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Evaluation of Proposed Flowsheet Changes for the Highly Enriched Uranium Blenddown Program

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Abstract

H Canyon is considering a flowsheet change for Plutonium (Pu) Contaminated Scrap (PuCS) material. The proposed change is to route dissolved PuCS material directly to a uranium (U) storage tank. As a result, the PuCS solution will bypass Head End and First U Cycle, and will be purified by solvent extraction in Second U Cycle. The PuCS solution contains appreciable amounts of boron (B) and fluoride (F^-), which are currently at trace levels in the U storage tank. Though unlikely, if the B concentration in the U storage tank were to reach 1.8 g B/g U, the entire contents of the U storage tank would likely require a second pass through Second U Cycle to provide sufficient decontamination to meet the Tennessee Valley Authority (TVA) Blend Grade Highly Enriched Uranium (HEU) specification for B, which is 30 μg/g U. In addition, Second U Cycle is expected to provide sufficient decontamination of F^- and Pu regardless of the amount of PuCS solution sent to the storage tank.

Though aluminum (Al) is not present in the PuCS solution, B can be credited as a complexant of F^-. Both stability constants from the literature and Savannah River National Laboratory (SRNL) corrosion studies were documented to demonstrate that B complexation of F^- in nitric acid solutions is sufficient to prevent excessive corrosion. Though B and Al complex F^- to a similar degree, neither completely eliminates the presence of free F^- in solution. Therefore, a limited amount of corrosion is expected even with complexed F^- solutions. Tanks maintained at ambient temperature are not expected to experience significant corrosion. However, the Low Activity Waste (LAW) evaporators may be subjected to a corrosion rate of about 25 mils per year (mpy) as they reach their highest F^- concentrations. The feed adjustment evaporator would only be subjected to the corrosion rate of about 25 mpy in the latter stages of the PuCS campaign. An issue that must be addressed as part of the proposed PuCS flowsheet change is that B has limited solubility in concentrated nitric acid solutions. As the proposed PuCS campaign progresses, the B concentration will increase in the U storage tank, in Second U Cycle feed, and in the 1DW stream sent to the LAW evaporators. Limitations on the B concentration in the LAW evaporators will be needed to prevent formation of boron-containing solids.

Introduction

As part of the Highly Enriched Uranium (HEU) Blenddown campaign, the Savannah River Site (SRS) began dissolving, purifying and blending HEU fuel tubes in 2003. The HEU solution is blended with natural uranium to achieve a 4.95% enriched product solution that is converted to fuel and burned in a Tennessee Valley Authority (TVA) reactor to produce electricity. A large portion of the HEU designated for Blenddown by the Department of Energy (DOE) was

aluminum-clad fuel. As the fuel portion of the campaign approached completion, SRS prepared for flowsheet changes to dissolve metal scrap. This paper describes the evaluation performed prior to implementing the needed flowsheet changes. The metal scrap is often referred to as plutonium-contaminated scrap (PuCS). A schematic of the flowsheet is presented in Figure 1.

Proposed Flowsheet Change

The flowsheet for uranium (U) fuel involves dissolution in nitric acid with a mercuric nitrate catalyst to promote aluminum (Al) dissolution. The resulting solution is centrifuged then purified via two cycles of solvent extraction (i.e., 1st and 2nd U Cycle). The flowsheet for metal scrap involves dissolution in nitric acid/potassium fluoride (KF) with boron (B) as a neutron poison. Initially, dissolved metal scrap was blended with dissolved uranium fuel and processed in the same way. The proposed flowsheet change involved not blending the dissolved metal, but rather placing it in a U storage tank and purifying it via one cycle of solvent extraction (2nd U Cycle). Prior to implementation, an evaluation was needed to determine if the 2nd U Cycle alone would provide sufficient decontamination of B, F^- and Pu to meet TVA Blend Grade HEU specifications. Table 1 lists key characteristics of the dissolved PuCS solution and the current U storage tank solution.

Table 1. Key Characteristics of PuCS Solution and Current U Storage Tank Solution.

PuCS Solution	U Storage Tank Contents
1,800 gallons per dissolution	80,000 gallons
2.5 g U/L	8.4 g U/L
3.5 M nitric acid	0.7 M nitric acid
0.03 M F^-	trace
2.5 g B/L	trace
1.0 x 10^4 Pu dpm/mL	1.0 x 10^4 Pu dpm/mL

As shown in Table 1, the PuCS solution contains two species, B and F^-, which are currently at trace levels in the U storage tank. If PuCS solution is re-routed to the U storage tank, the concentrations of both B and F^- in the tank would steadily increase throughout the campaign. Periodic removals from the U storage tank during the PuCS campaign would yield higher B and F^- concentrations in the U tank. Thus, an evaluation was needed as to what B and F^- concentrations could be attained in the U storage tank without requiring more than one cycle of solvent extraction.

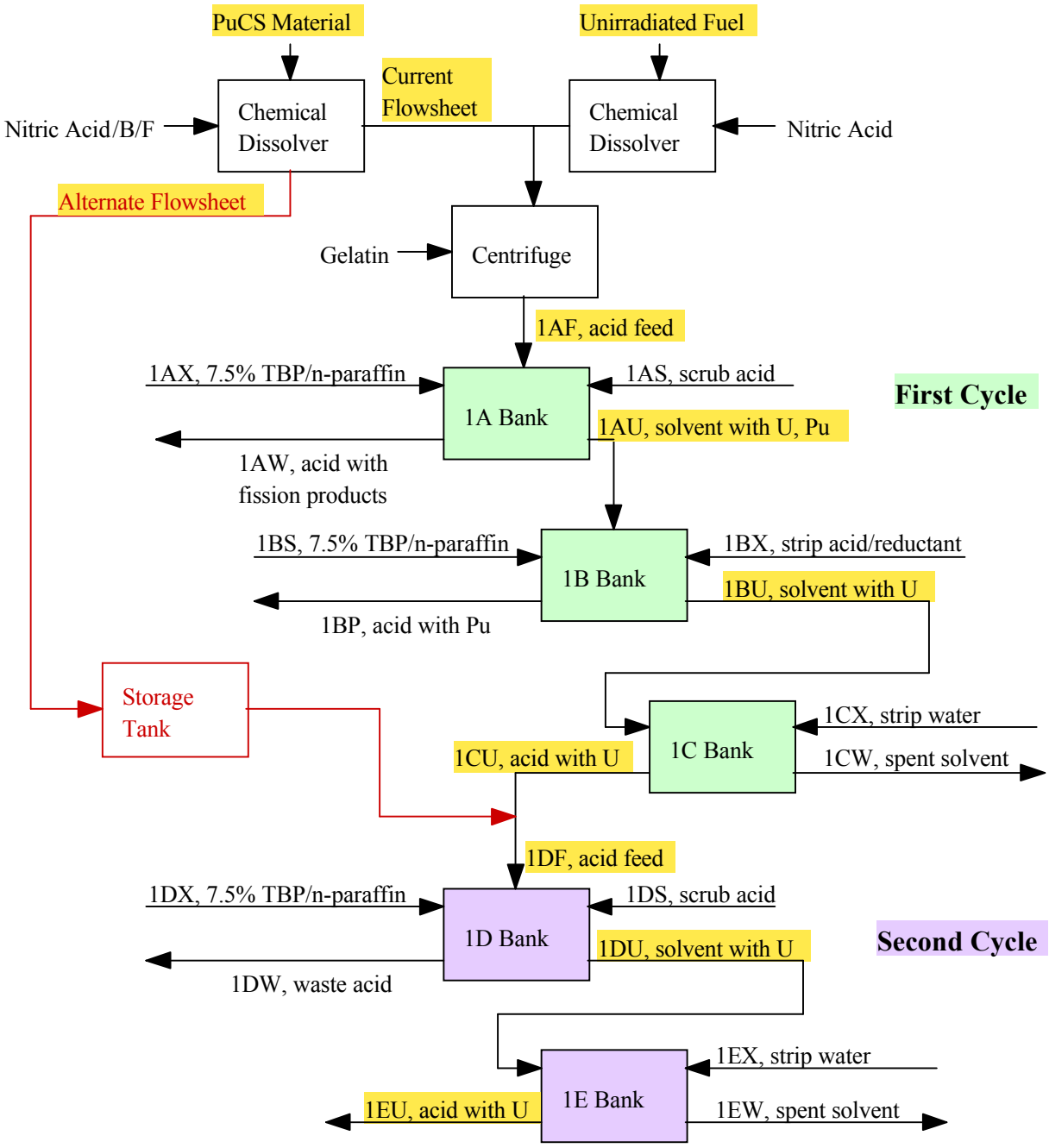

Figure 1. Flowsheet for U purification [1] with proposed changes.

Decontamination

Boron Decontamination

Recent H Canyon processing experience with blended PuCS/fuel solution demonstrated that most B was rejected by the A Bank in First U Cycle and that the resulting B concentration in the 1BP stream was equivalent to 60 µg/g U, which is two times the TVA Blend Grade specification. The First Cycle product stream, 1CU, was < 7 µg B/g U, which was well below the 30 µg/g U specification for B. Stated another way, the decontamination factor[i] (DF) for B in A Bank was slightly more than 1000 and the combined First Cycle DF was over 20,000 and may have been higher but the B level in the product was below detection limits. These DF values represent the combined effects of the distribution coefficient, number of stages, relative organic/aqueous flow rate, stage efficiency, entrainment, and interactions with other chemicals in the process. Since so many factors affect DF, it is difficult to predict if the B DF in Second U Cycle would be better or worse than that of First U Cycle.

Distribution coefficients[ii] for hydrogen fluoride (HF) and boric acid (H_3BO_3) between nitric acid solutions and tri-butyl phosphate (TBP) are shown in Table 3. The distribution coefficients for B (and HF) in 7.5 % TBP, as used in H Canyon, would be significantly less than the 100% (or 30%) TBP values because the diluent material, n-paraffin, which makes up the remaining 92.5 % of the solvent, does not extract either B or HF. Hence, a high degree of B decontamination would be expected in First or Second U Cycle. Based on the distribution coefficients in Table 3, it appears that stage efficiency and/or entrainment are the main factors limiting the B DF in A Bank to 1000.

Table 3. Distribution Coefficients for HF and H_3BO_3 between Nitric Acid Solutions and TBP [2].

Nitric acid mol/L	Distribution coefficient, $D_{o/a}$ 30% TBP	100% TBP
	HF	H_3BO_3
0.5	0.4	
1	0.3	0.15
2	0.2	0.08
3	0.15	0.035
4	0.1	0.007
5	0.05	
6	0.03	

Using the distribution coefficients in Table 3, and nitric acid distribution coefficients from Thompson et al [3], calculations were performed using a SASSE model (a Spreadsheet Algorithm for Stagewise Solvent Extraction [4]). Decontamination factors were calculated for typical A Bank and D Bank stage separation efficiencies and organic in aqueous and aqueous in

[i] Decontamination factor or DF = Volumetric feed concentration/volumetric product concentration.
[ii] Distribution coefficient or Do/a = Concentration in organic phase/concentration in aqueous phase at equilibrium.

organic entrainments. Stage efficiencies of 50% and 70% and entrained fractions of 0.02 and 0.05 (for both the organic and aqueous phases) were selected as representative values for A and D Bank operation. SASSE calculation results for selected cases are shown in Table 4. In all cases, the DF's for D Bank exceed those for A Bank. In particular, the DF for B is about three times greater in D Bank at a stage efficiency of 0.5 and entrained fractions of 0.05, which are the same parameters that yield an A Bank DF close to the observed value of 1000. Parametric calculations indicate that the improved separation efficiency in D Bank may be attributed to the relatively higher D Bank aqueous scrub flow rate.

Table 4. Calculated DF's for A Bank and D Bank

Stage Efficiency	Entrained Fraction	A Bank DF		D Bank DF	
		Boron	Fluoride	Boron	Fluoride
0.5	0.02	3400	11	6300	34
0.5	0.05	777	7.5	2200	28
0.7	0.02	58,000	39	60,000	67
0.7	0.05	1600	18	7100	47

Table 4 compares A and D Banks assuming that stage efficiency and entrainment levels are the same in both banks. Though efficiency and entrainment are likely to be similar in the two banks, the actual levels have not been determined. Therefore, it is reasonable to consider a few estimates of the B DF that may be achieved by Second U Cycle. These are shown in Table 5, along with the expected point at which a second solvent extraction pass would be required for the solution in the U storage tank. The obvious consequence of a second pass is an increased processing burden that would slow production significantly. For traditional mixer-settler performance, a stage efficiency of 70% and an entrainment fraction of 0.02 is often assumed. If these assumed values are correct, the PuCS solution would never reach a point where a second pass is needed. The PuCS solution starts with a 1:1 U:B ratio. A D Bank DF of 60,000 would provide sufficient purification for a feed concentration of 1.8 g B/g U. Therefore, B contamination is not expected to be a problem during the PuCS campaign after flowsheet changes are made.

Table 5. Effect of B Decontamination Factor on Need for Second Pass of Solvent Extraction for Solution in U Storage Tank.

Assumed B DF in Second U Cycle	B concentration in tank that would require a second pass
1000	3×10^4 µg B/g U
2000	6×10^4 µg B/g U
5000	1.5×10^5 µg B/g U
60,000	1.8×10^6 µg B/g U

Fluoride and Plutonium Decontamination

Recent H Canyon processing experience has shown that a single pass of the current U storage tank solution through Second U Cycle provides sufficient Pu decontamination to meet the TVA Blend Grade HEU specification for Pu of 400 µg/g U. Since the Pu concentration of PuCS solution is similar to the concentration of the current U storage tank solution, Second U Cycle should provide sufficient Pu decontamination regardless of the amount of PuCS solution sent to the U storage tank. However, purification by Second U Cycle may become more difficult if PuCS solution picked up additional Pu or other isotopes, such as neptunium-237, during transfer. Therefore, flushing of transfer lines prior to use in the proposed PuCS flowsheet change is recommended.

The DFs for F^- shown in Table 4 are all less than 70, but are based on the distribution coefficients of pure nitric acid/HF solutions. In solutions containing B and other components, F^- forms complex ionic species that are polar and are not as likely to extract. A recent sample from First U Cycle showed a DF for F^- greater than 40 for A Bank alone. (Since the F^- content in the 1BP sample was below the method detection limit, the actual DF would be greater than 40.) Due to the presence of B and other species, as well as the A Bank DF for F^- of 40, it is reasonable to expect a D Bank DF for F^- of at least 40. The TVA Blend Grade HEU specification for F^- is a relatively high value of 1,150 µg/g U, and the F^- concentration in the U storage tank is expected to approach 32,000 µg/g U at the end of the PuCS campaign (see Table 2). Thus, a DF for F^- of 40 would be sufficient for processing PuCS solutions from the U storage tank.

Effect of Solids

The PuCS material has no Al which is a normal source of silica. Silica content should be limited to small impurity levels from the carbon steel cans which contain the PuCS material. Historically, First Cycle feed is about 20 ppm silicon (Si). A sample of PuCS solution was analyzed and found to have less Si content than is normally seen in current First Cycle feed. In addition, the sample was visually inspected and solids were not observed. Therefore, it is unlikely that the PuCS solution has appreciable solids. However, insoluble corrosion products can build up in tanks and process piping. To ensure the success of the PuCS campaign, affected tanks and piping should be flushed to remove solids and other undesirable constituents prior to the addition of PuCS solution to the U storage tank.

Complexation of Fluoride by Boron

At the Savannah River Site, the dissolution of metals and refractory materials containing Pu or U is typically performed in nitric acid solutions with the addition of small amounts of F^-. Fluoride is added as a catalyst to increase the rate of dissolution. Once the dissolution is complete, the F^- is complexed with Al to reduce the corrosion of downstream processing equipment. With the addition of Al, evaporation of the waste solutions from purification processes can be performed without unacceptable corrosion in the evaporators. The PuCS material is currently being

dissolved using a 3.5 M nitric acid solution containing 0.03 M F^-. When the PuCS solution is blended with the solution from the dissolution of reactor fuel, the Al contained in the fuel is more than sufficient to complex the small amount of F^-. However, if the PuCS solution is transferred directly to the U storage tank instead of blending with the fuel, no Al is added to the solution. One option for the subsequent processing of the PuCS solution is to add Al to comply with the normal procedure for solutions containing F^-. The second option is to take credit for the complexation of F^- by the B which is added to the solution as a nuclear safety poison. However, the adequacy of B for the prevention of excessive corrosion during evaporation of the waste solutions must be assured.

The stability of F^- complexes formed by B and Al was evaluated by Hammer [5] as a function of temperature in 0.96 M and 2.88 M nitric acid. The data are reproduced in Table 6. Inspection of the data shows that the stability constants for the highest order F^- complexes for B are greater than those for Al. This information indicates that the free F^- concentration in the B system will not be more than the concentration in the Al system (assuming equal concentrations of B and Al). During the development of a flowsheet for the dissolution of sand, slag, and crucible (SS&C) residues in the F Canyon dissolvers, free F^- concentrations were measured in a solution of 9.3 M nitric acid containing 0.025 M total F^- as a function of the B concentration [6]. At nominally 0.05 M B (0.5 g B/L), the free F^- concentration was approximately 40 mg/L and dropped to approximately 10 mg/L when the B concentration was increased to 0.23 M (2.5 g/L). For comparison, the free F^- concentration measured in a solution containing 9.3 M nitric acid, 0.05 M F^-, and nominally 0.05 M Al (1.5 g/L) was approximately 80 mg/L. When the Al concentration was increased to nominally 0.25 M (7 g/L), the free F^- concentration dropped to approximately 10 mg/L. These data demonstrate that the complexation of F^- in nitric acid solutions by B is similar in effectiveness to Al for equal concentrations of the cations, and B can be credited as a complexant for F^-.

Table 6. Stability Constants for Al and B Fluoride Complexes in 0.96 and 2.88 M Nitric Acid
(Reproduced from Hammer [5])

System	25 °C		35 °C		45 °C		60 °C	
	0.96 M	2.88 M	0.96 M	2.88 M	0.96 M	2.88 M	0.96 M	2.88 M
Al^{3+} — HF — HNO_3								
$Al^{3+} + HF = AlF^{2+} + H^+$	1607	1507	1512	1301	1313	1051	1152	1143
$Al^{3+} + 2HF = AlF_2^+ + 2H^+$	$1.96x10^5$	$2.16x10^5$	$2.04x10^5$	$1.83x10^5$	$1.58x10^5$	$1.99x10^5$	$8.61x10^4$	$1.16x10^5$
$Al^{3+} + 3HF = AlF_3 + 3H^+$	$1.14x10^6$	$2.26x10^6$	$1.36x10^6$	$2.24x10^6$	$8.00x10^5$	$3.65x10^6$	$5.71x10^5$	$5.57x10^5$
$Al^{3+} + 4HF = AlF_4^- + 4H^+$	$1.62x10^6$		$\sim 1.39x10^6$				$\sim 7x10^4$	
H_3BO_3 — HF — HNO_3								
$H_3BO_3 + HF = H_4BO_3F$	15.2	20	10.5	11	11	15	1.7	2.1
$H_3BO_3 + 2HF = H_3BO_2F_2 + H_2O$	460	330	0	235	87	60	114	179
$H_3BO_3 + 3HF = H_2BOF_3 + 2H_2O$	$7.03x10^5$	$9.58x10^5$	$3.65x10^5$	$3.41x10^5$	$1.75x10^5$	$2.24x10^5$	$3.01x10^4$	$6.15x10^4$
$H_3BO_3 + 4HF = H^+ + BF_4^- + 3H_2O$	$3.80x10^7$	$1.24x10^8$	$2.08x10^7$	$8.06x10^7$	$1.13x10^7$	$4.06x10^7$	$1.52x10^6$	$2.54x10^6$

General Processing Concerns

While B would aid the proposed flowsheet change by complexing F^- and thus limit corrosion, B presents challenges in that it has limited solubility in concentrated nitric acid solutions. Literature data are provided in Table 7 [7].

Table 7. Solubility of H_3BO_3 in Aqueous Nitric Acid at 25 °C.

Nitric Acid M	Solubility of H_3BO_3 M	Solubility of B g/L
0.078	0.882	9.54
0.534	0.803	8.68
1.58	0.669	7.23
3.12	0.544	5.89
3.81	0.498	5.38
4.70	0.447	4.83
8.41	0.296	3.20
10.95	0.2305	2.49
15.18	0.197	2.13

Since the LAW evaporator (which will eventually receive the B from PuCS solution) has multiple feed sources with different concentrations of nitric acid and other constituents, it is difficult to prescribe operational limits. However, as the proposed PuCS campaign progresses, appropriate limitations on the concentration factor achieved in the LAW evaporators would be needed to prevent formation of boron-containing solids.

Corrosion

During the development of a related flowsheet, a series of coupon immersion tests was performed to measure corrosion rates for various solutions containing nitric acid, F^-, B, and Al [6]. The test coupons were fabricated from 304L stainless steel; both welded and non-welded coupons were used. The solution temperatures were maintained at 85-90 °C. Selected data from these tests are reproduced in Table 8.

Table 8. Selected 304L Corrosion Rates in Nitric Acid/Fluoride Solutions

Solution Number	Nitric Acid (M)	Potassium Fluoride (M)	B (M)	Al (M)	Free Fluoride (mg/L)	Corrosion Rate (mpy)
1	9.3	0.05	0	0	128	77.5
2	9.3	0.05	0.23	0	17	24.7
3	9.3	0.1	0.23	0	24	26.5
6	9.3	0.2	0.23	0.8	10	3.90

mpy ≡ mils per year

The B and F^- concentrations used in solution 2 most nearly correspond to concentrations used in the PuCS dissolving solution. In this test, the free F^- concentration and corrosion rate were 17 mg/L and 24.7 mpy, respectively. It is expected that a slight decrease in the F^- concentration would lower these values. The nitric acid concentration in solution 2 is also nearly three times greater than the concentration (3.5 M) used for the dissolution of the PuCS material. A lower nitric acid concentration would be expected to result in a lower corrosion rate. The corrosion rates of 304L stainless steels are strongly impacted by both the F^- and nitric acid concentrations resulting in increasing rates for increasing concentrations.[8] The corrosion rates given in Table 8 were also measured at slightly lower temperatures (85-90 °C) than would be experienced in the LAW evaporators, which operate at boiling temperatures greater than 100 °C. Higher corrosion rates would be expected at the higher temperatures. For boiling solutions of 6 M nitric acid at 0.01 M HF and no Al or B, the corrosion rate for 304L stainless steel is 50 mpy.[9] It should be noted that the small amount of HF in the boiling solution would approach equilibrium with the vapor phase. Therefore, a small amount of HF would collect in evaporator overheads. For process vessels at ambient temperature, the corrosion induced by 0.01 M F^- and about 1 M nitric acid is quite low, well under 10 mpy even without complexation.[9]

The greatest degree of fluoride-induced corrosion is expected to occur in the LAW evaporators which will receive the aqueous 1DW waste stream containing B, Fe and F^- from processing of the PuCS. The corrosion could impact both tank walls and equipment exposed to the solution. A concentration factor up to 30x can be attained as the evaporator overheads are sent to acid recovery, although a 15x concentration is expected to be more typical. The estimated concentrations of B, Fe, F^- and nitric acid are shown in Table 9 for the waste feed and different evaporator concentration factors. These data are conservative considering that the waste feed figures shown below are prior to water addition to the evaporator feed tank.

Table 9. Selected 304L Corrosion Rates in Nitric Acid/Fluoride Solutions

Constituent	Concentration (M(g/L))		
	Feed	15 X	30X
Fe	0.005 (0.3)	0.081 (4.5)	0.16 (9)
B	0.056 (0.6)	0.83 (9)*	1.67 (18)*
F^-	0.007 (0.14)	0.12 (2.2)	0.22 (4.2)
Nitric acid	1	7	8

* Note that B solubility is 3-4 g/L in 7-8 M HNO_3 at 25 °C

Free Fluoride Concentration

The free F^- concentration, which is impacted by the soluble B concentration, is the primary factor that increases the corrosion in the PuCS nitric acid waste. B has a maximum soluble concentration that is a function of the nitric acid concentration, even

though the B load may get as high as 18 g/L. As shown in Table 7, the soluble B concentration in 8.4 M nitric acid is 3.20 g/L (0.296 M) at 25 °C. At the evaporator operating temperature (>100 °C), this concentration will likely be slightly greater (4-5 g/L). Also, since the initial B:F^- ratio in PuCS solution is 7.7:1, the B:F^- ratio is expected to remain above 1.0 even if some B precipitation occurs.

The available corrosion data for nitric acid solutions containing F^- and B have maximum B concentrations of approximately 2 g/L. The impact of B on free F^- concentration was most significant at B levels of 0.54 g/L as shown by the data for the Rocky Flats SS&C [6]. As the B concentration increased from 0.54 to 2.49 g/L, the free F^- concentration changed from 35 ppm to 12 ppm when the total F^- concentration was 431 ppm. The nitric acid concentration was 9.3 M. Extending this relationship to a B concentration of 4-5 g/L would yield a free F^- concentration of 8 ppm. The free acid concentration also affects the complexation of F^- in the presence of Al [6]. In lower acid solutions, Al complexed more F^-. For the evaporators, which will have a maximum acid concentration of 8 M, the free F^- concentration may drop as evaporation proceeds.

Evaporator Corrosion Rates

The corrosion rate for the starting condition in the LAW evaporators would be essentially that for 1 M nitric acid since with 0.056 M B most of the F^- would probably be complexed. The corrosion rate would be between 0-5 mpy [10]. For the 15x increase in concentration, the maximum corrosion rate will be less than 25 mpy based on experimental data shown above. For an 8 M nitric acid solution with 0.2 M F^- and 2 g/L B, corrosion rates of approximately 25 mpy were measured. Since the 15x concentration has a lower acidity, lower F^-, and higher B, the free F^- concentration and subsequently the corrosion rate should be lower. For the 30x evaporator concentration, the corrosion rate would be closer to 25 mpy since the F^- is similar to test data. This rate would be expected to be a maximum because the actual B concentration would be higher than the 2 g/L used in corrosion tests. Welded areas would be expected to have slightly higher corrosion rates (2-5 mpy) based on available data [8,11].

Thus, it is reasonable to use solutions 2 and 3 in Table 8 to gauge corrosion levels in the LAW evaporators for the proposed PuCS campaign. In the feed adjustment evaporator, solutions are typically concentrated about 20-40%, and the F^- in the feed is expected to increase gradually to a maximum of about 0.02 M F^- at the end of the PuCS campaign. Thus, the corrosion expected from F^- in the feed adjustment evaporator would be significantly less than that of the LAW evaporators.

Conclusions

The evaluation of the proposed flowsheet change leads to the following conclusions:

1. Modeling calculations for both A and D Banks show that a B DF of ~60,000 should be expected. Therefore, B contamination of the product solution is not a concern.

2. Boron can be credited as a complexant of F^- in nitric acid solutions, since B is similar to aluminum in reducing both free F^- concentration and corrosion of stainless steel.

3. Boron has limited solubility in concentrated solutions of nitric acid.

4. The proposed routing change for Plutonium-Contaminated Scrap solutions will introduce F^- into vessels that do not typically contain F^-. Though B complexes F^-, F^- solutions in evaporators will cause some corrosion, up to about 25 mils per year.

Recommendation

The conclusions of this study lead to the following recommendation:

1. Whenever B is used as a complexant for F^-, the limited solubility of B in concentrated solutions of nitric acid should be recognized. Especially during evaporation operations, adequate controls must be put in place to prevent precipitation of boron-containing solids.

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