Flowsheet enhancements are expected to include updates to the BBI, information about planned DST upgrades, improved technical bases, and improved knowledge of transfer dates and receipt tanks in the near term.

REFERENCES

1. L. H. Cree, *Direct-Feed Low-Activity Waste: First Feed Flowsheet*, Rev. 0, Washington River Protection Solutions, LLC, Richland, Washington, 2016, RPP-RPT-58985.

2. R. Carter and T. J. Pace, *Low-Activity Waste Pretreatment System gPROMS Model (LAWPS-SR-01) First Feed Vector Scenario Acceptance Test Report*, Rev. 0, Washington River Protection Solutions, LLC, Richland, Washington, 2016, RPP-RPT-59295.

3. T. J. Pace, *LAWPS gPROMS Model First Feed Vector Scenario*, Rev. 0, Washington River Protection Solutions, LLC, Richland, Washington, 2016, WRPS-1600366-OS.

4. R. Carter, *One System gPROMS Low-Activity Waste Pretreatment System Model (LAWPS-NM-01) Design Document*, Rev. 0, Washington River Protection Solutions, LLC, Richland, Washington, 2015, RPP-RPT-58360.

5. Y. Deng, *Dynamic (G2) Model Design Document*, Rev. 13, Bechtel National, Inc., Richland, Washington, 2014, 24590-WTP-MDD-PR-01-02.

6. Y. Deng, *DFLAW First Feed Campaign*, Rev. 0, Bechtel National, Inc., Richland, Washington, 2016, 24590-WTP-MRR-PENG-16-005.

7. Y. Deng, et al., *Flowsheet Bases, Assumptions, and Requirements*, Rev. 8, Bechtel National, Inc., Richland, Washington, 2016, 24590-WTP-RPT-PT-02-005.

8. J. G. Reynolds, *Waste Acceptance Criteria for the Low activity Waste Pretreatment System*, Rev. 1, Washington River Protection Solutions, LLC, Richland, Washington, 2016, RPP-RPT-58649.

9. 24590-WTP-ICD-MG-01-030, 2015, *ICD 30 – Interface Control Document for Direct LAW Feed*, Rev. 0, Bechtel National, Inc., Richland, Washington.

10. 10 CFR 61.55, “Waste Classification,” *Code of Federal Regulations*, as amended.