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Revision 0

DWPF RECYCLE EVAPORATOR FLOWSHEET EVALUATION (U)

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April 2005

Immobilization Technology Section
Savannah River National Laboratory
Aiken, SC 29808

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EXECUTIVE SUMMARY

Evaluations were performed based on simulant testing and computer modeling of the proposed Recycle Evaporator for the Defense Waste Processing Facility (DWPF) to determine flowsheet requirements and identify potential impacts of the evaporator on downstream processes. Sludge Batch 4 (SB4) was utilized as the basis for the downstream process assessment. The evaporator will reduce the 1.4 million gallons of recycle currently produced to approximately 45,000 gallons. The overheads from the evaporation process will be sent to the Effluent Treatment Plant (ETP) while the concentrate will be recycled to the Sludge Receipt Adjustment Tank (SRAT).

The following conclusions were drawn from the evaluation:

- Approximately 1000 kg/hr of steam and 30,000 kg/hr of cooling water were required for the recycle evaporator as determined by an OLI simulation on the "typical" recycle stream. These requirements will be doubled once salt waste processing is implemented at DWPF.
- Recycling the Off Gas Condensate Tank (OGCT) and Slurry Mix Evaporator Condensate Tank (SMECT) condensate does not significantly impact the glass properties or operating windows for the SB4 system.
- Recycling the coil decontamination solution can impact the glass properties and for Frit SB4/Frit 320 system will reduce the operating window significantly while the operating window is increased for the SB4/Frit 418 system.
- Recycling the High Efficiency Mist Eliminator (HEME) dissolution solution can eliminate the operating window for the SB4/Frit 320 and SB4/Frit 418 systems due to the high sodium content. Alternative frits may be identified that could restore the operating windows.
- The recycle purge requirements will vary from sludge batch to sludge batch and are dependent upon the ratio of the feed concentration to the process limits for DWPF and the percentage of the contaminant removed in the glass stream, offgas, and/or recycle condensate.
- The recycle stream could have significant impacts on the SRAT cycle in a variety of ways. Impacts include: changes to the acid addition strategy, increased erosion of process equipment due to frit introduction, potential change in rheology properties from insoluble solids added from the recycle, extended processing time due to higher boil-off volumes, and the foaminess of the SRAT slurry due to antifoam degradation products.
- The condensate from the recycle evaporator could exceed the ETP Waste Acceptance Criteria (WAC) for mercury, silicon, and Total Organic Carbon (TOC) based on simulant tests. Treatment for mercury removal could include pH adjustment and/or ion exchange processes.

The following recommendations were made from the conclusions of the evaluation:

- Further evaluation of the amount of HEME dissolution material that can be processed in one SRAT cycle without significantly impacting the glass composition operating window is required or alternative disposal routes for this material needs to be identified.
- A calandria evaporator is recommended based on the proven remotability of this system and the ability to utilize an agitator to mitigate solids settling. It is assumed that a suitable antifoam agent can be identified to mitigate any foaming concerns.
- The impact of the recycle stream on the DWPF SRAT cycle should be evaluated. This evaluation should be completed after the selection of the antifoam agent to be utilized for the recycle evaporator.
- The treatment of condensate for mercury removal should be evaluated to determine if pH adjustment will reduce mercury concentrations to required levels.
- Evaluation of condensate treatment options for siloxane and TOC species should be performed. This evaluation will require characterization of the species present in the condensate.

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ADS	Analytical Development Section
CPC	Chemical Process Cell
DF	Decontamination Factor
DWPF	Defense Waste Processing Facility
DWPF-E	Defense Waste Processing Facility: Engineering
DWTT	Decontamination Waste Treatment Tank
ETP	Effluent Treatment Plant
HEME	High Efficiency Mist Eliminator
ML	Mobile Lab
NAS	Sodium Alumino-silicate
OGCT	Off Gas Condensate Tank
PCCS	Process Composition Control System
RCT	Recycle Collection Tank
SB4	Sludge Batch 4
SMECT	Slurry Mix Evaporator Condensate Tank
SRAT	Sludge Receipt and Adjustment Tank
SRNL	Savannah River National Laboratory
TOC	Total Organic Carbon
WAC	Waste Acceptance Criteria
WL	Waste Loading

1.0 INTRODUCTION

The Defense Waste Processing Facility (DWPF) converts the high level waste slurries stored at the Savannah River Site into borosilicate glass for long-term storage. The vitrification process results in the generation of approximately five gallons of dilute recycle streams for each gallon of waste slurry vitrified. This dilute recycle stream is currently transferred to the H-area Tank Farm and amounts to approximately 1,400,000 gallons of effluent per year. Process changes to incorporate salt waste could increase the amount of effluent to approximately 2,900,000 gallons per year[1].

The recycle consists of two major streams and four smaller streams. The first major recycle stream is condensate from the Chemical Process Cell (CPC), and is collected in the Slurry Mix Evaporator Condensate Tank (SMECT). The second major recycle stream is the melter offgas which is collected in the Off Gas Condensate Tank (OGCT). The four smaller streams are the sample flushes, sump flushes, decon solution, and High Efficiency Mist Eliminator (HEME) dissolution solution. These streams are collected in the Decontamination Waste Treatment Tank (DWTT) or the Recycle Collection Tank (RCT). All recycle streams are currently combined in the RCT and treated with sodium nitrite and sodium hydroxide prior to transfer to the tank farm.

Tank Farm space limitations and previous outages in the 2H Evaporator system due to deposition of sodium alumino-silicates[2] have led to evaluation of alternative methods of dealing with the DWPF recycle. One option identified for processing the recycle was a dedicated evaporator to concentrate the recycle stream to allow the solids to be recycled to the DWPF Sludge Receipt and Adjustment Tank (SRAT) and the condensate from this evaporation process to be sent and treated in the Effluent Treatment Plant (ETP).

In order to meet process objectives, the recycle stream must be concentrated to 1/30th of the feed volume during the evaporation process. The concentrated stream must be pumpable to the DWPF SRAT vessel and should not precipitate solids to avoid fouling the evaporator vessel and heat transfer coils. The evaporation process must not generate excessive foam and must have a high Decontamination Factor (DF) for many species in the evaporator feed to allow the condensate to be transferred to the ETP. An initial scoping study was completed in 2001 to evaluate the feasibility of the evaporator[1] which concluded that the concentration objectives could be met. This initial study was based on initial estimates of recycle concentration and was based solely on OLI modeling of the evaporation process.

The Savannah River National Laboratory (SRNL) has completed additional studies using simulated recycle streams[3] and OLI® simulations[4]. Based on this work, the proposed flowsheet for the recycle evaporator was evaluated for feasibility, evaporator design considerations, and impact on the DWPF process. This work was in accordance with guidance from DWPF-E[5] and was performed in accordance with the Technical Task and Quality Assurance Plan[3].

2.0 DISCUSSION

2.1 Material and Energy Balance

An OLI simulation was performed to provide an overall material and energy balance for typical operation of the recycle evaporator. This simulation was performed without additions from the HEME dissolution and equipment decontamination processes, as shown in Figure 1. The additions of these two streams impacted the evaporation process and the amounts of each that can be allowed during recycle evaporation is currently being evaluated.

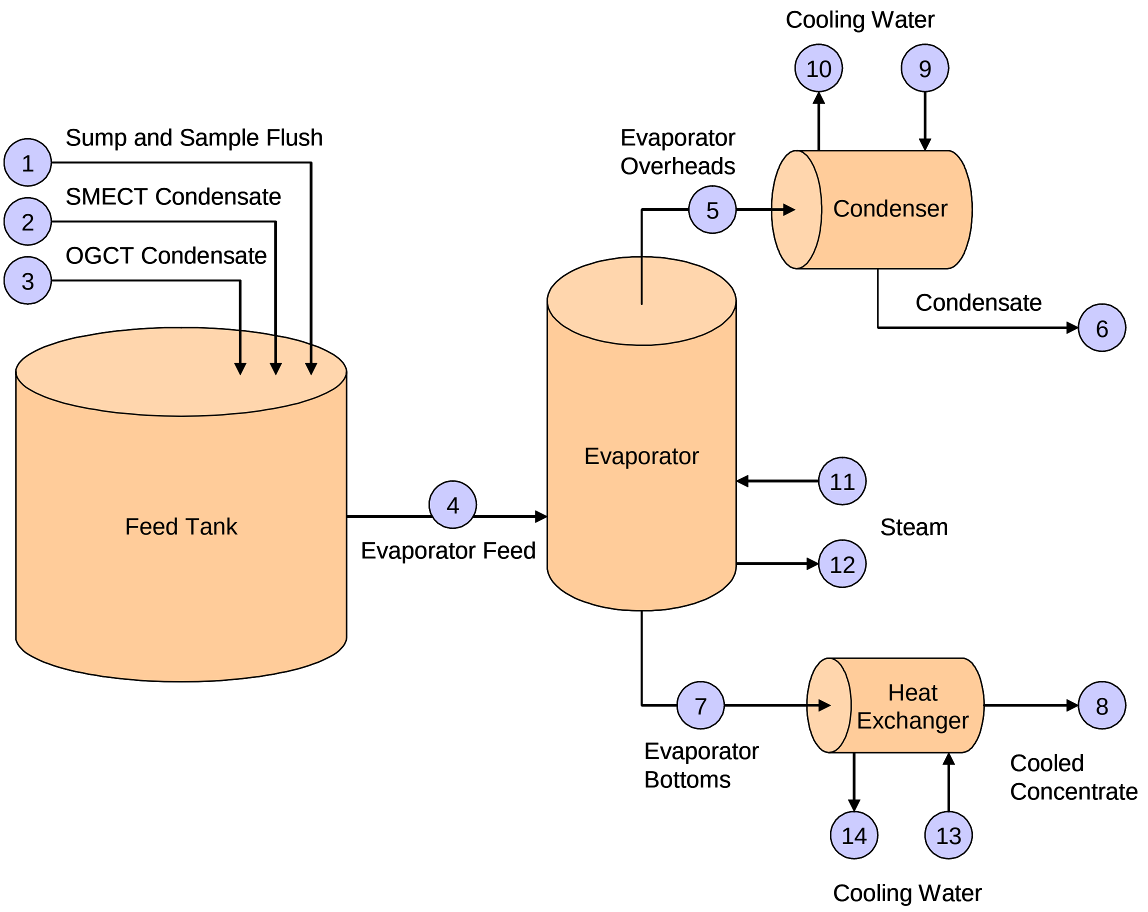

Figure 1. Flowsheet of OLI Model for Material and Energy Balance

The following assumptions were made to perform the mass balance:

1. 1,750,000 gallons of recycle evaporated per year
2. Volume ratios equal to Scenario #1 of the Task Plan[3]
3. 0.1% entrainment of evaporator feed into overheads
4. 30X volume reduction during evaporation
5. Non-condensable gases (such as air purge) not included in the balance

The 1.75 million gallons per year capacity was determined by assuming 80% attainment of evaporator operation and 1.4 million gallons of recycle processed per year. It should be noted that the values shown are nominal values and design factors have not been incorporated. These assumptions did not include the additional water that will be put into the system once salt operations are started. Salt operations are expected to approximately double the volume throughputs as shown in this flowsheet, but solids content is expected to be roughly unchanged. During salt operation, the evaporator will be required to reduce the volume by a factor of 60X to maintain the desired 1000 gallons of recycle per SRAT batch.

Based on the assumptions, the input streams were determined by OLI to have the composition and properties as shown in Table 1. Note that the sump and sample flushes were assumed to be water. The input streams were combined with the resulting composition shown for Stream 4. The albitelow shown for the OGCT condensate was a sodium aluminosilicate from the feldspar family. The resulting compositions for the concentrate and condensate as determined by the OLI simulation are shown in Table 2. After evaporation, the concentrate was approximately 6 wt% total solids and 8 wt% nitric acid. A “-“ in Table 1 and Table 2 indicate that the species was not present in that stream.

Table 1. Input streams to the Material and Energy Balance

Stream	OGCT Condensate	SMECT Condensate	Sample/Sump Flush	Evaporator Feed
Stream Designation	1	2	3	4
Temperature, C	25	25	25	25
Pressure, atm	1.000E+00	1.000E+00	1.000E+00	9.750E-01
pH	3.472E+00	1.153E+00	6.997E+00	1.564E+00
Total mol/hr	1.799E+04	2.106E+04	2.760E+03	4.181E+04
Species units	Wt fraction	Wt fraction	Wt fraction	Wt fraction
Al_2O_3	1.54E-04	-	-	1.72E-04
Albitelow	2.56E-04	-	-	-
$Al(OH)_3$	3.01E-04	-	-	-
$Am(OH)_3$	5.31E-08	5.31E-08	-	4.96E-08
CaO	3.22E-05	-	-	1.39E-05
$CoSiO_4$	2.52E-11	-	-	2.36E-11
$Co(OH)_2$	-	2.23E-11	-	-
CsOH	5.22E-08	5.22E-08	-	4.88E-08
EuO	2.91E-10	2.91E-10	-	2.72E-10
Fe_2O_3	2.90E-03	-	-	1.25E-03
Formic Acid	-	2.91E-04	-	1.47E-04
HF	2.00E-05	6.93E-11	-	8.61E-06
HgO	2.15E-04	2.16E-04	-	2.02E-04
Nitric Acid	1.62E-03	5.18E-03	-	3.31E-03
La_2O_3	1.15E-06	1.15E-06	-	1.08E-06
MgO	9.45E-05	-	-	4.07E-05
MnO	1.56E-04	-	-	6.71E-05
Na_2O	1.94E-04	-	-	9.66E-05
NiO	6.74E-05	-	-	2.90E-05
$Pu(OH)_4$	1.10E-06	1.10E-06	-	1.03E-06
SiO_2	4.24E-07	-	-	7.61E-05
$Ru(OH)_3$	3.06E-11	3.06E-11	-	2.86E-11
Sb_2O_3	8.66E-11	8.66E-11	-	8.09E-11
SO_3	5.09E-05	-	-	2.19E-05
TcO_2	7.98E-10	3.28E-08	-	7.63E-09
$TcO_2 \cdot 2H_2O$	2.05E-07	1.64E-07	-	1.83E-07
UO_2	2.70E-04	2.70E-04	-	2.53E-04
Water	9.94E-01	9.94E-01	1.00E+00	9.94E-01
Total kg/hr	3.26E+02	3.81E+02	4.97E+01	7.57E+02
Volume, L/hr	3.25E+02	3.81E+02	4.99E+01	7.56E+02
Enthalpy, J/hr	-5.15E+09	-6.02E+09	-7.89E+08	-1.20E+10
Solid fraction	4.47E-04	1.81E-05	-	1.76E-04
Abs Visc, cP	8.96E-01	8.93E-01	8.91E-01	8.95E-01
Relative Viscosity	1.01E+00	1.00E+00	1.00E+00	1.01E+00

Table 2. Evaporator Condensate and Concentrate Composition and Properties

Stream	Evaporator Overhead	Condensate	Evaporator Bottoms	Cooled Concentrate
Stream Designation	5	6	7	8
Temperature, C	1.002E+02	2.500E+01	1.002E+02	2.500E+01
Pressure, atm	9.750E-01	1.000E+00	9.750E-01	1.000E+00
pH	4.427E-01	2.113E+00	4.427E-01	9.225E-01
Total mol/hr	4.044E+04	4.044E+04	1.166E+03	1.376E+03
Species Units	Wt Fraction	Wt Fraction	Wt Fraction	Wt Fraction
Al_2O_3	5.534E-06	5.534E-06	5.284E-03	4.556E-03
$Am(OH)_3$	1.595E-09	1.595E-09	1.523E-06	1.313E-06
CaO	4.454E-07	4.454E-07	4.252E-04	3.667E-04
$CoSiO_4$	7.575E-13	7.575E-13	7.232E-10	6.236E-10
CsOH	1.567E-09	1.567E-09	1.496E-06	1.290E-06
EuO	8.727E-12	8.727E-12	8.332E-09	7.185E-09
Fe_2O_3	1.281E-06	1.281E-06	3.950E-02	3.406E-02
Formic Acid (dimer)	6.574E-12	-	-	-
H_2F_2	8.242E-15	-	-	-
H_2SO_4	3.706E-15	-	-	-
Formic Acid	1.435E-04	1.435E-04	2.700E-04	2.328E-04
HF	5.598E-07	5.598E-07	2.555E-04	2.203E-04
HgO	6.474E-06	6.474E-06	6.181E-03	5.330E-03
Nitric Acid	5.660E-04	5.660E-04	8.742E-02	7.538E-02
La_2O_3	3.455E-08	3.455E-08	3.298E-05	2.844E-05
MgO	1.307E-06	1.307E-06	1.248E-03	1.076E-03
MnO	2.156E-06	2.156E-06	2.059E-03	1.775E-03
Na_2O	3.103E-06	3.103E-06	2.963E-03	2.555E-03
NiO	9.323E-07	9.323E-07	8.901E-04	7.675E-04
$Pu(OH)_4$	3.308E-08	3.308E-08	3.159E-05	2.724E-05
SiO_2	4.374E-08	4.374E-08	4.683E-05	2.075E-03
$Ru(OH)_3$	9.193E-13	9.193E-13	8.777E-10	7.568E-10
Sb_2O_3	2.600E-12	2.600E-12	2.482E-09	2.141E-09
SiF_4	1.040E-17	-	-	-
SO_3	7.035E-07	7.035E-07	6.716E-04	5.791E-04
TcO_2	4.843E-09	1.856E-09	4.624E-06	3.844E-07
$TcO_2 \cdot 2H_2O$	-	3.809E-09	-	4.594E-06
UO_2	5.853E-08	5.853E-08	7.998E-03	6.896E-03
Water	9.993E-01	9.993E-01	8.447E-01	8.641E-01
Total kg/hr	7.29E+02	7.29E+02	2.39E+01	2.77E+01
Volume, L/hr	1.26E+06	7.31E+02	2.25E+01	2.52E+01
Enthalpy, J/hr	-9.68E+09	-1.16E+10	-3.31E+08	-3.98E+08
Vapor fraction	9.99E-01	-	-	-
Solid fraction	-	1.073E-07	5.52E-03	3.69E-03
Osmotic Pressure, atm	4.71E+01	4.82E-01	4.71E+01	4.15E+01
Abs Viscosity, cP	-	8.91E-01	-	9.94E-01
Relative Viscosity	-	1.00E+00	-	1.12E+00

2.1.1 Steam and Cooling Water Requirements

Calculations were performed using Excel® spreadsheets to determine the amount of steam and cooling water required to meet the heating and cooling requirements determined by the OLI simulation. The following assumptions were made:

1. Saturated steam is available at 85 psig.
2. Cooling water is available at 35° C
3. Allowable increase in cooling water temperature was 15° C.
4. Steam amount increased 10% to account for heat loss from evaporator.

The change in enthalpy from the OLI calculations was used to determine the heating or cooling required. The OLI run was conducted assuming a temperature of 25° C for the cooled condensate and concentrate. Cooling water service is available at 35° C, this water was used without corrections in the OLI enthalpy calculations for the differences in temperature. The difference in the cooling water requirements for the condenser will be very small since the bulk of the heat load is required to condense the vapor while the requirements for the concentrate heat exchanger are likely ~15% lower than stated. The steam calculation assumed saturated steam inlet and saturated condensate at the system pressure of 85 psig, which was rounded to 7.0 bar absolute pressure for property determinations. A heat capacity of 4189 J/kg-C was utilized for the cooling water calculation, therefore stream enthalpies were not required and are not shown in Table 3. It should be noted that requirements shown in Table 3 will be approximately doubled once salt operation commences due to the additional condensate from the salt waste.

Table 3. Steam and Cooling Water Requirements

Stream	Condenser Cooling Water Inlet	Condenser Cooling Water Outlet	Steam Inlet	Steam Condensate Outlet	Heat Exchanger Cooling Water Inlet	Heat Exchanger Cooling Water Outlet
Stream Designation	9	10	11	12	13	14
Temperature, °C	35	50	165	165	35	50
Pressure, psig	-	-	85	85	-	-
Flowrate, kg/hr	29886	29886	1004	1004	109	109
Volume, L/hr	30013	30106	274050	1112	110	109
Enthalpy, kJ/hr	-	-	2762000	697100	-	-
Vapor fraction	0.0	0.0	1.0	0.0	0.0	0.0

2.1.2 Antifoam Additions

Antifoam additions are not shown in the material balance as the antifoam type and addition strategy have not been finalized. Dow Corning 2210 has been identified as a candidate, but evaluation of this antifoam is still in progress. Addition of antifoam to the process could have two major impacts on the material balance. First, degradation products from the antifoam could add significant quantities of silicon and organic carbon to the condensate. Second, large antifoam additions could add a significant amount of solids to the process and impact the composition of the concentrate recycle. Additional downstream impacts of the antifoam are discussed in the appropriate sections later in the text.

2.2 ETP WAC Review

A comparison of experimental runs and OLI model results to the ETP Waste Acceptance Criteria (WAC) indicates that some species have the potential to exceed the WAC limits for the evaporator condensate. Mercury, silicon, and Total Organic Carbon (TOC) were noted in the evaporator condensate above the WAC limits during at least one of the experimental runs. OLI simulations indicated that Hg, Co, Mn, Li, B, Pu, and TOC limits could be violated given certain feed compositions and that Co, Eu, Sb, and Am always violated the WAC limits. It should be noted that 0.1% entrainment was assumed during the OLI runs and that many of the WAC limits were exceeded as a result of entrainment, not evaporation from the vessel.

The mercury in the overhead condensate will be at saturation in the condensate and elemental mercury will collect in the condensate collection vessel sump, based on results from the experimental tests. The amount of mercury dissolved in the condensate will be a function of the pH of the condensate and the amount of oxidants and reductants present. Oxidation of elemental mercury promotes higher solubility as the Hg^{+2} ion is easily dissolved. Neutralization of the condensate stream to pH 7 would minimize mercury solubility, but addition mercury removal may still be necessary. Removal of mercury from the condensate can be accomplished via an ion exchange process.

The silicon in the evaporator condensate is primarily semi-volatile siloxane compounds. The source of the silicon in the recycle stream is the breakdown of the siloxane-based antifoam (IIT-747) added to the DWPF CPC process. The siloxane compounds are readily dissolved in water at low concentrations. Control of the foaming noted during the boil-down may require additional siloxane based antifoams to be added to the evaporator feed, increasing the potential to exceed the silicon ETP WAC limit in the condensate. Siloxane compounds are known to foul filtration systems, further complicating condensate processing. It should be noted that the silicon contained in the evaporator overheads would be expected to be in the overheads of any subsequent evaporation process. The amount of siloxanes in the evaporator condensate could be substantially increased from degradation products if a silicon based antifoam is selected to reduce foaming during evaporation.

Two sources exist for organic carbon compounds in the evaporator feed: formate from the formic acid additions during the SRAT/SME process and siloxanes from the IIT-747 antifoam. During the Scenario 5 run (a 50-50 blend of OGCT and decon solution)[3], the large amount of formate in the coil decontamination solution caused the formate in the evaporator condensate to exceed the TOC specification in the ETP WAC. Blending this feed in slowly may prevent excessive TOC in the evaporator overheads. Also, the amount of organics in the evaporator condensate could be substantially increased from degradation products of the antifoam selected to reduce foaming during evaporation.

Evaporation of actual DWPF recycle streams will be performed in the Shielded Cells and species that exceed the ETP WAC will be identified. Control of the other species that exceeded the ETP WAC limits due to entrainment should be accomplished by design of the evaporator to minimize entrainment. The 0.1% entrainment assumed during OLI runs represents a maximum DF of 1000 for any component in the feed. Higher DF's were achieved for selected species during the simulant boil-down tests and should be achievable for the process evaporator.

2.3 Evaporator Design Recommendations

Evaporators are available in a wide range of design configurations. Designs reviewed during initial evaluations of the evaporator design included forced and natural circulation evaporators as well as calandrias. The processing characteristics as well as strengths and weaknesses of each type of evaporator design are well studied. Design selection for commercial operations focus on efficiency, maintainability, and capital cost. While these requirements are important for the recycle evaporator, the ability of the design to be remotely operated and maintained must be the primary design consideration.

The recycle evaporator will be sized to process the maximum expected recycle volume from the DWPF process. The maximum volume assumes that both steam atomized scrubbers are in operation and that salt processing is adding significant amounts of water to the process. The evaporator must also be able to operate at reduced capacity when DWPF operation generates less than the maximum recycle volume. An evaporator which can handle "turn-down" is therefore desirable to allow varying process volumes to be processed without excessive cycling of equipment. This turn-down capability is also required since the evaporator should be designed to handle the expected doubling of recycle volume once salt waste processing begins. Until the salt waste is processed, the evaporator load will be ~ 50% of design capacity.

Selected recycles streams are high in solids content and most streams have some insoluble solids. The amount of concentration required to limit the recycle volume per SRAT cycle to 1000 gallons will be 30 to 60X and will lead to significant amounts (up to 10 wt %) of insoluble solids in the concentrate. The ability to handle solids during both operation, idle, and startup conditions is required.

Recycle solids include process frit. The frit enters the recycle through entrainment in the melter, sample flushes, and equipment decontaminations. The frit is extremely abrasive and requires special materials for equipment in high shear contact with the process fluid.

Foaming was noted during laboratory tests[3]. The foaming was most severe during initial startup, but some foam was present in all runs at the conclusion of testing. Evaluation of chemical antifoamers to reduce the amount of foam during processing is in progress, but the ability to process with some amount of foaming is required.

The combination of requirements for remotability, turn-down ability, solids handling, and foam mitigation lead to compromises in the evaporator design process. Forced circulation evaporators are generally the most flexible design for most evaporation processes. Drawbacks to the forced circulation include the susceptibility of pumps to erosion from abrasive particles, startup with settled solids, and remotability of maintenance. An air sparge system would be required to prevent settled solids during pump outages and frequent replacement of the recirculation pump would be required. The design of this evaporator precludes using an agitator to prevent solids settling since the agitator would not prevent settling in the piping to the heat exchanger.

Natural circulation evaporators rely on high boilup rates to provide circulation through the heat exchanger tubes to prevent fouling of the tubes. A natural circulation evaporator is not recommended due to the inability to operate in a turn-down condition. The design of this evaporator precludes using an agitator to prevent solids settling since the agitator would not prevent settling in the piping to the heat exchanger.

A calandria type evaporator similar to the DWPF SRAT/SME vessel has proven remotability and solids handling capabilities. Drawbacks include low efficiency, a greater potential for fouling the steam coils than a forced circulation system, and reduced foam handling characteristics. A central agitator would provide solids handling capability and allow startup after solids have settled. This agitator would also aid in the turndown ability of the evaporator by increasing circulation under low boiling conditions.

2.4 Chemical Process Cell Impacts

The evaporator concentrate will be recycled to the DWPF SRAT cycle. Approximately 1000 gallons of recycle will be added to each SRAT cycle if a concentration factor of 30X can be achieved during recycle evaporation. A number of different impacts have been identified and must be addressed prior to implementation of recycle to the SRAT cycle.

2.4.1 Frit in SRAT process

The recycle will introduce significant quantities of frit (approximately 2000 lb per coil decontamination) to the SRAT vessel when equipment decontamination solutions are blended with the recycle. The frit will increase erosion of the SRAT vessel and equipment, particularly the coil, coil supports, agitator blades, and process pumps. This increased erosion could cause premature failure of these components. Retrofitting the SRAT vessel to materials of construction similar to the SME would minimize the amount of erosion if premature failures are noted.

2.4.2 Silica in SRAT process

The recycle streams from equipment decontamination and HEME dissolution contain large amounts of silica. This increase in silica concentration could result in the formation of significant quantities of sodium alumino-silicates in the SRAT. However, the SRAT cycle is typically only at elevated temperatures during highly caustic conditions during the initial stages of the acid addition. The generation of the sodium alumino-silicate (NAS) solids is expected to be a relatively slow process since the silica must be leached into the solution from the frit prior to NAS formation. Given the limited time the SRAT is typically at temperature under caustic conditions, NAS solids formation is not expected to cause process problems, but an evaluation of the NAS formation should occur prior to incorporation of the recycle evaporator into the DWPF process.

2.4.3 Siloxane compound impacts

The condensate from the CPC process (SMECT condensate) contains siloxane compounds from the carryover and/or degradation of the 747 antifoam utilized in the SRAT and SME processes[6]. The amount of siloxane added will likely be insignificant when compared to the amount of siloxane added by the 747 antifoam, but this assumption should be confirmed by testing. Although these compounds will be volatile in the recycle evaporator, some fraction of these compounds will be recycled to the SRAT. The SMECT condensate from simulant testing of the CPC process foams when shaken, likely due to the presence of these compounds, therefore foaming in the SRAT cycle could be impacted; however, the foaming from these compounds will likely be controllable by the 747 antifoam.

2.4.4 Acid calculation impacts

The nitric acid content of the recycle will impact the acid requirements during the SRAT process. Nitric and formic acids are added during the SRAT cycle to adjust the chemistry and the rheology of the sludge. The amount of nitric and formic acids are added during the SRAT cycle is based on an acid stoichiometry calculation and experimentally determined stoichiometric factor. Operating windows are experimentally determined by evaluation of the minimum amount of acid required to destroy the nitrite in the feed while the maximum amount of acid is determined by evaluation of the hydrogen evolution during processing. The ratio of nitric acid to formic acid is determined by a prediction of the glass redox based on the amount of oxidants and reductants in the feed. Addition of nitric acid to the feed will impact the overall amounts of acid required and the ratios of nitric to formic acid utilized. Titration tests should be conducted on feeds blended with various recycle streams to quantify this impact.

2.4.5 Dilution of Feed

The recycle addition will add approximately 1000 gallons to the typical SRAT cycle. This recycle volume would increase as the salt processing streams are added to the DWPF process. Given a boil-up rate of 2000 lbs/hr obtained during SRAT processing, the additional 1000 gallons of water will require approximately four hours of additional boiling time in the SRAT cycle.

2.4.6 Additional Solids in SRAT Product

Several of the recycle streams contain significant amounts of solids and will increase the solids content of the SRAT product. If the recycle stream is 1000 gallons at 5 wt% solid and the SRAT product without recycle is 6000 gallons at 25 wt% solids, then the SRAT product with recycle will increase in solids content by approximately 1 wt%. This increase in solids is relatively small, but could have a large impact on rheological properties since the yield stress follows an exponential function with solids content[7]. The impact on rheology is difficult to predict and must be determined experimentally.

2.4.7 New antifoam agent

The recycle evaporator simulant testing indicated that antifoam additions will be required for recycle evaporator operation. The antifoams currently approved for use by DWPF (Dow Corning 544 and 747 antifoams) are not effective in the acidic conditions of the recycle evaporator. Preliminary evaluations of antifoams for the recycle evaporator have identified Dow Corning 2210 as a candidate. Further evaluation of this antifoam is currently in progress. Tests should be conducted to evaluate the impact of the selected antifoam on the DWPF process.

2.5 Melter Operation Impacts

The recycle stream could impact melter operations in two ways. First, the buildup of selected species, such as sulfate, in the recycle stream could result in exceeding individual solubility limits that are controlled utilizing the Process Composition Control System (PCCS) algorithms. These limits are imposed to reduce risks of melter processing issues such as excessive corrosion which limit the melter life. Second, the addition of an antifoam agent could lead to excessive TOC in the melter feed. The TOC melter feed limit is imposed to prevent flammable atmospheres in the melter offgas system. Assuming that a purge stream is applied to prevent buildup of semivolatiles like sulfate in the melter feed and the restriction of antifoam additions to meet the TOC requirements, no impacts on melter operation are expected.

2.6 Glass Impacts

Based on the projected SB4 compositions and candidate frits, assessments were made using predictions from models currently implemented in the DWPF PCCS over the waste loading (WL) interval of interest (25 – 60 wt%)[8]. The primary property predictions assessed included those for liquidus temperature (T_L), viscosity (η), and durability (normalized boron release – NL[B]). The predicted properties were then assessed against PCCS acceptance criteria to determine the projected operating windows for each system. The projected operating windows (defined in terms of a minimum and maximum waste loading over which all the predicted properties are acceptable) for each system are shown in Table 4. In addition, the property which restricted access to higher waste loading is shown in parentheses in the table. Nominal compositions (expressed in mass fractions) for each system are shown in Table 5.

Table 4. PCCS Results

System	Frit 418	Frit 320
1100 Canister Baseline	25 – 42 (T_L)	25 – 44 (low η)
Scenario 1: Typical Operation	25 – 40 (T_L)	25 – 42 (low η)
Scenario 2: Melter Idled – SMECT + Flushes	25 – 42 (T_L)	25 – 43 (low η)
Scenario 3: CPC Idled – OGCT Only	25 – 40 (T_L)	25 – 41 (low η)
Scenario 4: HEME Dissolution Added	25 (ΔG_P)	- (ΔG_P)
Scenario 5: 50% Decon – 50% OGCT	25 – 44 (T_L)	25 – 38 (low η)

Table 5. Melter Feed Oxide Compositions from Test Cases

Oxide	"Typical Operation"	Melter Idled – SMECT Only	CPC Idled – OGCT Only	HEME Dissolution Added	50% Decon Solution – 50% OGCT
Ag_2O	0.000000	0.000001	-	-	0.000027
Al_2O_3	0.214313	0.225030	0.210308	0.180070	0.200324
B_2O_3	0.000078	0.000044	0.000078	0.004447	0.000356
BaO	0.001526	0.001606	0.001501	0.001275	0.001422
CaO	0.021392	0.022289	0.021145	0.018693	0.022032
Ce_2O_3	0.001953	0.002046	0.001928	0.001638	0.001741
Cr_2O_3	0.002371	0.002485	0.002339	0.001986	0.002123
CuO	0.000796	0.000843	0.000777	0.000663	0.000885
Fe_2O_3	0.293099	0.263502	0.302117	0.218764	0.296344
Gd_2O_3	0.000012	0.000015	0.000007	-	0.000179
K_2O	0.009661	0.010134	0.009509	0.008728	0.009235
La_2O_3	0.000873	0.000914	0.000862	0.000732	0.000778
Li_2O	0.000019	0.000022	-	-	0.000726
MgO	0.019136	0.019529	0.019135	0.015943	0.022018
MnO	0.056391	0.058028	0.055933	0.046416	0.057333
MoO_3	0.000001	0.000001	0.000001	0.000075	0.000001
Na_2O	0.213023	0.221076	0.211040	0.360005	0.235171
NiO	0.035366	0.036665	0.035140	0.029381	0.032147
P_2O_5	0.000018	0.000038	-	-	0.000197
PbO	0.001560	0.001638	0.001536	0.001304	0.001475
SO_4^{2-}	0.010901	0.010953	0.011005	0.008849	0.010643
SiO_2	0.026023	0.027307	0.025346	0.024326	0.022959
SrO	0.000001	0.000001	0.000001	-	0.000006
ThO_2	0.000327	0.000343	0.000323	0.000274	0.000292
TiO_2	0.000202	0.000213	0.000198	0.000168	0.000198
U_3O_8	0.087115	0.091236	0.085996	0.073053	0.077662
ZnO	0.001225	0.001299	0.001194	0.001009	0.001392
ZrO_2	0.002618	0.002742	0.002584	0.002202	0.002335
Total	1.000000	1.000000	1.000000	1.000000	1.000000

The evaluation indicated that the "typical" recycle stream consisting of OGCT and SMECT condensate could be incorporated into the process without significantly impacting the glass properties as denoted by the minimal impact to the projected operating window as compared to the 1100 Canister Baseline option. In addition, the Scenarios 2 and 3 also did not significantly impact the projected operating windows. The solids content from these recycle streams was small in comparison to the amount of solids in a typical SRAT batch. Operating windows were slightly reduced by the recycle addition (only a 1% reduction in the Frit 320 system and a 2 or 3% reduction in “Typical” and OGCT only systems).

Addition of HEME recycle (33% by volume) in the Scenario 4 run dramatically impacted the projected operating windows (i.e., either complete elimination of the window for the Frit 320 based case or only a single targeted waste loading for the Frit 418 system). The amount of solids in this recycle stream was over three times the amount in the SMECT and OGCT streams. The high solids content precludes concentration of the stream by 30X, therefore it was assumed that the stream was concentrated to 50% solids content (~ 10X evaporation). The solids are primarily sodium nitrate (after neutralization of the stream to pH = 7). The increased sodium resulted in predictions of durability limiting both the Frit 320 and Frit 418 based systems.

The coil decontamination solution also contains a larger amount of solids than the OGCT and SMECT condensate, but evaporation of a 50-50 blend of OGCT condensate and coil decon solution resulted in an evaporator feed that could be concentrated 30X, as demonstrated in Scenario 5. This stream reduced the projected operating window for the Frit 320 system, but increased the operating window for the Frit 418 system.

2.7 Evaluation of Purge Requirements

A mass balance can be drawn around the DWPF process with recycle evaporation that highlights only the inlets and outlet streams, as shown in Figure 2. The process outlets are the glass product, stack emissions, mercury sumps, and the recycle evaporator overheads. Most of the cations (Fe, Al, Na) fed to the system exit as glass product while most of the anions (formate, nitrate) exit the stack after conversion to CO_2, N_2, N_2O, or NO_x. Water, along with other semivolatiles, will typically exit the DWPF process through the recycle evaporator.

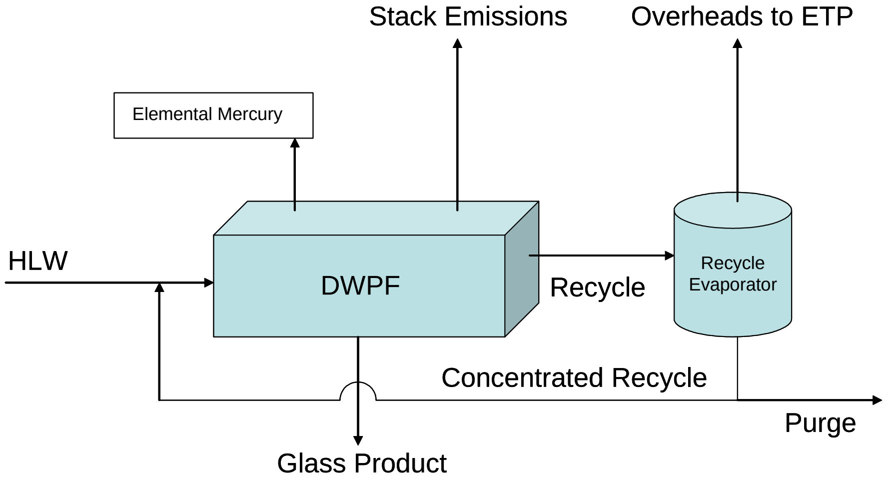

Figure 2. DWPF Mass Balance Flowsheet with Recycle Evaporation

Selected species, such as sulfate, chlorine, fluorine, and mercury are volatile in the melter and are efficiently captured by the offgas scrubbing process. These species are not appreciably volatile when dissolved in water and will remain in the evaporator concentrate from the recycle evaporator. Buildup of these components in the DWPF system will occur when the recycle stream is added. The amount of buildup in the melter feed is dependent on the amount in the feed, the percent removed in the glass or other streams, the percent captured by the offgas system, and the amount of purge to the tank farm. Mercury represents a special case due to the reduction and removal of mercury during the SRAT cycle. This additional purge point will prevent accumulation of mercury.

The initial evaluation of the recycle evaporator assumed that a 10% purge stream would adequately prevent accumulation of these species. However, the sludge washing strategy has changed significantly since the initial evaluation and the incoming feed to the DWPF contains much higher levels of these soluble species. Additional streams added to the waste prior to vitrification (e.g. Np and Pu streams from separations decommissioning processes) can also add increased levels of soluble species. Sulfur concentration, in particular, is much higher in the current sludge batch and evaluations were conducted to raise the sulfur limit in preparations for this batch. Given the variations in feed compositions, a single purge stream calculation is no longer adequate. An approach is presented below that determines the purge required based on the ratio of the feed concentration of a contaminant and the process limit.

Assuming that the inlet concentration of one of these components is equal to 1 (units cancel during this calculation and are unnecessary), a mass balance can be performed for various assumptions of percentages in the glass product and purge amounts. For this evaluation, capture by the offgas system was assumed to be 100%. The mass balance was performed assuming that all recycle from a cycle is recycled during the next feed cycle. The results of the mass balance calculations are shown in Table 6, Table 7, Table 8, and Table 9.

Table 6. Accumulation of Contaminant Assuming 75% Glass Retention

Purge (% of Recycle)	0	10	20	30	40	50	60	70	80	90	100
Feed Cycle	Concentration of Contaminant in Melter Feed										
1	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
2	1.25	1.23	1.20	1.18	1.15	1.13	1.10	1.08	1.05	1.03	1.00
3	1.31	1.28	1.24	1.21	1.17	1.14	1.11	1.08	1.05	1.03	1.00
4	1.33	1.29	1.25	1.21	1.18	1.14	1.11	1.08	1.05	1.03	1.00
5	1.33	1.29	1.25	1.21	1.18	1.14	1.11	1.08	1.05	1.03	1.00
6	1.33	1.29	1.25	1.21	1.18	1.14	1.11	1.08	1.05	1.03	1.00
7	1.33	1.29	1.25	1.21	1.18	1.14	1.11	1.08	1.05	1.03	1.00
8	1.33	1.29	1.25	1.21	1.18	1.14	1.11	1.08	1.05	1.03	1.00
9	1.33	1.29	1.25	1.21	1.18	1.14	1.11	1.08	1.05	1.03	1.00
10	1.33	1.29	1.25	1.21	1.18	1.14	1.11	1.08	1.05	1.03	1.00

Table 7. Accumulation of Contaminant Assuming 50% Glass Retention

Purge (% of Recycle)	0	10	20	30	40	50	60	70	80	90	100
Feed Cycle	Concentration of Contaminant in Melter Feed										
1	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
2	1.50	1.45	1.40	1.35	1.30	1.25	1.20	1.15	1.10	1.05	1.00
3	1.75	1.65	1.56	1.47	1.39	1.31	1.24	1.17	1.11	1.05	1.00
4	1.88	1.74	1.62	1.52	1.42	1.33	1.25	1.18	1.11	1.05	1.00
5	1.94	1.78	1.65	1.53	1.43	1.33	1.25	1.18	1.11	1.05	1.00
6	1.97	1.80	1.66	1.54	1.43	1.33	1.25	1.18	1.11	1.05	1.00
7	1.98	1.81	1.66	1.54	1.43	1.33	1.25	1.18	1.11	1.05	1.00
8	1.99	1.82	1.67	1.54	1.43	1.33	1.25	1.18	1.11	1.05	1.00
9	2.00	1.82	1.67	1.54	1.43	1.33	1.25	1.18	1.11	1.05	1.00
10	2.00	1.82	1.67	1.54	1.43	1.33	1.25	1.18	1.11	1.05	1.00

Table 8. Accumulation of Contaminant Assuming 25% Glass Retention

Purge (% of Recycle)	0	10	20	30	40	50	60	70	80	90	100
Feed Cycle	Concentration of Contaminant in Melter Feed										
1	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
2	1.75	1.68	1.60	1.53	1.45	1.38	1.30	1.23	1.15	1.08	1.00
3	2.31	2.13	1.96	1.80	1.65	1.52	1.39	1.28	1.17	1.08	1.00
4	2.73	2.44	2.18	1.95	1.74	1.57	1.42	1.29	1.18	1.08	1.00
5	3.05	2.65	2.31	2.02	1.78	1.59	1.43	1.29	1.18	1.08	1.00
6	3.29	2.79	2.38	2.06	1.80	1.60	1.43	1.29	1.18	1.08	1.00
7	3.47	2.88	2.43	2.08	1.81	1.60	1.43	1.29	1.18	1.08	1.00
8	3.60	2.94	2.46	2.09	1.82	1.60	1.43	1.29	1.18	1.08	1.00
9	3.70	2.99	2.47	2.10	1.82	1.60	1.43	1.29	1.18	1.08	1.00
10	3.77	3.02	2.48	2.10	1.82	1.60	1.43	1.29	1.18	1.08	1.00
11	3.83	3.04	2.49	2.10	1.82	1.60	1.43	1.29	1.18	1.08	1.00
12	3.87	3.05	2.49	2.10	1.82	1.60	1.43	1.29	1.18	1.08	1.00
13	3.90	3.06	2.50	2.10	1.82	1.60	1.43	1.29	1.18	1.08	1.00
14	3.93	3.06	2.50	2.11	1.82	1.60	1.43	1.29	1.18	1.08	1.00
15	3.95	3.07	2.50	2.11	1.82	1.60	1.43	1.29	1.18	1.08	1.00

Table 9. Accumulation of Contaminant Assuming 0% Glass Retention

Purge (% of Recycle)	0	10	20	30	40	50	60	70	80	90	100
Feed Cycle	Concentration of Contaminant in Melter Feed										
1	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
2	2.00	1.90	1.80	1.70	1.60	1.50	1.40	1.30	1.20	1.10	1.00
3	3.00	2.71	2.44	2.19	1.96	1.75	1.56	1.39	1.24	1.11	1.00
4	4.00	3.44	2.95	2.53	2.18	1.88	1.62	1.42	1.25	1.11	1.00
5	5.00	4.10	3.36	2.77	2.31	1.94	1.65	1.43	1.25	1.11	1.00
6	6.00	4.69	3.69	2.94	2.38	1.97	1.66	1.43	1.25	1.11	1.00
7	7.00	5.22	3.95	3.06	2.43	1.98	1.66	1.43	1.25	1.11	1.00
8	8.00	5.70	4.16	3.14	2.46	1.99	1.67	1.43	1.25	1.11	1.00
9	9.00	6.13	4.33	3.20	2.47	2.00	1.67	1.43	1.25	1.11	1.00
10	10.0	6.51	4.46	3.24	2.48	2.00	1.67	1.43	1.25	1.11	1.00
11	11.0	6.86	4.57	3.27	2.49	2.00	1.67	1.43	1.25	1.11	1.00
12	12.0	7.18	4.66	3.29	2.49	2.00	1.67	1.43	1.25	1.11	1.00
13	13.0	7.46	4.73	3.30	2.50	2.00	1.67	1.43	1.25	1.11	1.00
14	14.0	7.71	4.78	3.31	2.50	2.00	1.67	1.43	1.25	1.11	1.00
15	15.0	7.94	4.82	3.32	2.50	2.00	1.67	1.43	1.25	1.11	1.00

Thus, if a contaminant is only 50% retained in the glass and is non-volatile in an evaporator, a purge would be required unless the process limit is twice the amount in the initial feed stream. The amount of purge required is dependent on the ratio of the process limit to the initial feed. If the limit is 1.7X the feed concentration, then a 20% purge stream would allow the process to remain under the limit, since the process reaches a steady-state value of 1.67 for these conditions. The number of cycles required to reach steady-state varies as the glass retention and purge are changed, but most conditions reach steady-state in just a few cycles.

2.8 Recycle Volume Impacts

The addition of the recycle stream to the SRAT process will increase the amount of condensate generated during CPC processing. The increased condensate will be added to the recycle stream, increasing the amount of recycle. A small amount of water accumulation will occur in the system due to the increase recycle volumes, but not to the extent reported during the initial study[1]. The initial study indicated an exponential increase in recycle volume due to the accumulation of additional condensate as more recycle is returned to the SRAT during each cycle. As shown in Figure 3, the amount of recycle generated reaches a steady-state volume in approximately five SRAT cycles and exponential volume increase is avoided by the removal of water in the evaporator overheads. This calculation assumed a worst-case scenario of 10 gallons of recycle for each gallon of HLW fed and a 10X concentration factor during recycle evaporation.

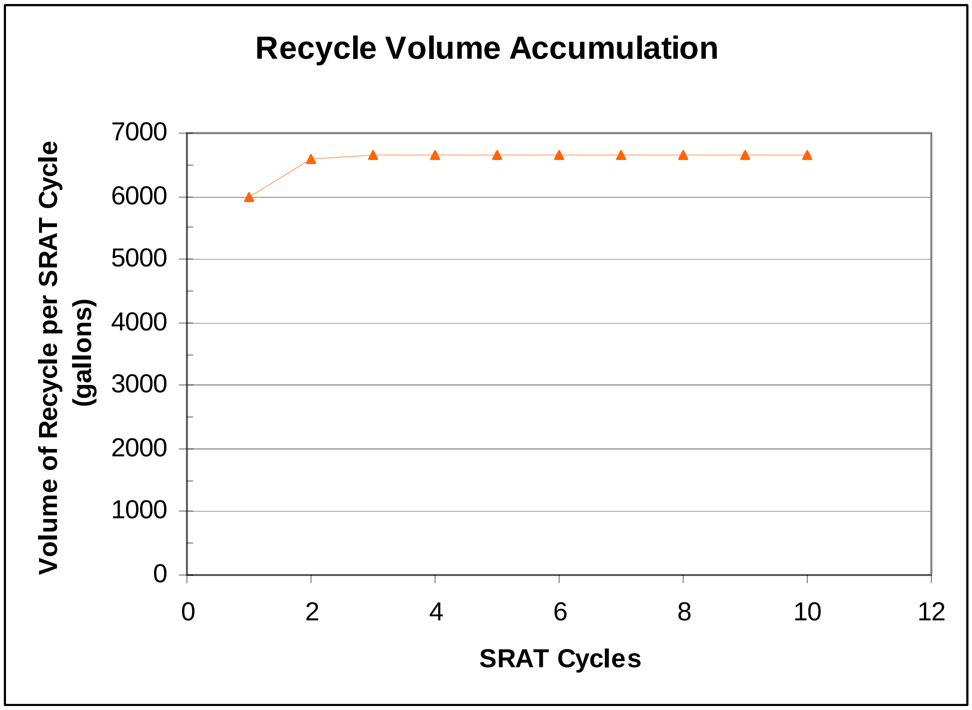

Figure 3. Water Accumulation during Recycle Evaporation

3.0 CONCLUSIONS

- Approximately 1000 kg/hr of steam and 30,000 kg/hr of cooling water were required for the recycle evaporator as determined by an OLI simulation on the "typical" recycle stream. These requirements will be doubled once salt waste processing is implemented at DWPF.
- Recycling the Off Gas Condensate Tank (OGCT) and Slurry Mix Evaporator Condensate Tank (SMECT) condensate does not significantly impact the glass properties or operating windows for the SB4 system.
- Recycling the coil decontamination solution can impact the glass properties and for Frit SB4/Frit 320 system will reduce the operating window significantly while the operating window is increased for the SB4/Frit 418 system.
- Recycling the High Efficiency Mist Eliminator (HEME) dissolution solution can eliminate the operating window for the SB4/Frit 320 and SB4/Frit 418 systems due to the high sodium content. Alternative frits may be identified that could restore the operating windows.
- The recycle purge requirements will vary from sludge batch to sludge batch and are dependent upon the ratio of the feed concentration to the process limits for DWPF and the percentage of the contaminant removed in the glass stream, offgas, and/or recycle condensate.
- The recycle stream could have significant impacts on the SRAT cycle in a variety of ways. Impacts include: changes to the acid addition strategy, increased erosion of process equipment due to frit introduction, potential change in rheology properties from insoluble solids added from the recycle, extended processing time due to higher boil-off volumes, and the foaminess of the SRAT slurry due to antifoam degradation products.
- The condensate from the recycle evaporator could exceed the ETP Waste Acceptance Criteria (WAC) for mercury, silicon, and Total Organic Carbon (TOC) based on simulant tests. Treatment for mercury removal could include pH adjustment and/or ion exchange processes.

4.0 RECOMMENDATIONS

- Further evaluation of the amount of HEME dissolution material that can be processed in one SRAT cycle without significantly impacting the glass composition operating window is required or alternative disposal routes for this material needs to be identified.
- A calandria evaporator is recommended based on the proven remotability of this system and the ability to utilize an agitator to mitigate solids settling. It is assumed that a suitable antifoam agent can be identified to mitigate any foaming concerns.
- The impact of the recycle stream on the DWPF SRAT cycle should be evaluated. This evaluation should be completed after the selection of the antifoam agent to be utilized for the recycle evaporator.
- The treatment of condensate for mercury removal should be evaluated to determine if pH adjustment will reduce mercury concentrations to required levels.

- Evaluation of condensate treatment options for siloxane and TOC species should be performed. This evaluation will require characterization of the species present in the condensate.

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