Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Russell, Renee , Snow, Lanée and Peterson, Reid(2010) 'Methods to Avoid Post-Filtration Precipitation in Treatment of High-Level Waste', Separation Science and Technology, 45: 12, 1814 — 1821
To link to this Article: DOI: 10.1080/01496395.2010.493806
URL: http://dx.doi.org/10.1080/01496395.2010.493806

Separation Science and Technology, 45: 1814–1821, 2010

ISSN: 0149-6395 print/1520-5754 online
DOI: 10.1080/01496395.2010.493806

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Methods to Avoid Post-Filtration Precipitation in Treatment of High-Level Waste

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The Hanford Tank Waste Treatment and Immobilization Plant, currently under construction for treating high level waste at the Hanford Site, will rely on ultrafiltration to provide solids/liquid separation as a core part of the treatment process. A series of bench-scale simulant tests have been performed to evaluate the potential for post-filtration precipitation. These tests focused on identifying precipitation from a range of potential feed compositions and providing the data required to evaluate mitigation options. This data is also important to the Savannah River Site as they are also attempting to dissolve saltcake from their tanks. Inadvertent solids formation after saltcake dissolution, either within a staging tank or in transfer piping can have serious repercussions on the eventual retrieval and transfer of the waste. A series of tests were performed using a variety of simulant samples. These tests identified the expected extent of supersaturation that develops under normal operations and identified and characterized the solids phases that are expected to form when the filtrate solutions are stored. In addition, tests identified the potential to mitigate the formation of these solids through both dilution and the application of increased temperature.

Keywords Hanford; high-level waste; kinetics; post-filtration; precipitation; simulant

INTRODUCTION

The Hanford Tank Waste Treatment and Immobilization Plant (WTP) is currently under construction for treating high-level waste (HLW) at the U.S. Department of Energy's Hanford Site near Richland, Washington. The plant will rely on ultrafiltration to provide solids/liquid separation as a core part of the treatment process. Leaching and ultrafiltration processes for WTP were recently demonstrated using an engineering-scale Pretreatment Engineering Platform (PEP), which was designed to simulate the WTP pretreatment caustic leaching, oxidative leaching, ultrafiltration solids concentration, and slurry washing processes. During that demonstration, significant post-filtration precipitation was observed in the post-caustic leachate and wash solutions. Precipitation in this stream could lead to precipitates accumulating in filtrate receipt vessels and in the feed to ion exchange columns. Post-filtration precipitation in the feed to the ion exchange column would have a very significant effect on the operability of the unit and the overall pretreatment process. Therefore, it is essential to develop an understanding of both the solubility and kinetics of the post-filtration precipitation phenomenon.

Several solubility studies have been performed previously with the anions of interest under several different conditions (1–3); however, this study includes the kinetics of precipitation of these anions with solubility data to augment the previous data. Kinetics and the temperature dependence of the precipitation are important to the WTP because they will determine how much time will be available to process the solution before precipitation occurs and how maintaining a certain temperature may keep the precipitation from occurring.

This data is also important to the Savannah River Site (SRS) as they are also attempting to dissolve saltcake from their tanks and have studied and modeled the dissolution process extensively (4–6). Inadvertent solids formation after dissolution, either within a staging tank or in transfer piping, can have serious repercussions on the eventual retrieval and transfer of the waste. Previous salt cake dissolution work has shown that solids re-precipitation can occur when different dissolution fractions are mixed together (7). The propensity for additional solids precipitation, should other streams be considered within the total process, is not known, but is likely to depend on the composition, ionic strength, and volume of the streams being mixed.

EXPERIMENTAL

Determining the Rate of Approach to Saturation Concentrations

To determine the rate at which the anions of interest (phosphate, oxalate, sulfate, silicate, and fluoride) approach equilibrium solution composition in post-caustic-leach slurry at 25°C before filtration, a set of tests was performed using simplified simulated post-caustic-leach slurry (derived from the PEP simulant composition) with the

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Received 1 November 2009; accepted 16 March 2010.

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TABLE 1
Simulated post-caustic-leach slurry composition

Chemical	Amount (g/kg)
Fe-rich slurry	40.0
$NaNO_3$	75.3
$NaNO_2$	18.5
NaOH	149.6
$NaAl(OH)_4$	36.9
Na_2CO_3	7.42
H_2O	677.2

composition shown in Table 1. The Fe-rich slurry was composed of primarily iron oxy-hydroxide with small amounts of other metal hydroxides. The insoluble metal hydroxide solids were produced when NaOH was added to a metal nitrate solution to a pH of 10–11. The excess nitrate was then washed from the slurry. This slurry was based on previous actual Hanford tank waste compositions (8).

The simulated post-caustic-leach slurry was spiked with each anion of interest (phosphate, oxalate, sulfate, silicate, and fluoride) in the form of a sodium salt as shown in Table 2 in an amount sufficient to super-saturate the solution.

These 11 test slurries were placed in an oven at 85°C for 25.5 hrs. They were hand-shaken once every hour for the first 12 hours to dissolve the added salts and super-saturate the solution with respect to room temperature. Then they were allowed to sit in the oven for the remaining 13.5 hours. After 25.5 hours at 85°C, the test slurries were removed from the oven and sampled by placing approximately 1.5 mL of supernate (filtered through a 0.45-μm filter immediately) into a sample bottle containing de-ionized water to dilute it to approximately 1:10. Then the test slurries were placed into an incubator, which was at a temperature of 71.1°C, and cooled over 5.5 hours to 25 ± 1°C while shaking at 150 rpm to well mix them.

These test slurries were then held at 25 ± 1°C for 24 hours in the incubator while shaking at 150 rpm. They were each sampled at 0, 1, 2, 4, 8, 12, and 24 hours at 25 ± 1°C. After holding for 24 hours at 25 ± 1°C, the temperature of the incubator was lowered to 16 ± 1°C over 4.3 hours while continuing to shake at 150 rpm. Another sample was taken from each of the test slurries after they had been held at 16 ± 1°C for 20 hours. The temperature of the incubator was again increased to 25 ± 1°C over 3.5 hours and held at temperature for ~93 hours. Samples were taken from each of the test slurries at 24 and 93 hours after the temperature had been raised with the shaker remaining at 150 rpm. Each sample was taken by placing approximately 1.5 mL of supernate (filtered through a 0.45-μm filter immediately) into a sample bottle containing de-ionized water to dilute it to approximately 1:10.

Tests to Identify and Characterize the Precipitate Formed in Post-Caustic-Leach Filtrate

A second set of tests were performed also using simulated post-caustic-leach slurry (derived from the PEP simulant) with the composition shown in Table 1. The simulated post-caustic-leach slurry was spiked with each anion of interest (phosphate, oxalate, sulfate, silicate, and fluoride) in the form of a sodium salt with the same composition as shown in Table 2 to identify precipitates formed at ambient temperature (20°C) in the presence of phosphate, oxalate, sulfate, silicate, and fluoride anions in the post-caustic-leachate solution.

The test slurries were placed in the incubator at 35 ± 1°C and held there for at least 46 hours while shaking at

TABLE 2
Test matrix identification for rate of approach to saturation tests

Test ID	Post-caustic leach slurry (g)	$Na_2C_2O_4$-$2H_2O$ (g)	Na_3PO_4-$12H_2O$ (g)	Na_2SO_4 (g)	NaF (g)	Na_2SiO_3-$9H_2O$ (g)
T2–1a	1000.06	80.88	52.63	14.75	13.59	14.33
T2–1b	1000.03	80.88	52.63	14.75	13.59	14.33
T2–1c	1000.06	80.88	52.63	14.75	13.59	14.33
T2–2	1000.07	80.88	52.63	14.75	0.00	0.00
T2–3	1000.03	0.00	52.63	0.00	13.59	14.33
T2–4	1000.05	0.00	52.63	0.00	0.00	0.00
T2–5	1000.08	0.00	52.63	14.75	13.59	14.33
T2–6	1000.01	0.00	0.00	14.75	0.00	0.00
T2–7	1000.07	80.88	0.00	0.00	13.59	14.33
T2–8	1000.03	80.88	0.00	0.00	0.00	0.00
T2–9	1000.05	0.00	0.00	0.00	0.00	0.00

150 rpm to well mix them. After 46 hours, the test slurries were centrifuged and then filtered through a 0.45-μm filter at 35°C. They were sampled by placing approximately 1.5 mL of supernate (filtered through a 0.45-μm filter) into a sample bottle with de-ionized water to dilute it to approximately 1:10. This filtering was difficult because of filter clogging from iron oxy-hydroxide. Multiple filters were used during the filtering of each test sample, which required more time per slurry (~1 to 2 hours) and increased the time the slurry was held at 35 ± 1°C. Filtering continued for 22 hours to filter all of the test slurries. Immediately after filtering, the test slurries were split by pouring the filtered supernatant into two bottles—one 250-mL bottle and one 1-L bottle. The smaller bottle was used for a settling test and crystal size testing. The larger bottle was used for all of the other analyses. The filtered test supernates were cooled to 20 ± 1°C in the incubator over 5 hours while shaking at 165 rpm to provide better mixing. After being held at 20 ± 1°C for 92 to 96 hours, another sample was taken by placing approximately 1.5 mL of supernate (filtered through a 0.45 μm filter) into a sample bottle with de-ionized water to dilute it to approximately 1:10.

Testing of the Dilution Required to Re-Dissolve the Precipitate

The next set of tests was performed to determine the potential to re-dissolve the post-filtration precipitate from the PEP testing wash solutions through dilution with de-ionized water at 25 ± 1°C. These tests were performed in duplicate by placing 100 mL of the test solutions into 200-mL centrifuge tubes and holding them at 25 ± 1°C for 48 hours while shaking at 150 rpm. After 48 hours, the samples were centrifuged, the volume of solids was measured, and then 5 mL of de-ionized water was added. The samples were then shaken to re-suspend the solids and then held at 25 ± 1°C for 24 hours while continuing to shake at 150 rpm. The process was repeated until the solids completely dissolved or the centrifuge tubes became full.

Tests to Determine the Super-Saturation in Post-Caustic-Leach Filtrates from PEP

The objective of these tests was to determine the solution super-saturation in the post-caustic-leach filtrate during the dewatering and washing period based on the samples collected during the PEP testing. Samples were tested by placing them in an incubator at 20 ± 1°C for 72 hours while shaking at 150 rpm. After 72 hours at 20 ± 1°C, supernate samples were taken by placing approximately 1.5 mL of supernate (filtered through a 0.45-μm filter immediately) into a sample bottle containing de-ionized water to dilute it to approximately 1:10.

The temperature was then increased 1°C every 24 hours and the samples allowed to equilibrate. After each 24-hour period, the samples were inspected for the remaining solids. This continued until the temperature reached 30°C. After which, the solids remained in the majority of the samples. Therefore, the temperature was increased to 35°C, and the samples were allowed to equilibrate for 24 hours. Supernate samples were taken at 25°C and at 35°C by placing approximately 1.5 mL of supernate (filtered through a 0.45-μm filter immediately) into a sample bottle containing de-ionized water to dilute it to approximately 1:10.

Analytical Techniques Used

Each supernate sample was analyzed by inductively coupled plasma (ICP) for Al, Na, K, P, S, Si, and Fe and by ion chromatography (IC) for oxalate, phosphate, sulfate, fluoride, nitrate, and nitrite.

These crystals were then analyzed by polarized light microscopy and a scanning electron microscope equipped with an energy dispersive spectrometer (EDS) to determine their composition by fixing the crystals onto carbon sticky tape and coating the crystals with a conductive coating of Pd using a sputter coater. Note that EDS-measured compositions are limited to elements with a mass equal to or greater than carbon.

RESULTS AND DISCUSSION

Determination of the Rate of Approach to Saturation Concentrations

The ICP results of the supernatant samples from the tests shown in Table 2 showed very low levels of silicon present and no evidence of alumino-silicate precipitation.

Based on the tests in Table 2, Fig. 1 shows the precipitation of oxalate in the samples by the decrease in oxalate concentration. Oxalate appeared to be fairly slow to precipitate, taking nearly 24 hours before reaching equilibrium. The presence of phosphate, sulfate, or fluoride did not significantly change the equilibrium concentration of

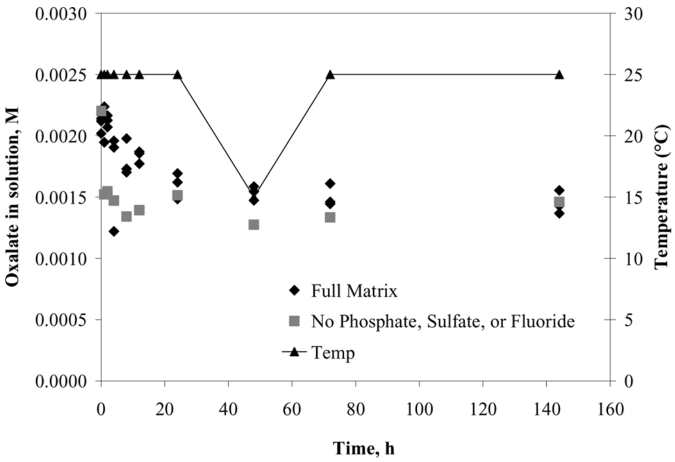

FIG. 1. Oxalate precipitation in the rate to saturation tests.

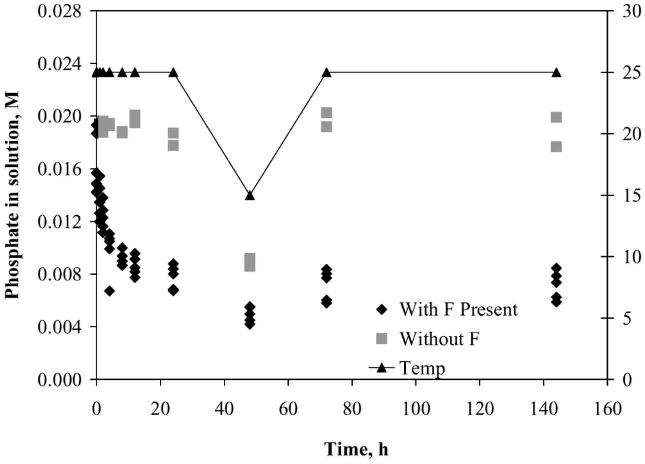

FIG. 2. Phosphate precipitation in the rate to saturation tests.

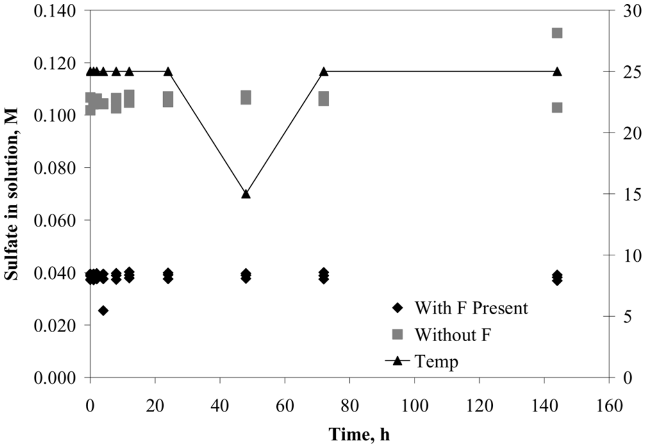

FIG. 4. Sulfate precipitation in the rate to saturation tests.

oxalate, but the kinetics appeared to be slower in the presence of the other anions. The average oxalate concentration was 1.56 M at long times. Note that the initial concentration in the full matrix is significantly higher, up to 2.27 M. Results discussed below show that oxalate solubility is relatively insensitive to temperature. The solubility at 35°C in this matrix was approximately 1.82 M (see Fig. 7). Thus, this initial supersaturation was significantly in excess of 10°C.

Figure 2 shows the precipitation of phosphate in the samples. In the absence of fluoride, phosphate appeared to equilibrate very quickly. However, in the presence of fluoride, phosphate equilibrated much slower and to a significantly lower solubility. This was demonstrated in a previous study, namely, that adding fluoride to solutions containing phosphate significantly decreases the amount of phosphate that can remain in solution (9). Figure 3 shows the average of the last two phosphate and fluoride measurements from each of the test slurries in Table 2 that contained both phosphate and fluoride. The solubility of phosphate supersaturation seen for sodium fluoride phosphate is approximately 6.32 M. This extent of phosphate supersaturation (in the absence of fluoride) is again significantly more than 10°C of supersaturation. The observed saturation at 35°C (see Fig. 6) was approximately 10.5 M, while the initial concentration at 25°C was over 16.8 M.

Figure 4 shows that sulfate appeared to equilibrate almost immediately regardless of the presence of fluoride. However, the presence of fluoride did appear to impact the sulfate solubility by significantly decreasing it. It should be noted that for the tests without fluoride, the quantity of sulfate in the supernatant was likely the entire sulfate in the simulant. As such, this test was not a good measure of sulfate kinetics or solubility, but it did indicate that sulfate is very soluble in this matrix.

Interestingly, fluoride appeared to equilibrate within a few hours for all the samples as shown in Fig. 5. However, the presence of oxalate appeared to impact

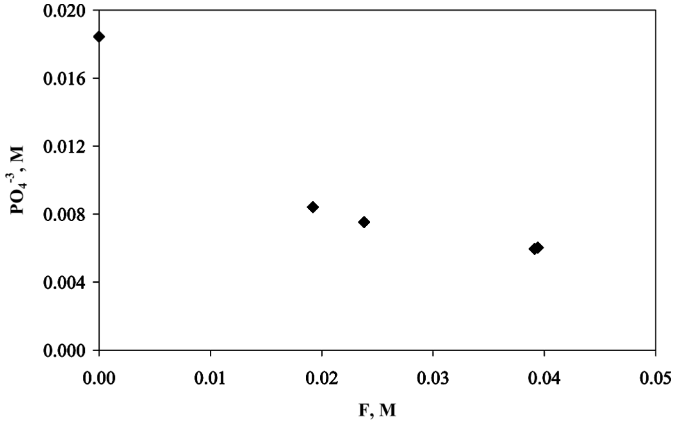

FIG. 3. Phosphate solubility as a function of fluoride concentration.

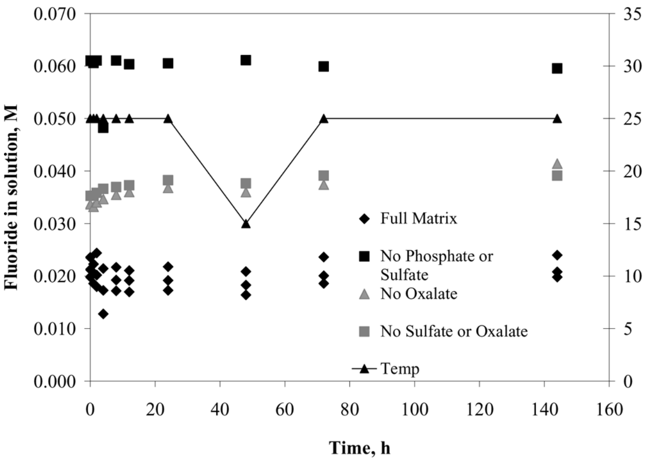

FIG. 5. Fluoride precipitation in the rate to saturation tests.

the fluoride solubility, but fluoride did not impact the oxalate solubility.

Identification and Characterization of the Precipitate Formed in Post-Caustic-leach Filtrate

Tests T3–2, T3–6, T3–7, T3–8, and T3–9 (see Table 2 for test compositions) did not produce solids. Test T3–4 with only phosphate added produced the most solids. Tests T3–1a, T3–1b, T3–1c, T3–3, and T3–5 only produced approximately 1 mL or less of solids. Tests T3–2, T3–6, T3–7, T3–8, and T3–9 were seeded with one drop of supernate containing crystals from each of Test T3–4 and T3–1a and allowed to mix over a weekend. However, they still did not produce any crystals after 96 hours at 20°C.

Table 3 provides the measured difference in ion concentrations between the 35°C and 20°C samples. A greater amount of precipitation is expected for larger concentration differences. The blank entries represent tests where the species of interest was omitted from the slurry matrix. This table indicates that tests T3–1a, T3–1b, T3–1c, T3–3, T3–4, and T3–5 should have a significant amount of precipitation. Tests T3–2, T3–7, and T3–8 should have a small amount of precipitation with Test T3–6 and Test T3–9 having no precipitation. These results indicate that very small quantities of oxalate and sulfate will precipitate upon cooling. The results indicate that larger quantities of sodium fluoride phosphate will precipitate upon cooling.

Sodium phosphate solubility as a function of fluoride concentration from the results of all tests discussed above is shown in Fig. 6. Data are presented for equilibrium temperatures of ~15°C, 20°C, 25°C, and 35°C. The amount of phosphate in solution at 35°C in Test T3–2 appeared to be anomalously low. At this point, there is not a good explanation for this low value, but the sample did not appear to be at equilibrium at 35°C. This may be due to either the sample re-equilibrating quickly upon cooling during the separation process or the sample not yet reaching equilibrium during storage at temperature. The amount of phosphate in solution was about 1/4 of the expected amount based on Test T3–4.

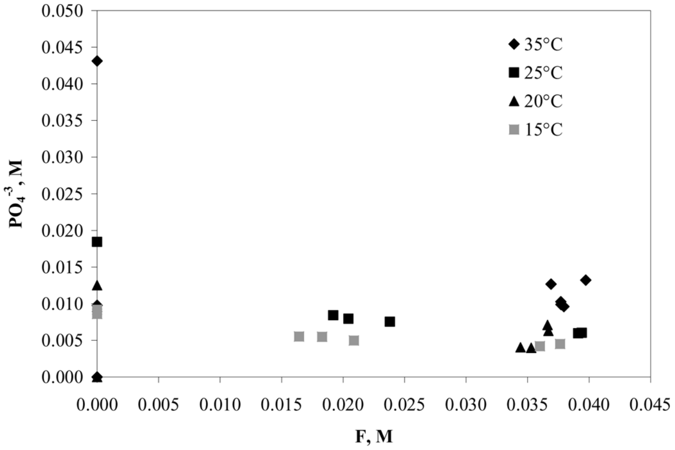

FIG. 6. Phosphate solubility as a function of fluoride concentration.

Figure 7 shows the impact of temperature on solubility of the different anions measured at 24 hours for tests at each of these temperatures. This shows that only sodium phosphate and sodium fluoride phosphate exhibited any significant change in solubility with temperature. The other main components appeared to be relatively insensitive to temperature.

Table 2 sample solutions containing precipitated crystals after all testing was completed were analyzed by optical

TABLE 3
Measured difference in ion concentration between 35°C and 20°C

Test ID	F (μg/mL)	C_2O_4 (μg/mL)	PO_4 (μg/mL)	SO_4 (μg/mL)
T3–1a	62	25	590	−20
T3–1b	24	7	315	−16
T3–1c	21	−5	265	−43
T3–2	—	9	19	4
T3–3	84	—	876	—
T3–4	—	—	2906	—
T3–5	79	—	817	13
T3–6	—	—	—	−94
T3–7	−6	14	—	—
T3–8	—	15	—	—
T3–9	—	—	—	—

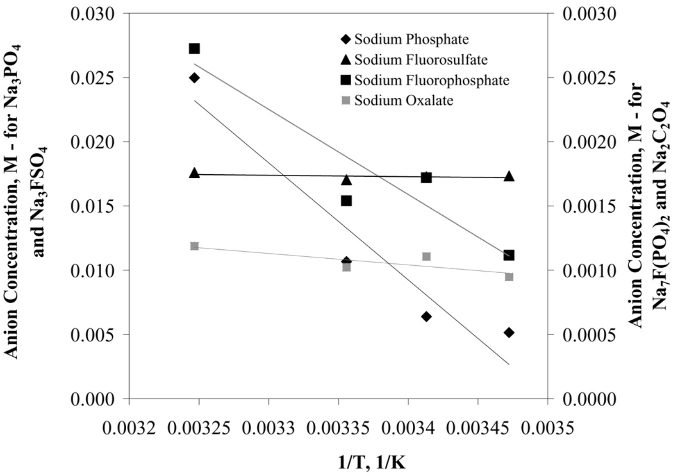

FIG. 7. Impact of temperature on solubility measured at 24 hours.

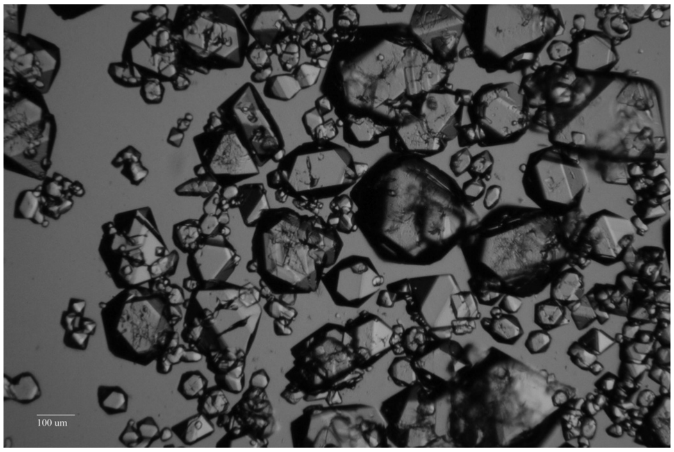

FIG. 8. Sample #T3–5 optical microscopy in polarized light at 50 × (likely $Na_7F(PO_4)_2 \cdot 19H_2O$).

microscopy to gather optical properties of the crystals. All of the samples in Table 2, with the exception of T3–4, contained the same cubic crystal type. The crystals were determined to be cubic from the observed morphology in polarized light, as shown in Fig. 8, and when under cross-polarized light, there was no change. These crystals were $Na_7F(PO_4)_2 \cdot 19H_2O$ based on their high refractive index and comparisons with previous compilations (10).

Sample T3–4 contains long, thin, needle-shaped crystals that appeared white under cross-polarized light. When a full wave plate was inserted into the cross-polarized light, the crystals appeared blue when the long axes of the crystal and the wave plate were parallel and yellowish-orange when perpendicular, as shown in Fig. 9. These crystals were most likely $Na_3PO_4 \cdot 12H_2O$.

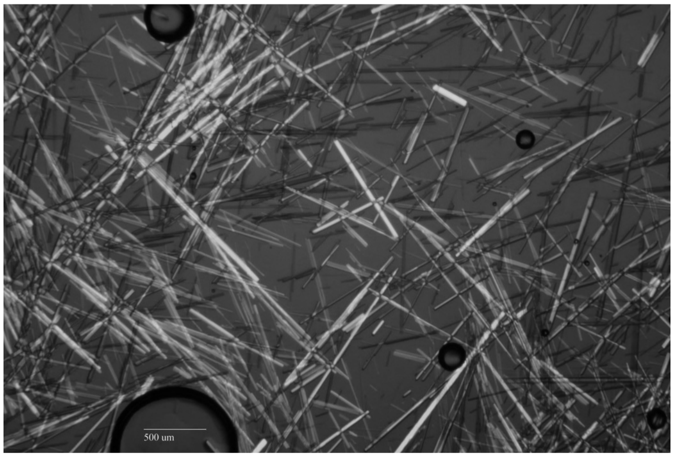

FIG. 9. Sample #T3–4 crystals in OM cross-polarized light with full wave plate at 12.5 × (likely $Na_3PO_4 \cdot 12H_2O$).

TABLE 4
Amount de-ionized water required to achieve complete dissolution of solids

Test ID	Sample Amt (mL)	De-ionized water added (mL)	Comments
T4-A-2367a	105	30	No solids at this point
T4-A-2367b	110	30	No solids at this point
T4-A-2587a	105	30	No solids at this point
T4-A-2587b	105	30	No solids at this point
T4-B-4232a	100	30	No solids at this point
T4-B-4232b	105	30	No solids at this point
T4-B-4267a	110	85	Stopped with skiff of solids left
T4-B-4267b	113	85	Stopped with skiff of solids left
T4-D-5652a	98	85	Stopped with skiff of solids left
T4-D-5652b	99	85	Stopped with skiff of solids left
T4-D-5457a	101	50	No solids at this point
T4-D-5457b	103	50	No solids at this point
T4-D-5478a	105	55	No solids at this point
T4-D-5478b	104	50	No solids at this point

Testing of the Dilution Required to Re-Dissolve the Precipitate

Table 4 shows the amount of dilution required for each test to achieve complete solids dissolution or when the centrifuge tube became full. Figure 10 shows how the solids decreased as a function of dilution with 10% error bars included. After a certain point in several of the samples, the solids dissolved an unobservable amount so that it

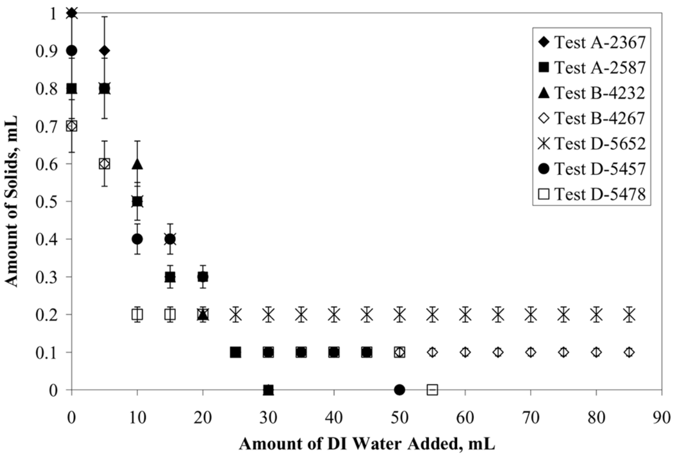

FIG. 10. Solids dissolution with de-ionized water addition.

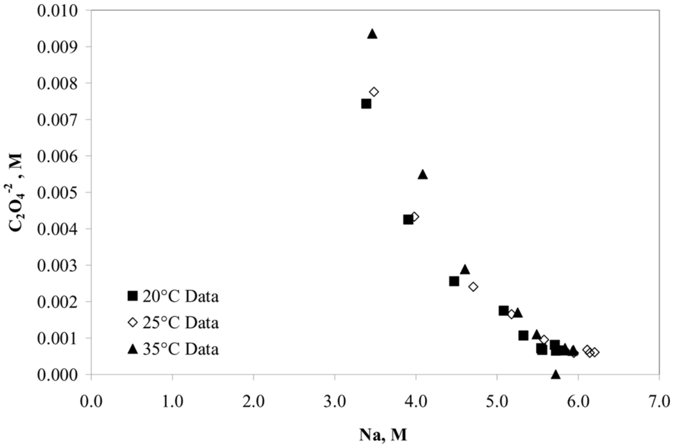

FIG. 11. Oxalate concentration relative to sodium concentration during PEP test washes.

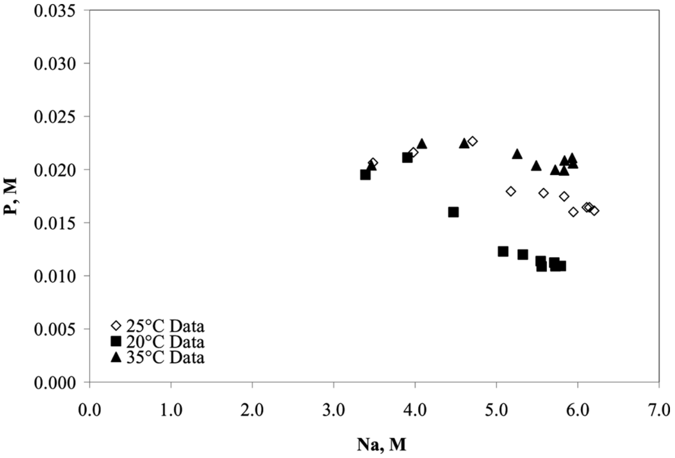

FIG. 12. Phosphorus concentration relative to sodium concentration during PEP test washes.

appeared they were not dissolving. The dilution was stopped after 85 mL of de-ionized water was added due to the centrifuge tube being full. However, these results show that the solids are essentially soluble if enough de-ionized water is added. The dried remaining solids weighed 50 mg for T4-D-5652 and 5 mg for T4-B-4267.

Determination of the Super-Saturation in Post-Caustic Leach Filtrates from PEP Testing

The results of the oxalate dissolution for these samples are shown in Fig. 11 with amount of washing increasing from right to left in the figure. Figure 11 shows that there was little to no dissolution of oxalate after heating the solutions from 20°C to 35°C except for washes # 15 and #18 (the two on the far left of the figure) where an increase occurred from 25°C to 35°C. This can be explained by the fact that in the earlier washes, oxalate was not very soluble due to all of the other dissolved ions present, so no precipitated oxalate dissolved upon increasing the temperature. However, in washes #15 and #18, the concentration of the other ions was low enough for the oxalate to dissolve. Therefore, when the temperature was increased, the amount of the oxalate in the supernate increased in these samples.

The results of the phosphorus (phosphate) dissolution for these samples are shown in Fig. 12, with the number of washes increasing from right to left in the figure. It can be seen in Fig. 12 that the phosphorus (phosphate) concentration in solution increased significantly upon heating to 35°C for most of the samples with the exception of washes #12, #15, and #18 (the three on the far left of the figure). This can be explained by the fact that by this point in PEP processing, most of the phosphorus (phosphate) had been dissolved and washed out, and therefore, there was no more to dissolve when the temperature was increased in these tests.

At the end of the test, the ~1-L samples were filtered immediately at 35°C using 0.45-μm filters to determine how many solids were left in each test bottle. These solids were air-dried and weighed, and the results are shown in Table 5. Each sample contained brownish solids on the filter, and some of them had crystals also.

EDS analysis of the crystals reveals a chemistry of mainly Na, O, and C, with minor amounts of Al, S, and Cu. Samples from washes #12, #15, and #18 were all similar in chemical composition to wash #9. These samples all appeared to be $Na_2C_2O_4$, based on the thin needle crystals and measured chemistry.

Sample #4153 from the PEP process supernatant prior to washing was quite different from the other samples in terms of morphology and chemistry of the residual solids. The morphology of the crystals was wide, large crystals that appear to have cracked significantly, presumably upon drying. The measured chemistry of this sample was mainly Na, P, and O, with minor amounts of Al, C, Cu, and S. This sample appeared to be $Na_3PO_4 \cdot 12H_2O$, based on the measured chemistry and the large needle-shaped crystals.

TABLE 5
Solids left in samples after test completed

Sample ID	Wash #	Solids left (g)	Sample ID	Wash #	Solids left (g)
4153	supernate	0.1591	4229	7	0.1608
4154	supernate	0.1350	4231	9	0.1532
4223	1	0.2116	4234	12	0.2535
4225	3	0.1094	4237	15	0.3445
4227	5	0.1098	4240	18	0.5374

CONCLUSIONS

The following conclusions can be made from this testing:

- Oxalate appears to be fairly slow to precipitate, taking nearly 24 hours before reaching equilibrium. The presence of phosphate, sulfate, or fluoride did not significantly change the equilibrium concentration of oxalate, but the kinetics appeared to be slower in the presence of the other anions.
- In the absence of fluoride, phosphate appears to equilibrate very quickly. However, in the presence of fluoride, phosphate equilibrates much slower and to a significantly lower solubility. Phosphate solubility appears to be a strong function of fluoride.
- Sulfate appears to equilibrate quickly regardless of the presence of fluoride. However, the presence of fluoride does appear to impact the sulfate solubility by significantly decreasing it.
- Fluoride appears to equilibrate fairly quickly for all compositions. However, while the presence of oxalate appears to impact the fluoride solubility, fluoride did not impact the oxalate solubility.
- Only sodium phosphate and sodium fluoride phosphate exhibit any significant change in solubility with temperature. The other main components appear to be relatively insensitive to temperature.
- The solids are essentially soluble if enough de-ionized water is added.
- There was little to no dissolution of oxalate after heating the solutions from 20°C to 35°C until the concentration of the other ions was low enough for the oxalate to dissolve. Therefore, when the temperature was increased, the amount of oxalate in the supernate increased in these samples.
- The phosphorus (phosphate) concentration in solution increased significantly upon heating to 35°C for most of the samples until most of the phosphorus (phosphate) had been dissolved and washed out, and therefore there was no more to dissolve when the temperature was increased.

ACKNOWLEDGEMENTS

Pacific Northwest National Laboratory is operated for the U.S. Department of Energy by Battelle under Contract DE-AC05-76RL01830. This work was funded by the U.S. Department of Energy through the Office of Environmental Management and under the guidance of Bechtel National, Inc.

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