SRNL-STI-2012-00400
Revision 0

Keywords: *Tank 26F, 2F Evaporator, Precipitation, Solids Formation*

Retention: *Permanent*

Tank 26F-2F Evaporator Study

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August 2012

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Prepared for the U.S. Department of Energy under contract number DE-AC09-08SR22470.

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EXECUTIVE SUMMARY

Tank 26F supernate sample was sent by Savannah River Remediation to Savannah River National Laboratory for evaporation test to help understand the underlying cause of the recent gravity drain line (GDL) pluggage during operation of the 2F Evaporator system.

The supernate sample was characterized prior to the evaporation test. The evaporation test involved boiling the supernate in an open beaker until the density of the concentrate (evaporation product) was between 1.4 to 1.5 g/mL. It was followed by filtering and washing of the precipitated solids with deionized water. The concentrate supernate (or concentrate filtrate), the damp unwashed precipitated solids, and the wash filtrates were characterized.

All the precipitated solids dissolved during water washing. A semi-quantitative X-ray diffraction (XRD) analysis on the unwashed precipitated solids revealed the solids are composed of the following.

Trona - $Na_3H(CO_3)_2 \cdot 2H_2O$ = 42 wt%
Thermonatrite - $Na_2CO_3 \cdot H_2O$ = 32 wt%
Nitratine - $NaNO_3$ = 23 wt%
Silicon Oxide - SiO_2 = 3 wt%
Natrite - Na_2CO_3 = Trace

All the above compounds with the exception of silica (silicon oxide) are known to be readily soluble in water. Hence, their dissolution during water washing is not unexpected. Even though silica is a sparingly water-soluble compound, its dissolution is also not surprising. This stems from its small fraction in the solids as a whole and also its relative freshness. Assuming similar supernate characteristics, flushing the GDL with water (preferably warm) should facilitate dissolution and removal of future pluggage events as long as build up/aging of the sparingly soluble constituent (silica) is limited. On the other hand, since the amount of silica formed is relatively small, it is quite possible dissolution of the more soluble larger fraction will cause disintegration or fragmentation of the sparingly soluble smaller fraction (that may be embedded in the larger soluble solid mass) and allow its removal via suspension in the flushing water.

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LIST OF ABBREVIATIONS

ARS	As-Received Supernate (Pre-Evaporation Sample)
CPS	Concentrate Precipitated Solids
CS	Concentrate Supernate
CSS	Concentrate Soluble Solids
CTS	Concentrate Total Solids
DI	deionized (water)
E&CPT	Environmental & Chemical Process Technology
FDS	Filtrate Dissolved Solids
GDL	Gravity Drain Line
IC	Ion Chromatography
ICP-ES	Inductively Coupled Plasma-Emission Spectroscopy
n/a	not applicable
nm	not measured
PUREX	Plutonium-Uranium Extraction
RSD	Relative Standard Deviation
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
SRS	Savannah River Site
TC	Total Carbon
TIC	Total Inorganic Carbon
TOC	Total Organic Carbon
TTQAP	Task Technical and Quality Assurance Plan
TTR	Technical Task Request
XRD	X-Ray Diffraction
1WF	1st Wash Filtrate
2T4WF	2nd to 4th Wash Filtrates
2T4WF	2nd to 4th Wash Filtrates

1.0 Introduction

The 2F Evaporator system has been experiencing pluggage in the gravity drain line (GDL) that drops evaporator concentrate into Tank 25F. It is envisaged that the pluggage issue may be due to the formation of solids in the evaporator which deposit in the GDL due to its limited slope and known low points. As a result, two samples were collected from Tank 26F (which is the feed tank for the 2F Evaporator) by Savannah River Remediation (SRR) and sent to Savannah River National Laboratory (SRNL) to perform work based on the Technical Task Request (TTR)[1] and a subsequent Task Technical and Quality Assurance Plan (TTQAP)[2] developed for the work.

However, upon opening the samples bottles at SRNL after a three-month interim storage period in the SRNL shielded cells during the winter months, the samples were found to contain solids.[3] A second set of Tank 26F sample was obtained and sent to SRNL to perform the work outlined in the TTR and TTQAP mentioned above. The objective of the work is to perform a simple batch evaporation test to characterize the solids, the potential evaporator feeds that contribute to the formation of solids and to identify the preferred medium to help dissolve and remove plugged solids in the evaporator GDL.

2.0 Experimental

2.1 Characterization of As-Received Tank 26F Supernate Sample

The Tank 26F sample (ID: FTF-26-12-3) was received in a 200-mL stainless steel liquid dip vial. It was transferred to a 250-mL polypropylene bottle and weighed. It was inspected visibly for the presence of solids. As depicted below in Figure 2-1, the as-received supernate did not have any solids. The mass of the as-received supernate was 265 grams.

Duplicate aliquots of the as-received supernate were submitted for the following analyses: Inductively Coupled Plasma-Emission Spectroscopy (ICP-ES), Ion Chromatography (IC) for Anions, Hydroxide (OH), Total Inorganic Carbon/Total Organic Carbon (TIC/TOC), and Dissolved Solids via Total Solids. Note that Rheology measurements (i.e, yield stress and viscosity) were not done. The rheology measurements require a large sample volume (~80 mL). Hence, it was skipped to allow more sample to be available for the evaporation test and the post-evaporation processing/characterization with agreement from the customer (SRR).

To enable the supernate samples to be removed from the shielded cell, the aliquots for the ICP-ES analysis were diluted with 3M nitric acid (1 mL supernate to 9 mL nitric acid). Similarly, the aliquots for the IC Anions, OH, and TIC/TOC analyses were diluted with deionized (DI) water (1 mL supernate to 9 mL DI water).

The density was also measured in triplicate using 2-mL Kimax Class A # 28014 (Fisher Scientific, Pittsburgh, Pennsylvania) density measuring tubes. As a standard check, the density of DI water was measured before and after the supernate density measurements. All the density measurements were performed at cell ambient temperature.

2.2 Evaporation Test

The evaporation test involved filling a 400-mL glass beaker to the 175-mL mark with the as-received Tank 26F supernate followed by weighing and heating to boiling while stirring simultaneously with a stir bar. The greater than 2X beaker capacity to supernate volume ratio prevented splashing of supernate out of the beaker during boiling.

The heating dial of the stirring hot plate was turned off when the liquid reached the 150-mL mark to allow the liquid to cool to the cell ambient temperature. Note that stirring was continued while the liquid was cooling. The duration of the evaporation process was about 35 minutes.

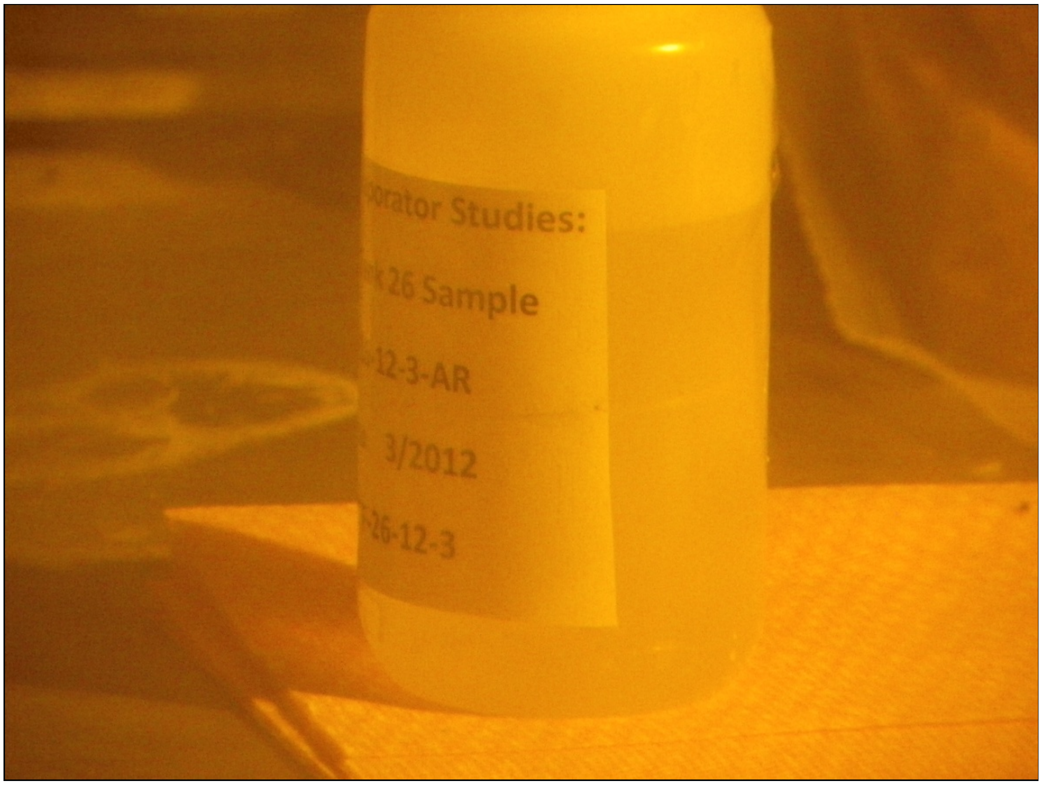

Figure 2-1. As-Received Tank 26F Supernate Sample.

The density of the cooled concentrate or evaporation product was measured as described earlier in subsection 2.1. Since the density was within the target range of 1.4 and 1.5 g/mL, the density measurement was repeated two additional times to confirm. Because the goal was to attain a concentrate with a density of between 1.4 and 1.5 g/mL (typical evaporator operating density range), the evaporation would have been repeated had the density of the concentrate been less than 1.4 g/mL.

The concentrate plus the stir bar were transferred to a polypropylene bottle and weighed. It was then allowed to sit for a week at the cell ambient temperature (~26 ^{o}C) to allow a more complete solids precipitation.

2.3 Post-Evaporation Processing and Characterization

A portion of the concentrate was filtered under vacuum using a 0.45 μm nylon Nalgene filter unit (Rochester, New York). To ensure minimal amount of residual solution in the filter cake (i.e., solids), the filtration was continued until the solution dripping ceased. The damp unwashed filter cake (or solids) was then sampled for X-ray Diffraction (XRD) analysis. The supernate (or filtrate) was set aside for later use.

Using another filtrate receiver bottle, the remaining damp unwashed filter cake was washed with 10 mL of DI water as follows. The DI water was poured into the top of the filter unit (filter cup) and allowed to soak for 5 minutes before turning on the vacuum to start the filtration. Again, the filtration was continued until the solution dripping ceased.

The vacuum was turned off and the filter unit was disconnected from the vacuum source. The filter unit top was unscrewed from the filtrate receiver bottle and the filtrate, referred to as 1st wash filtrate, was transferred to a 20-mL polypropylene vial. The same filtrate receiver bottle was reconnected to the filter unit top (containing the damp solids) and to the vacuum source. The 10-mL DI water washing was repeated three additional times. Note that the 2nd to 4th wash filtrates were collected as one composite solution. In other words, the same filtrate receiver bottle was used for the 2nd, 3rd, and 4th washings.

All the solids dissolved during the washing operation. To make sure or confirm no insoluble solids remained during washing, the filtration/washing was repeated with almost all the remaining or leftover concentrate. Hence, there was not enough concentrate for the planned rheology measurements (i.e, yield stress and viscosity). Again, all the solids dissolved during the repeat washing.

All the corresponding solids and solution from the repeat filtration/washing were combined with the respective solids and solutions from the initial filtration/washing i.e.,

Initial concentrate supernate and repeat concentrate supernate.
Initial damp unwashed filter cake (or solids) sampled for XRD analysis and
repeat damp unwashed filter cake (or solids) sampled for XRD analysis.
Initial 1st wash filtrate and repeat 1st wash filtrate.
Initial composite 2nd to 4th wash filtrates and repeat composite 2nd to 4th wash filtrates.

The solids dissolution was complete by the end of the 2nd washing. Because there were no insoluble solids or no solids remained after washing, the planned analyses below were not done: Acid digestion followed by ICPES, IC Anions, and TIC/TOC; XRD; and Moisture Content.

<u>Concentrate Supernate</u>
Duplicate aliquots of the concentrate supernate set aside earlier were submitted for the following analyses: ICP-ES, IC Anions, OH, TIC/TOC, and Dissolved Solids via Total Solids.

To enable the concentrate supernate samples to be removed from the shielded cell, the aliquots for the ICP-ES analysis were diluted with 3M nitric acid (1 mL supernate to 9 mL nitric acid). Similarly, the aliquots for the IC Anions, OH, and TIC/TOC analyses were diluted with deionized (DI) water (1 mL concentrate supernate to 9 mL DI water). The density was also measured in triplicate as described in subsection 2.1.

<u>1st Wash Filtrate</u>
Duplicate aliquots of the 1st wash filtrate were submitted for the following analyses after diluting with DI water (2 mL concentrate supernate to 248 mL DI water) to allow removal from the shielded cell: ICP-ES, IC Anions, and TIC/TOC. The density was also measured in triplicate as described in subsection 2.1.

<u>2nd to 4th Wash Filtrates</u>
Duplicate aliquots of the composite 2nd to 4th wash filtrates were submitted for the following analyses: ICP-ES, IC Anions, and TIC/TOC.

Concentrate
Duplicate aliquots of the remaining concentrate were submitted for Total Solids analysis.

3.0 Results and Discussion

3.1 Characterization of As-Received Tank 26F Supernate Sample

Table 3-1 gives the characterization data for the as-received Tank 26F supernate sample. As mentioned earlier, visual inspection indicated the supernate did not contain any solids. The major components on a molar basis include sodium, hydroxide, nitrate, nitrite, aluminum (Al), carbonate (or TIC), TOC, sulfate (or S), and potassium in that order. Note that oxalate was below the detection limit. The data compares reasonably well with those for fairly similar samples analyzed at F/H Area laboratories provided in Table 3-2.[4] With the exception of Al and phosphate, the remaining analytes are in good agreement. The data in Table 3-1 also agree with data for another fairly similar supernate samples collected on December 2, 2010 and analyzed at F/H Area laboratories given in Table 3-5 of Reference # 3.

3.2 Post-Evaporation Processing and Characterization

Figure 3-1 is a picture of the concentrate (evaporation product) taken two days after the evaporation test ended. The picture shows precipitated solids at the bottom of the bottle.

Table 3-3 provides the composition of the solids (i.e., total, soluble, and precipitated) in the concentrate (or slurry) along with similar solids data for the concentrate filtrate and the as-received supernate. As expected the pre-evaporation solution total solids content (i.e., as-received supernate soluble solids) increased after evaporation.

Amount of total solids in as-received (pre-evaporation) supernate = 92.18 mg
Amount of total solids in concentrate = 88.16 mg
Ratio of concentrate total solids to as-received supernate total solids = 0.96

The above indicate good overall total solids mass balance.

Figure 3-2 provides the XRD patterns for the unwashed precipitated solids. A semi-quantitative analysis gives the composition of the solids as follows.

Trona - $Na_3H(CO_3)_2 \cdot 2H_2O$ = 42 wt%
Thermonatrite - $Na_2CO_3 \cdot H_2O$ = 32 wt%
Nitratine - $NaNO_3$ = 23 wt%
Silicon Oxide - SiO_2 = 3 wt%
Natrite - Na_2CO_3 = Trace

The composition indicates a large portion (74 wt%) of the unwashed precipitated solids is sodium carbonate/bicarbonate compounds. Note that the interstices or voids in the unwashed damp precipitated solids contained residual supernate. It is quite possible the dried residual supernate (especially $NaNO_3$) may have contributed to the solids on the filter membrane.

As a comparison, the unwashed solids from the earlier work[3] contained the following compounds: Natroxalate [$Na_2C_2O_4$], Clarkeite [$Na((UO_2)O)(OH) \cdot (H_2O)$], and Nitratine [$NaNO_3$]; while the water-washed solids consisted of following: Hematite [Fe_2O_3], Clarkeite [$Na((UO_2)O(OH))$], Natroxalate [$Na_2C_2O_4$], Iron Oxalate Hydrate [$C_2FeO_4 \cdot 2H_2O$], and Sodium Alumino-Silicate Nitrate [$Na_8(Al_6Si_6O_{24})(NO_3)_2 \cdot 4H_2O$].

Table 3-1. Composition of As-Received Tank 26F Supernate Sample (ID: FTF-26-12-3).

	Replicate 1	Replicate 2	Average	Average	RSD
Analyte	**mg/L**	**mg/L**	**mg/L**	**M**	**%**
Ag	<1.43E+01	<1.43E+01	<1.43E+01	<1.33E-04	n/a
Al	**1.16E+04**	**1.19E+04**	**1.17E+04**	**4.34E-01**	1.7
B	**9.80E+01**	**1.03E+02**	**1.00E+02**	**9.30E-03**	3.5
Ba	<6.18E+00	<6.18E+00	<6.18E+00	<4.50E-05	n/a
Be	<7.85E-01	<7.84E-01	<7.85E-01	<8.71E-05	n/a
Ca	**5.59E+00**	**5.59E+00**	**5.59E+00**	**1.40E-04**	0.05
Cd	<6.28E+00	<6.28E+00	<6.28E+00	<5.59E-05	n/a
Ce	<5.92E+01	<5.91E+01	<5.91E+01	<4.22E-04	n/a
Co	<9.52E+00	<9.51E+00	<9.51E+00	<1.61E-04	n/a
Cr	**1.84E+02**	**1.90E+02**	**1.87E+02**	**3.60E-03**	2.2
Cu	<6.18E+00	<6.18E+00	<6.18E+00	<9.73E-05	n/a
Fe	**1.53E+01**	**1.57E+01**	**1.55E+01**	**2.78E-04**	1.7
Gd	<7.65E+00	<7.65E+00	<7.65E+00	<4.87E-05	n/a
K	**1.67E+03**	**1.73E+03**	**1.70E+03**	**4.34E-02**	2.4
La	<5.30E+00	<5.30E+00	<5.30E+00	<3.81E-05	n/a
Li	<1.43E+01	<1.43E+01	<1.43E+01	<2.06E-03	n/a
Mg	**2.16E+00**	**3.38E+00**	**2.77E+00**	**1.14E-04**	31
Mn	<2.06E+00	<2.06E+00	<2.06E+00	<3.75E-05	n/a
Mo	**5.35E+01**	**5.68E+01**	**5.51E+01**	**5.75E-04**	4.2
Na	**1.93E+05**	**1.99E+05**	**1.96E+05**	**8.53E+00**	2.1
Ni	<1.57E+01	<1.57E+01	<1.57E+01	<2.67E-04	n/a
P	**3.33E+02**	**3.65E+02**	**3.49E+02**	**1.13E-02**	6.52
Pb	<7.03E+01	<7.02E+01	<7.02E+01	<3.39E-04	n/a
S	**1.10E+03**	**2.12E+03**	**1.61E+03**	**5.02E-02**	45
Sb	<1.05E+02	<1.05E+02	<1.05E+02	<8.62E-04	n/a
Si	<4.31E+01	<4.30E+01	<4.31E+01	<1.53E-03	n/a
Sn	<5.50E+01	<5.50E+01	<5.50E+01	<4.64E-04	n/a
Sr	<4.91E-01	<4.90E-01	<4.90E-01	<5.60E-06	n/a
Th	<2.63E+01	<2.63E+01	<2.63E+01	<1.13E-04	n/a
Ti	<1.28E+00	<1.27E+00	<1.28E+00	<2.66E-05	n/a
U	<3.21E+02	<3.21E+02	<3.21E+02	<1.35E-03	n/a
V	<4.61E+00	<4.61E+00	<4.61E+00	<9.05E-05	n/a
Zn	**8.93E+00**	**1.17E+01**	**1.03E+01**	**1.58E-04**	19
Zr	<2.45E+00	<2.45E+00	<2.45E+00	<2.69E-05	n/a
Fluoride	<1.06E+02	<1.03E+02	<1.04E+02	<5.50E-03	n/a
Formate	<1.06E+02	<1.03E+02	<1.04E+02	<2.32E-03	n/a
Chloride	<5.31E+02	<5.13E+02	<5.22E+02	<1.47E-02	n/a
Nitrite	**4.80E+04**	**4.89E+04**	**4.85E+04**	**1.05E+00**	1.3
Bromide	<1.06E+03	<1.03E+03	<1.04E+03	<1.31E-02	n/a
Nitrate	**1.30E+05**	**1.31E+05**	**1.30E+05**	**2.10E+00**	0.88
Phosphate	**6.59E+02**	**6.57E+02**	**6.58E+02**	**6.93E-03**	0.27
Sulfate	**4.91E+03**	**4.75E+03**	**4.83E+03**	**5.03E-02**	2.4
Oxalate	<1.06E+02	<1.03E+02	<1.04E+02	<1.19E-03	n/a
TIC	**2.80E+03**	**2.75E+03**	**2.77E+03**	**2.31E-01**	1.2

	Replicate 1	**Replicate 2**	**Average**	**Average**	**RSD**
Analyte	**mg/L**	**mg/L**	**mg/L**	**M**	**%**
TOC	**2.49E+03**	**2.38E+03**	**2.43E+03**	**2.03E-01**	3.1
TC	**5.28E+03**	**5.13E+03**	**5.21E+03**	**4.33E-01**	2.1
Carbonate*	**1.40E+04**	**1.37E+04**	**1.39E+04**	**2.31E-01**	1.2
	M	**M**	**M**		**%**
Hydroxide	**3.28E+00**	**3.11E+00**	**3.20E+00**		3.9
	Replicate 1	**Replicate 2**	**Replicate 3**	**Average**	**RSD**
	g/mL	**g/mL**	**g/mL**	**g/mL**	**%**
Density	1.34	1.32	1.35	1.34	1.1

RSD = Relative Standard Deviation
TIC = Total Inorganic Carbon
TOC = Total Organic Carbon
TC = Total Carbon
* Calculated from corresponding TIC values
n/a = Not applicable
Values with "<" symbols are detection limits. Note: Non-detection limit values are in bold.

Table 3-2. Composition of Tank 26F Supernate Samples Analyzed at F/H Area Laboratories.

Sample ID	**200590539 (CC/EFQ)**	**200590540 (EFQ) 161"**
Collection Date	**2/9/2012**	**2/9/2012**
Analyte	**M**	**M**
Al	2.44E-01	2.12E-01
Na	7.35E+00	7.55E+00
Si	5.00E-04	5.00E-04
Nitrite	1.29E+00	No value was given
Nitrate	2.20E+00	No value was given
Phosphate	1.10E-02	1.05E-02
Sulfate	5.88E-02	5.57E-02
Oxalate	<5.70E-03	<5.20E-03
Carbonate	2.42E-01	2.16E-01
Hydroxide	3.40E+00	3.48E+00
	g/mL	**g/mL**
Density	1.35	1.37

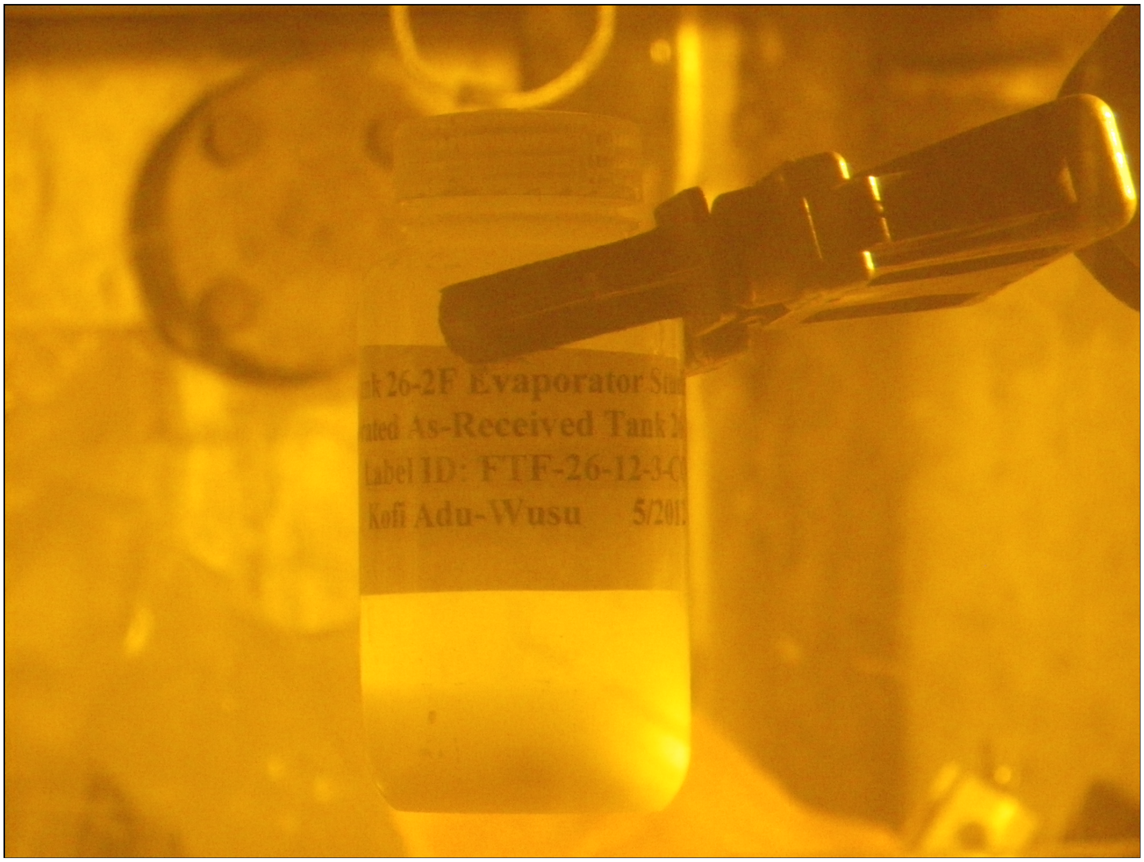

Figure 3-1. Concentrate (Evaporation Product) With Solids at the Bottom of the Bottle.

Table 3-3. Composition of Solids in Concentrate, Concentrate Supernate, and As-Received Supernate.

	Replicate 1	**Replicate 2**	**Average**	**RSD**
	Wt %	**Wt %**	**Wt %**	**%**
Concentrate Total Solids (CTS)	48.5	48.1	48.3	0.59
Concentrate Filtrate Dissolved Solids (FDS)	45.5	45.7	45.6	0.31
As-Received Supernate Soluble Solids	38.8	38.8	38.8	0.08
Concentrate Precipitated Solids (CPS)*	5.0 wt%			
Concentrate Soluble Solids (CSS)**	43.3 wt%			

* Calculated from CPS = (CTS – FDS)/(1 – FDS/100), using the respective average values.
** Calculated from CSS = CTS – CPS, using the CTS average value.

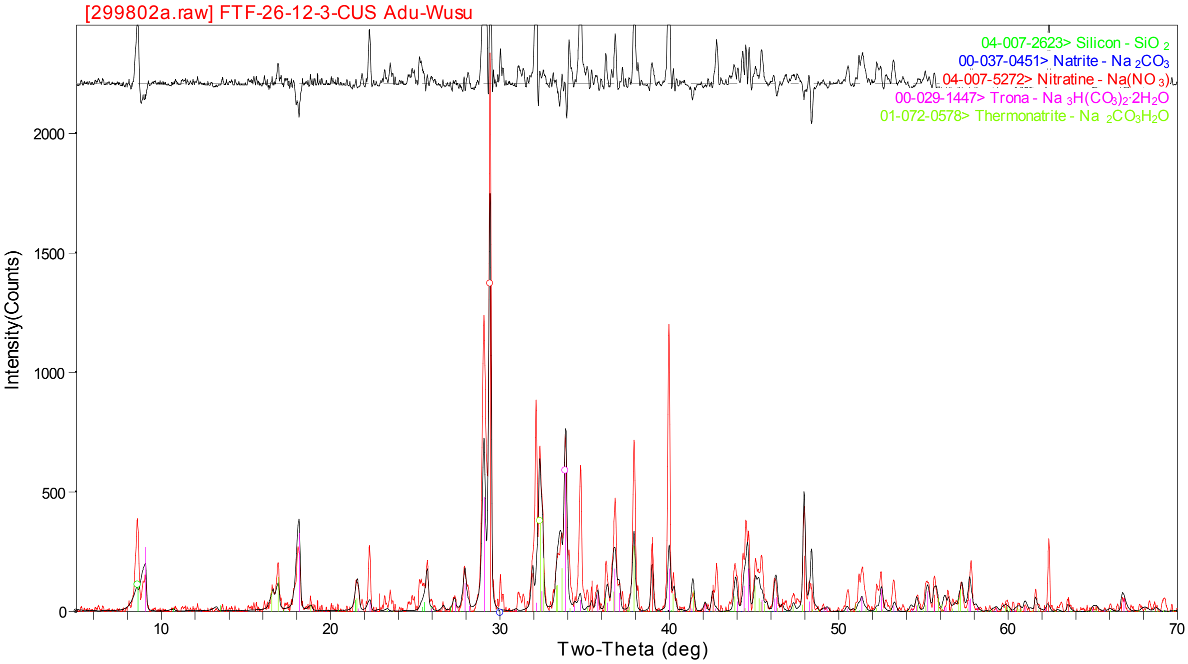

Figure 3-2. XRD Patterns for the Damp Unwashed Precipitated Solids.

Table 3-4 provides the analysis data for the concentrate supernate or concentrate filtrate. Expectedly, the analyte concentrations for the concentrate supernate are generally higher than the corresponding as-received supernate concentrations (see Table 3-1). Ca, S, and Mg are exceptions because one or both of the %RSDs values is relatively high. Carbonate and TIC values also deviate due to the fact that a greater fraction of the solids formed are carbonates/bicarbonates.

For TOC values, the deviation is hard to explain. The values for formate and oxalate do not help either because they are all below detection limits. In addition, the detection limits for formate and oxalate in the concentrate supernate are an order of magnitude higher than those for the as-received supernate. It is quite possible other organics not analyzed for decomposed via heating during evaporation. The other possibility is that small fractions (of the total solids formed) of several organic solid compounds not detectable by XRD formed. But it seems more unlikely. Note that the percent difference between the two average TOC values is 23% which is close to the error in the analysis of 20%. The TOC analysis data are questionable.

As mentioned in the experimental section, all the precipitated solids on the filter membrane dissolved during washing with DI water. In other words, no water-insoluble solids remained after washing. This observation is not unexpected based on the composition of the as-received sample. First, all the solid compounds identified by XRD with the exception silica (silicon oxide) have fairly high solubilities in water.

Secondly, sparingly soluble compounds like silica generally form first as amorphous phases before slowly transforming to crystalline phases. The transformation is typically slow. It is of the order of weeks to months and possibly years in some instances especially at room or low temperatures. Hence, it is quite possible silica and any other insoluble compounds that may have formed dissolved readily. This is because amorphous materials are in general more soluble than their crystalline counterparts; and relatively fresh crystals are also generally more soluble than aged crystals. Lastly, silica, at the relatively low levels in the precipitated solids will most likely dissolve.

The foregoing seems to imply the following for 2F Evaporator operations. Formation of solids occurs in the 2F Evaporator system (i.e., GDL and drop tank) and the solids that form are water soluble. However, there are differences between the laboratory evaporation test conducted here and the 2F Evaporator system.

- The 2F Evaporator operation is continuous versus the batch laboratory evaporation test. This means solids that form in the GDL increase or build up with time to a level that may be above the respective solubilities of the solid compounds. Further, aged crystals are generally less soluble than freshly precipitated crystals.

- The drop tank for the 2F Evaporator system is at a temperature of about 50 ^{o}C versus shielded cell ambient temperature of 26 ^{o}C for the laboratory evaporation test. This implies the temperature of the GDL will be at least 50 ^{o}C. The degree of crystallinity of solid compounds generally increases with increasing temperature; and the more crystalline a material is, the less soluble it will be.

In the light of the above differences between the 2F Evaporator operation and the laboratory evaporation test, it could be inferred that the solids that form in the 2F Evaporator system may be less soluble in water than their laboratory evaporation test counterparts especially the sparingly soluble component, silica. Put differently, even though all the solids that formed in the laboratory evaporation test dissolved readily during washing, there is a small likelihood that it may not be the case for some of the solid constituents that form in the 2F Evaporator system.

Note that contrary to the above, the earlier work[3] identified a number of water-insoluble compounds (clarkeite, oxalates, hematite, aluminosilicates etc.) in the 2F Evaporator educator feed pump solids and the solids that formed in the Tank 26F sample. The reason for the different solid types (soluble/insoluble) formed is most likely from the change in feed composition. The samples for the earlier work were from 2F Evaporator feed that contained neutralized spent oxalic acid solutions resulting from plutonium-uranium extraction (PUREX) sludge dissolutions. The sample for this work did not contain any residual oxalic acid.

The data for the 1st wash filtrate are given in Table 3-5. Most of the data are below detection limit because of the large dilution factor (~125) used. The analytes that are above detection limits by and large reflect the solid compounds identified by XRD – Na, nitrate, and carbonate with Al, S, and TOC as exceptions. The Al, S, and TOC are mainly from the concentrate supernate in the voids of the solids. Note that the Al, S, and TOC concentrations in the concentrate supernate are relatively high (see Table 3-4).

Table 3-6 gives the analysis data for the composite 2nd to 4th wash filtrates. More analytes are above their detection limits than the data for 1st wash filtrate (Table 3-5) because they were not diluted. Since dissolution was complete by the end of the 2nd water wash they were considered dilute enough to allow removal from the shielded cell. Because the 3rd and 4th water washes were essentially dilution of the solution at the end of the 2nd wash, the concentrations are generally lower than their respective counterparts for the 1st wash filtrate.

A rough mass balance for all analytes where values are available (i.e., are not less than detection limits) are provided in Table 3-7. The basis of the mass balance is as follows. The meaning of the abbreviations is given in the first two rows of the Table and also at the bottom of the Table.

ARS = CS + Precipitated Solids
Precipitated Solids + Residual CS in the voids of the solids = 1WF + 2T4WF
ARS = CS + 1WF + 2T4WF

The ratios in the last-but-one column (also referred to as mass balance column) indicate good overall mass balance. Ideally, all the ratios should be 1. In other words, the closer a ratio value is to 1, the better the mass balance. Note that a portion of the Precipitated Solids + Residual CS in the interstices and voids in the solids (i.e., damp unwashed solids) were taken for XRD analysis. This implies the values for 1WF and 2T4WF (and therefore the last-but-one column) in Table 3-7 especially the former will be lower than expected.

Also, TIC and carbonate values should be much lower than expected because of the high concentration of carbonate/bicarbonate measured in the solids by XRD. Similarly, the nitrate value should be much lower than expected because it comprises at least a fifth of the solids formed. The Na ratio value is not as low as expected when compared with the values for TIC, carbonate, and nitrate even though it is part of the solids formed. This is due to its extremely high concentration in both the ARS and the CS.

Based on the above, it could be argued that other analytes (e.g., Al, Fe, Zn, etc.) may also have formed solids but either their fractions in the total solids were too small and/or amorphous for XRD to detect. An additional requirement for solids formation will be used to elucidate this later. The ratios should further be viewed with the perspective that the error in the analysis of the concentration values (from Table 3-1, and Table 3-4 to Table 3-6) used to calculate the amounts of the analytes (in Table 3-7) is 20%.

The mass balance for B is a little on the high side. Calcium's and magnesium's mass balance are off. The main reason for this may be that for Ca, the %RSD for its CS analysis is relatively high (see Table 3-4); and for Mg, the %RSDs for its ARS and CS analyses are relatively high (see Table 3-1 and Table 3-4). Also, for Ca, the 1WF analysis was below detection limit; and for Mg, the 1WF and 2T4WF analyses were below detection limits. As a result, no amounts for 1WF and 2T4WF were calculated as indicated by the blank cells. Lastly, the amounts of Ca and Mg in the ARS were relatively small to begin with.

The ratio for sulfate is low because of the less than detection limit values for CS and 1WF. However, the value for S is quite good. The TOC ratio has a significant deviation. As mentioned earlier, the TOC analyses data look suspicious.

Comparing the amount of analyte in ARS and CS as indicated by the ratio values in the last column of Table 3-7 gives a hint as to which analytes may have formed solids. However, because the mass balance ratios in the last-but-one column (i.e., mass balance column) deviate from 1 due to errors in the analysis as mentioned earlier, the additional requirement for a hint in solids formation is for the ratio in the last column to be less than the corresponding ratio in the last-but-one column.

In other words, an analyte with equal (e.g., B, Fe, Mo, P, Zn, etc.) or approximately equal (e.g., Al, Cr, K, nitrite, etc.**)** ratio values in the last-but-one column and the last column virtually did not form any solids. The values or data for Na, nitrate, TIC, and carbonate (which formed solids from XRD analysis) confirm this especially when it is considered that their values in the last-but-one column should have been higher due to the fact that a portion of the solids was taken for XRD analysis.

S deviates from the above requirement because it did not form measureable solids based on the XRD analysis. It may be due to the %RSD of its ARS analysis being relatively high (see Table 3-1). Ca sort of deviates from the above requirements because it did not form solids based on the XRD analysis. It may also be due to the %RSD of its CS analysis being relatively high (see Table 3-4). Si formed solids (relatively small). However, because its ARS analysis was less than the detection limit, no ratios could be calculated, let alone check to see if it conforms to the requirement.

The ratio values for TOC seem to suggest formation of organic solids. The XRD patterns did not indicate the formation of any organic solids. A comparison of the TIC or carbonate amounts to the TOC amounts (especially the 1WF and 2T4WF) indicates if organic solids had formed, they would have been relatively substantial; which implies the last-but-one column ratio should have been much less than 1 like the TIC ratio value. As it stands, the TOC ratio value indicates more than the TOC needed for mass balance.

On the other hand, since for XRD did not detect any organic solids, it could be argued that small fractions (of the total solids formed) of several organic solid compounds (though substantial in total in terms of TOC) not detectable by XRD were formed. It looks more likely that the IWF and 2T4WF TOC analysis data are also questionable (too high) or somehow the samples may have been contaminated.

The Si concentration in ARS was less than detection limit due most likely to its dilution prior to submission for analysis (see Table 3-1). This in no way should be construed to mean there is no Si in the Tank 26F supernate. Table 3-2 gives composition for fairly similar supernate samples analyzed at F/H Area laboratories. They contain Si. Also, information on Tank 26F supernate analysis that goes back to 2009 all indicates the presence of Si.[4]

As shown in Table 3-7, the maximum amount of Si that can be in ARS and 1WF is 7.63 and 3.14 mg respectively based on the detection limit values given in Table 3-1 and Table 3-5. The amount of Si in CS (26.6 mg) is higher than the maximum amount that can be in the ARS (i.e., the pre-evaporation solution). This suggests Si most likely dissolved from the glass beaker used for the evaporation test. Hence, the silica solids that formed can be attributed to two sources: Si already in the Tank 26F supernate and Si from glass beaker dissolution. Previous characterization by King et al.[3] measured the presence of Si compounds in the feed, and demonstrated the formation of Si compounds.

Two types of samples were received for the earlier work. The first sample was supposed to be a Tank 26F supernate (Sample ID: FTF-26-10-57 and FTF-26-10-58). The second sample was a solid material from the 2F Evaporator educator feed pump. Both samples were stored in the SRNL shielded cells for about 3 months before the containers were opened. The storage temperature ranged from 12-22 °C. The first sample turned out to be a slurry instead of a supernate from the precipitation of solids during storage.

All the earlier work was done in plastic containers. Figure 3-3 of the earlier work's report[3] listed Sodium Aluminosilicate Nitrate as one of the compounds identified by XRD in the water-washed solids of the first sample. Figure 3-4 of the same report mentioned Illite, a crystalline Si containing compound, as one of the compounds identified by XRD in the water-washed solids of the second sample.

Table 3-4. Composition of Concentrate Supernate.

Analyte	**Replicate 1 mg/L**	**Replicate 2 mg/L**	**Average mg/L**	**Average M**	**RSD %**
Ag	<2.38E+01	<2.27E+01	<2.32E+01	<2.15E-04	n/a
Al	**1.58E+04**	**1.55E+04**	**1.56E+04**	**5.79E-01**	1.5
B	**2.09E+02**	**2.12E+02**	**2.10E+02**	**1.95E-02**	0.93
Ba	<2.16E+00	<2.06E+00	<2.11E+00	<1.53E-05	n/a
Be	<5.34E-01	<5.08E-01	<5.21E-01	<5.78E-05	n/a
Ca	**3.81E+00**	**2.99E+00**	**3.40E+00**	**8.49E-05**	17
Cd	<2.96E+00	<2.82E+00	<2.89E+00	<2.57E-05	n/a
Ce	<1.76E+01	<1.67E+01	<1.72E+01	<1.22E-04	n/a
Co	<4.76E+00	<4.53E+00	<4.65E+00	<7.89E-05	n/a
Cr	**2.57E+02**	**2.64E+02**	**2.60E+02**	**5.01E-03**	1.9
Cu	<2.89E+00	<2.75E+00	<2.82E+00	<4.44E-05	n/a
Fe	**1.81E+01**	**1.83E+01**	**1.82E+01**	**3.26E-04**	0.77
Gd	<4.27E+00	<4.07E+00	<4.17E+00	<2.65E-05	n/a
K	**2.56E+03**	**2.64E+03**	**2.60E+03**	**6.64E-02**	2.2
La	<4.25E+00	<4.05E+00	<4.15E+00	<2.99E-05	n/a
Li	<3.38E+00	<3.22E+00	<3.30E+00	<4.76E-04	n/a
Mg	**4.23E-01**	**3.60E-01**	**3.91E-01**	**1.61E-05**	11
Mn	<7.56E-01	<7.20E-01	<7.38E-01	<1.34E-05	n/a
Mo	**9.47E+01**	**9.87E+01**	**9.67E+01**	**1.01E-03**	3.0
Na	**2.56E+05**	**2.53E+05**	**2.55E+05**	**1.11E+01**	0.73
Ni	<2.20E+01	<2.10E+01	<2.15E+01	<3.66E-04	n/a
P	**4.95E+02**	**5.39E+02**	**5.17E+02**	**1.67E-02**	6.1
Pb	<1.30E+02	<1.24E+02	<1.27E+02	<6.13E-04	n/a
S	**1.19E+03**	**1.25E+03**	**1.22E+03**	**3.81E-02**	3.5
Sb	<8.05E+01	<7.67E+01	<7.86E+01	<6.46E-04	n/a
Si	**2.29E+02**	**2.34E+02**	**2.32E+02**	**8.25E-03**	1.5
Sn	<1.02E+02	<9.76E+01	<1.00E+02	<8.43E-04	n/a
Sr	<3.34E-01	<3.18E-01	<3.26E-01	<3.72E-06	n/a
Th	<2.16E+01	<2.06E+01	<2.11E+01	<9.08E-05	n/a
Ti	<8.90E-01	<8.47E-01	<8.69E-01	<1.81E-05	n/a
U	<1.30E+02	<1.24E+02	<1.27E+02	<5.34E-04	n/a
V	<1.16E+00	<1.10E+00	<1.13E+00	<2.22E-05	n/a
Zn	**1.40E+01**	**1.35E+01**	**1.37E+01**	**2.10E-04**	2.9
Zr	<1.25E+00	<1.19E+00	<1.22E+00	<1.33E-05	n/a
Fluoride	<1.08E+03	<1.07E+03	<1.07E+03	<5.65E-02	n/a
Formate	<1.08E+03	<1.07E+03	<1.07E+03	<2.38E-02	n/a
Chloride	<1.08E+03	<1.07E+03	<1.07E+03	<3.03E-02	n/a
Nitrite	**6.26E+04**	**6.97E+04**	**6.61E+04**	**1.44E+00**	7.5
Bromide	<5.40E+03	<5.33E+03	<5.36E+03	<6.71E-02	n/a
Nitrate	**1.46E+05**	**1.53E+05**	**1.50E+05**	**2.41E+00**	3.6
Phosphate	<1.08E+03	<1.07E+03	<1.07E+03	<1.13E-02	n/a
Sulfate	<1.08E+03	<1.07E+03	<1.07E+03	<1.12E-02	n/a
Oxalate	<2.16E+03	<2.13E+03	<2.15E+03	<2.44E-02	n/a
TIC	**2.01E+03**	**2.05E+03**	**2.03E+03**	**1.69E-01**	1.3

	Replicate 1	**Replicate 2**	**Average**	**Average**	**RSD**
Analyte	**mg/L**	**mg/L**	**mg/L**	**M**	**%**
TOC	**1.93E+03**	**1.92E+03**	**1.93E+03**	**1.60E-01**	0.59
TC	**3.94E+03**	**3.96E+03**	**3.95E+03**	**3.29E-01**	0.36
Carbonate*	**1.00E+04**	**1.02E+04**	**1.01E+04**	**1.69E-01**	1.2
	M	**M**	**M**		**%**
Hydroxide	**5.30E+00**	**5.13E+00**	**5.22E+00**		2.3
	Replicate 1	**Replicate 2**	**Replicate 3**	**Average**	**RSD**
	g/mL	**g/mL**	**g/mL**	**g/mL**	**%**
Density	1.43	1.42	1.45	1.43	0.88

RSD = Relative Standard Deviation
TIC = Total Inorganic Carbon
TOC = Total Organic Carbon
TC = Total Carbon
* Calculated from corresponding TIC values
n/a = Not applicable
Values with "<" symbols are detection limits. Note: Non-detection limit values are in bold.

Table 3-5. Composition of the First Wash Filtrate.

Analyte	**Replicate 1 mg/L**	**Replicate 2 mg/L**	**Average mg/L**	**Average M**	**RSD %**
Ag	<4.27E+01	<4.27E+01	<4.27E+01	<3.96E-04	n/a
Al	**2.00E+03**	**2.04E+03**	**2.02E+03**	**7.48E-02**	1.3
B	<2.77E+01	<2.77E+01	<2.77E+01	<2.56E-03	n/a
Ba	<2.38E+01	<2.38E+01	<2.38E+01	<1.73E-04	n/a
Be	<5.89E+00	<5.89E+00	<5.89E+00	<6.53E-04	n/a
Ca	<3.66E+01	<3.66E+01	<3.66E+01	<9.12E-04	n/a
Cd	<2.97E+01	<2.97E+01	<2.97E+01	<2.64E-04	n/a
Ce	<1.94E+02	<1.94E+02	<1.94E+02	<1.38E-03	n/a
Co	<4.49E+01	<4.49E+01	<4.49E+01	<7.62E-04	n/a
Cr	<5.30E+01	<5.30E+01	<5.30E+01	<1.02E-03	n/a
Cu	<1.59E+01	<1.59E+01	<1.59E+01	<2.51E-04	n/a
Fe	<4.83E+01	<4.83E+01	<4.83E+01	<8.65E-04	n/a
Gd	<4.71E+01	<4.71E+01	<4.71E+01	<3.00E-04	n/a
K	<1.88E+03	<1.88E+03	<1.88E+03	<4.80E-02	n/a
La	<4.69E+01	<4.69E+01	<4.69E+01	<3.37E-04	n/a
Li	<5.52E+02	<5.52E+02	<5.52E+02	<7.95E-02	n/a
Mg	<2.70E+00	<2.70E+00	<2.70E+00	<1.11E-04	n/a
Mn	<8.34E+00	<8.34E+00	<8.34E+00	<1.52E-04	n/a
Mo	<2.74E+02	<2.74E+02	<2.74E+02	<2.85E-03	n/a
Na	**6.54E+04**	**6.51E+04**	**6.53E+04**	**2.84E+00**	0.27
Ni	<2.43E+02	<2.43E+02	<2.43E+02	<4.14E-03	n/a
P	<1.34E+03	<1.34E+03	<1.34E+03	<4.32E-02	n/a
Pb	<1.44E+03	<1.44E+03	<1.44E+03	<6.93E-03	n/a
S	**3.74E+03**	**3.72E+03**	**3.73E+03**	**1.16E-01**	0.47
Sb	<8.88E+02	<8.88E+02	<8.88E+02	<7.29E-03	n/a
Si	<1.53E+02	1.61E+02	<1.57E+02	<5.59E-03	n/a
Sn	<3.61E+02	<3.61E+02	<3.61E+02	<3.04E-03	n/a
Sr	<3.68E+00	<3.68E+00	<3.68E+00	<4.20E-05	n/a
Th	<2.38E+02	<2.38E+02	<2.38E+02	<1.03E-03	n/a
Ti	<9.81E+00	<9.81E+00	<9.81E+00	<2.05E-04	n/a
U	<1.44E+03	<1.44E+03	<1.44E+03	<6.03E-03	n/a
V	<1.28E+01	<1.28E+01	<1.28E+01	<2.50E-04	n/a
Zn	<3.21E+01	<3.21E+01	<3.21E+01	<4.92E-04	n/a
Zr	<1.37E+01	<1.37E+01	<1.37E+01	<1.51E-04	n/a
Fluoride	<1.23E+04	<1.23E+04	<1.23E+04	<6.46E-01	n/a
Formate	<1.23E+04	<1.23E+04	<1.23E+04	<2.72E-01	n/a
Chloride	<1.23E+04	<1.23E+04	<1.23E+04	<3.46E-01	n/a
Nitrite	<1.23E+04	<1.23E+04	<1.23E+04	<2.67E-01	n/a
Bromide	<6.13E+04	<6.13E+04	<6.13E+04	<7.68E-01	n/a
Nitrate	**4.86E+04**	**4.85E+04**	**4.85E+04**	**7.82E-01**	0.18
Phosphate	<1.23E+04	<1.23E+04	<1.23E+04	<1.29E-01	n/a
Sulfate	<1.23E+04	<1.23E+04	<1.23E+04	<1.28E-01	n/a
Oxalate	<2.45E+04	<2.45E+04	<2.45E+04	<2.79E-01	n/a
TIC	**4.44E+03**	**4.44E+03**	**4.44E+03**	**3.70E-01**	0.00

	Replicate 1	**Replicate 2**	**Average**	**Average**	**RSD**
Analyte	**mg/L**	**mg/L**	**mg/L**	**M**	**%**
TOC	**1.95E+04**	**2.01E+04**	**1.98E+04**	**1.65E+00**	2.2
TC	**2.39E+04**	**2.45E+04**	**2.42E+04**	**2.02E+00**	1.8
Carbonate*	**2.22E+04**	**2.22E+04**	**2.22E+04**	**3.70E-01**	0.00
Hydroxide	nm	nm	nm		
	Replicate 1	**Replicate 2**	**Replicate 3**	**Average**	**RSD**
	g/mL	**g/mL**	**g/mL**	**g/mL**	**%**
Density	1.13	1.12	1.12	1.12	0.38

RSD = Relative Standard Deviation
TIC = Total Inorganic Carbon
TOC = Total Organic Carbon
TC = Total Carbon
* Calculated from corresponding TIC values
n/a = Not applicable
nm = Not measured
Values with “<” symbols are detection limits. Note: Non-detection limit values are in bold.

Table 3-6. Composition of the Composite 2nd to 4th Wash Filtrates.

	Replicate 1	**Replicate 2**	**Average**	**Average**	**RSD**
Analyte	**mg/L**	**mg/L**	**mg/L**	**M**	**%**
Ag	<3.48E-01	<3.48E-01	<3.48E-01	<3.23E-06	n/a
Al	**2.39E+02**	**2.48E+02**	**2.44E+02**	**9.02E-03**	2.6
B	**2.26E+00**	**2.32E+00**	**2.29E+00**	**2.12E-04**	1.9
Ba	<1.94E-01	<1.94E-01	<1.94E-01	<1.41E-06	n/a
Be	<4.80E-02	<4.80E-02	<4.80E-02	<5.33E-06	n/a
Ca	7.20E-01	6.89E-01	7.05E-01	1.76E-05	3.1
Cd	<2.42E-01	<2.42E-01	<2.42E-01	<2.15E-06	n/a
Ce	<1.58E+00	<1.58E+00	<1.58E+00	<1.13E-05	n/a
Co	<3.66E-01	<3.66E-01	<3.66E-01	<6.21E-06	n/a
Cr	**4.08E+00**	**4.12E+00**	**4.10E+00**	**7.89E-05**	0.69
Cu	<1.30E-01	<1.30E-01	<1.30E-01	<2.05E-06	n/a
Fe	<3.94E-01	<3.94E-01	<3.94E-01	<7.05E-06	n/a
Gd	<3.84E-01	<3.84E-01	<3.84E-01	<2.44E-06	n/a
K	**3.81E+01**	**3.74E+01**	**3.78E+01**	**9.65E-04**	1.3
La	<3.82E-01	<3.82E-01	<3.82E-01	<2.75E-06	n/a
Li	<4.50E+00	<4.50E+00	<4.50E+00	<6.49E-04	n/a
Mg	<2.20E-02	<2.20E-02	<2.20E-02	<9.05E-07	n/a
Mn	<6.80E-02	<6.80E-02	<6.80E-02	<1.24E-06	n/a
Mo	<2.23E+00	<2.23E+00	<2.23E+00	<2.32E-05	n/a
Na	**1.21E+04**	**1.24E+04**	**1.23E+04**	**5.33E-01**	1.7
Ni	<1.98E+00	<1.98E+00	<1.98E+00	<3.37E-05	n/a
P	<1.09E+01	<1.09E+01	<1.09E+01	<3.52E-04	n/a
Pb	<1.17E+01	<1.17E+01	<1.17E+01	<5.65E-05	n/a
S	**1.27E+03**	**1.29E+03**	**1.28E+03**	**3.99E-02**	1.1
Sb	<7.24E+00	<7.24E+00	<7.24E+00	<5.95E-05	n/a
Si	**3.71E+00**	**4.62E+00**	**4.17E+00**	**1.48E-04**	15
Sn	<2.94E+00	<2.94E+00	<2.94E+00	<2.48E-05	n/a
Sr	<3.00E-02	<3.00E-02	<3.00E-02	<3.42E-07	n/a
Th	<1.94E+00	<1.94E+00	<1.94E+00	<8.36E-06	n/a
Ti	<8.00E-02	<8.00E-02	<8.00E-02	<1.67E-06	n/a
U	<1.17E+01	<1.17E+01	<1.17E+01	<4.92E-05	n/a
V	<1.04E-01	<1.04E-01	<1.04E-01	<2.04E-06	n/a
Zn	<2.62E-01	<2.62E-01	<2.62E-01	<4.01E-06	n/a
Zr	<1.12E-01	<1.12E-01	<1.12E-01	<1.23E-06	n/a
Fluoride	<1.00E+02	<1.00E+02	<1.00E+02	<5.26E-03	n/a
Formate	<1.00E+02	<1.00E+02	<1.00E+02	<2.22E-03	n/a
Chloride	<1.00E+02	<1.00E+02	<1.00E+02	<2.82E-03	n/a
Nitrite	**1.29E+03**	**1.30E+03**	**1.30E+03**	**2.81E-02**	0.55
Bromide	<5.00E+02	<5.00E+02	<5.00E+02	<6.26E-03	n/a
Nitrate	**9.12E+03**	**9.14E+03**	**9.13E+03**	**1.47E-01**	0.15
Phosphate	<1.00E+02	<1.00E+02	<1.00E+02	<1.05E-03	n/a
Sulfate	**3.01E+03**	**3.03E+03**	**3.02E+03**	**3.14E-02**	0.47
Oxalate	<2.00E+02	<2.00E+02	<2.00E+02	<2.27E-03	n/a
TIC	**1.58E+02**	**1.60E+02**	**1.59E+02**	**1.32E-02**	0.89

Analyte	Replicate 1 mg/L	Replicate 2 mg/L	Average mg/L	Average M	RSD %
TOC	**1.00E+03**	**1.01E+03**	**1.01E+03**	**8.37E-02**	0.70
TC	**1.16E+03**	**1.17E+03**	**1.17E+03**	**9.70E-02**	0.61
Carbonate*	**7.89E+02**	**7.99E+02**	**7.94E+02**	**1.32E-02**	0.89
Hydroxide	nm	nm	nm		
	Replicate 1	**Replicate 2**	**Replicate 3**		
Density	nm	nm	nm		

RSD = Relative Standard Deviation
TIC = Total Inorganic Carbon
TOC = Total Organic Carbon
TC = Total Carbon
* Calculated from corresponding TIC values
n/a = Not applicable
nm = Not measured
Values with "<" symbols are detection limits. Note: Non-detection limit values are in bold.

Table 3-7. Analyte Mass Balance from the Evaporation Test and the Subsequent Filtration/Washing.

Analyte	Amount in As-Received Supernate	Amount in Concentrate Supernate	Amount in 1st Wash Filtrate	Amount in 2nd to 4th Wash Filtrates	Total	Ratio of Total to ARS	Ratio of CS to ARS
	ARS	CS	1WF	2T4WF	CS + 1WF + 2T4WF	(CS + 1WF + 2T4WF)/ ARS	CS/ARS
	mg	mg	mg	mg	mg		
Ag							
Al	2.08E+03	1.79E+03	4.04E+01	1.46E+01	1.85E+03	0.89	0.86
B	1.78E+01	2.42E+01		1.37E-01	2.43E+01	1.36	1.36
Ba							
Be							
Ca	9.90E-01	3.90E-01		4.23E-02	4.33E-01	0.44	0.39
Cd							
Ce							
Co							
Cr	3.32E+01	2.99E+01		2.46E-01	3.01E+01	0.91	0.90
Cu							
Fe	2.74E+00	2.09E+00			2.09E+00	0.76	0.76
Gd							
K	3.01E+02	2.98E+02		2.27E+00	3.00E+02	1.00	0.99
La							
Li							
Mg	4.91E-01	4.49E-02			4.49E-02	0.09	0.09
Mn							
Mo	9.76E+00	1.11E+01			1.11E+01	1.14	1.14
Na	3.47E+04	2.92E+04	1.31E+03	7.35E+02	3.12E+04	0.90	0.84
Ni							
P	6.18E+01	5.93E+01			5.93E+01	0.96	0.96
Pb							
S	2.85E+02	1.40E+02	7.46E+01	7.68E+01	2.91E+02	1.02	0.49
Sb							
Si	<7.63E+00	2.66E+01	<3.14E+00	2.50E-01	2.68E+01	n/a	n/a
Sn							
Sr							
Th							
Ti							
U							
V							
Zn	1.82E+00	1.58E+00			1.58E+00	0.86	0.86
Zr							
Fluoride							
Formate							
Chloride							
Nitrite	8.59E+03	7.59E+03		7.77E+01	7.67E+03	0.89	0.88
Bromide							
Nitrate	2.31E+04	1.72E+04	9.70E+02	5.48E+02	1.87E+04	0.81	0.74
Phosphate	1.16E+02					n/a	n/a

Analyte	**Amount in As-Received Supernate**	**Amount in Concentrate Supernate**	**Amount in 1st Wash Filtrate**	**Amount in 2nd to 4th Wash Filtrates**	**Total**	**Ratio of Total to ARS**	**Ratio of CS to ARS**
	ARS	**CS**	**1WF**	**2T4WF**	**CS + 1WF + 2T4WF**	**(CS + 1WF + 2T4WF)/ ARS**	**CS/ARS**
	mg	**mg**	**mg**	**mg**	**mg**		
Sulfate	8.55E+02			1.81E+02	1.81E+02	0.21	n/a
Oxalate							
TIC	4.91E+02	2.33E+02	8.88E+01	9.54E+00	3.31E+02	0.67	0.47
TOC	4.31E+02	2.21E+02	3.96E+02	6.03E+01	6.77E+02	1.57	0.51
TC	9.22E+02	4.53E+02	4.85E+02	6.99E+01	1.01E+03	1.09	0.49
Carbonate	2.45E+03	1.16E+03	4.44E+02	4.77E+01	1.65E+03	0.67	0.47

Blank cell means value is less than the detection limit and is excluded in the ratio calculations. Note that the less than detection limit amounts for Si are provided even though they were excluded in the ratio calculations.

n/a = Not applicable

ARS = As-Received Supernate (Pre-Evaporation Solution)
CS = Concentrate Supernate
1WF = 1st Wash Filtrate
2T4WF = 2nd to 4th Wash Filtrates

Volume of As-Received Supernate = 177 mL
Volume of Concentrate Supernate = 115 mL. Equals volume of concentrate minus volume of solids in concentrate = 125 - 10 = 115 mL. Note: Volume of solids in concentrate was visually assumed to be 10 mL.
Volume of 1st Wash Filtrate = 20 mL
Volume of 2nd to 4th Wash Filtrates = 60 mL

4.0 Conclusions

Evaporation of the Tank 26F supernate resulted in the precipitation of solids upon cooling. All the solids formed dissolved upon washing with deionized water. The bulk of the solids formed [Trona - $Na_3H(CO_3)_2 \cdot 2H_2O$, Thermonatrite - $Na_2CO_3 \cdot H_2O$, Nitratine - $NaNO_3$, and Natrite - Na_2CO_3] are known to be fairly soluble in water. The only sparingly soluble compound identified in relatively small amount is Silicon Oxide or Silica (SiO_2).

Even though the silica that formed dissolved due to its small amount and also being relatively fresh, it may not be the case for the 2F Evaporator which operates in a continuous mode. Increasing amounts or build-up of aged silica with time may render the silica more insoluble. This is because relatively new crystals are also generally more soluble than aged crystals.

5.0 Recommendations

It seems water should be adequate to dissolve the solids that form from 2F Evaporator feed that does not contain residual oxalic acid. Increasing the temperature of the water may speed up the dissolution. The caution is that the solubility of sodium carbonate peaks around 35 ^oC even though the decrease beyond 35 ^oC is not drastic.

The only solid component that has a slight chance of being problematic is silica. However, limiting its build-up and aging will lead to a more complete dissolution of the silica.

6.0 References

1. C. E. Duffey, “2F Evaporator Pluggage Issues,” HLE-TTR-2011-002, Rev. 0, January, 2011.

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3. W. D. King, M. S. Hay, and C. J. Coleman, “Tank 26F Supernatant and 2F Evaporator Eductor Pump Sample Characterization Results,” SRNL-STI-2011-00375, Rev. 0, August 2011.

4. C. E. Duffey, “Re: Tank 26 Sample,” Email, March 15, 2012, 03:43 PM.

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