

Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U. S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1) warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2) representation that such use or results of such use would not infringe privately owned rights; or
- 3) endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.



Impact of Sodium Oxalate, Sodium Aluminosilicate, and Gibbsite/Boehmite on ARP Filter Performance

M. R. Poirier

P. R. Burket

November 2015

SRNL-STI-2015-00465, Revision 0



DISCLAIMER

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
2. representation that such use or results of such use would not infringe privately owned rights; or
3. endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

Printed in the United States of America

**Prepared for
U.S. Department of Energy**

Keywords: *ARP, Crossflow, 512-S,
Sintered-Metal Filter*

Retention: *Permanent*

Impact of Sodium Oxalate, Sodium Aluminosilicate, and Gibbsite/Boehmite on ARP Filter Performance

M. R. Poirier
P. R. Burket

November 2015

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



REVIEWS AND APPROVALS

AUTHORS:

M. R. Poirier, Hanford Mission Programs	Date
---	------

P. R. Burket, Engineering Processing Development	Date
--	------

TECHNICAL REVIEW:

C. A. Nash, Advanced Characterization and Process Technology, Reviewed per E7 2.60	Date
--	------

APPROVAL:

F. M. Pennebaker, Manager Process Technology Programs	Date
--	------

D. E. Dooley, Manager Environmental & Chemical Process Technology Research Programs	Date
--	------

E. J. Freed, Manager DWPF/Saltstone Facility Engineering	Date
---	------

EXECUTIVE SUMMARY

The Savannah River Site (SRS) is currently treating radioactive liquid waste with the Actinide Removal Process (ARP) and the Modular Caustic Side Solvent Extraction Unit (MCU). Recently, the low filter flux through the ARP of approximately 5 gallons per minute has limited the rate at which radioactive liquid waste can be treated. Salt Batch 6 had a lower processing rate and required frequent filter cleaning. Savannah River Remediation (SRR) has a desire to understand the causes of the low filter flux and to increase ARP/MCU throughput.

One plausible cause of the low filter flux is plugging of the primary (i.e., crossflow) or secondary filters. Three solids compounds that could be forming in the ARP Filter Feed Tank or within the ARP Filter System were identified. These solids are sodium oxalate, sodium aluminosilicate, and aluminum solids (i.e., gibbsite and boehmite).

SRR requested SRNL to conduct bench-scale filter tests to evaluate whether sodium oxalate, sodium aluminosilicate, or aluminum solids (i.e., gibbsite and boehmite) could be the cause of excessive fouling of the crossflow or secondary filter at ARP. The authors conducted the tests by preparing slurries containing 6.6 M sodium Salt Batch 6 supernate, 2.5 g MST/L slurry, and varying concentrations of sodium oxalate, sodium aluminosilicate, and aluminum solids, processing the slurry through a bench-scale filter unit that contains a crossflow primary filter and a dead-end secondary filter, and measuring filter flux and transmembrane pressure as a function of time.

The conclusions from this work follow.

- Testing with feed slurries containing sodium aluminosilicate and aluminum (gibbsite and boehmite)-containing solids gave no indication of a significant change in crossflow filter fouling compared to feeds containing MST only.
- While the sodium oxalate containing feeds had the same final crossflow filter flux as the baseline feed, the initial crossflow filter flux was significantly lower than the initial baseline crossflow filter flux. This difference could be from the presence of sodium oxalate or from other causes.
- The sodium oxalate containing feed showed no change in pressure drop of the secondary filter, but the pressure drop was larger than measured with the sodium aluminosilicate and gibbsite/boehmite containing feeds, indicating that some fouling may have occurred.
- The sodium aluminosilicate and aluminum solids containing feeds showed evidence of secondary filter fouling.
- All of the tests showed some evidence of fouling the secondary filter. This fouling could be from fine particles passing through the crossflow filter.
- The sodium oxalate containing feeds behaved differently than the sodium aluminosilicate and gibbsite/boehmite containing feeds.

TABLE OF CONTENTS

LIST OF TABLES	vii
LIST OF FIGURES	vii
LIST OF ABBREVIATIONS.....	viii
1.0 Introduction.....	1
2.0 Experimental	1
2.1 Equipment	1
2.1.1 Crossflow Filter	1
2.2 Test Protocol	3
2.3 Quality Assurance	6
3.0 Results.....	6
3.1 Crossflow Filter.....	6
3.2 Secondary Filter	11
4.0 Discussion.....	14
5.0 Conclusions.....	15
6.0 References.....	16

LIST OF TABLES

Table 1. Clean Water Flux Prior to Start of Testing	4
Table 2. Composition of Simulated Salt Solution.....	4
Table 3. Sodium Oxalate Concentration in Filter Feed.....	5
Table 4. Sodium Aluminosilicate Concentration in Filter Feed	5
Table 5. Aluminum Solids Concentration in Filter Feed	6

LIST OF FIGURES

Figure 1. Schematic of Laboratory-Scale Crossflow Filter Unit	3
Figure 2. Comparison of filter flux with and without Sodium Oxalate	7
Figure 3. Pressure Drop across Crossflow Filter for Sodium Oxalate Containing Feeds	7
Figure 4. Comparison of filter flux with and without Sodium Aluminosilicate	8
Figure 5. Pressure Drop across Crossflow Filter for Sodium Aluminosilicate Containing Feeds	8
Figure 6. Comparison of filter flux with and without Gibbsite/Boehmite Solids	9
Figure 7. Pressure Drop across Crossflow Filter for Gibbsite/Boehmite Containing Feeds.....	10
Figure 8. Pressure Drop across Secondary Filter for Baseline Feed.....	11
Figure 9. Secondary Filter Pressure Drop for Oxalate-Containing-Feed.....	12
Figure 10. Secondary Filter Pressure Drop for Sodium Aluminosilicate-Containing-Feed	13
Figure 11. Secondary Filter Pressure Drop for Aluminum-Containing-Feed.....	14

LIST OF ABBREVIATIONS

ARP	Actinide Removal Process
LWHT	Late Wash Hold Tank
LWPT	Late Wash Precipitate Tank
MCU	Modular Caustic Side Solvent Extraction Unit
MST	Monosodium Titanate
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
SRS	Savannah River Site
SS	Stainless Steel

1.0 Introduction

The Savannah River Site (SRS) is currently treating radioactive liquid waste with the Actinide Removal Process (ARP) and the Modular Caustic Side Solvent Extraction Unit (MCU). Recently, the low filter flux through the ARP of approximately 5 gallons per minute has limited the rate at which radioactive liquid waste can be treated. Salt Batch 6 had a lower processing rate and required frequent filter cleaning. There is a desire to understand the causes of the low filter flux and to increase ARP/MCU throughput.

One plausible cause of the low filter flux is plugging of the primary (i.e., crossflow) or secondary filters. Three solids compounds that could be forming in the ARP Filter Feed Tank or within the ARP Filter System were identified. These solids are sodium oxalate, sodium aluminosilicate, and aluminum solids (i.e., gibbsite and boehmite).

Oxalic acid is used to chemically clean the crossflow filter and secondary filter at ARP. Some oxalate may remain in the heels in the Late Wash Precipitate Tank (LWPT) and the Late Wash Hold Tank (LWHT) following chemical cleaning. When these heels are mixed with salt solution (~6.6 M sodium) or diluted salt solution, sodium oxalate could precipitate.¹ If the precipitate forms in the LWPT, it could cause fouling of the crossflow filter. If it forms in the LWHT or between the crossflow and secondary filter, it could cause fouling of the secondary filter.

The feed to ARP contains sodium aluminate, soluble silica, and free hydroxide. These chemical species have been observed to form sodium aluminosilicate in the SRS Tank Farm.² If sodium aluminosilicate formed in the ARP process, it could foul the crossflow or secondary filter.

The salt solution fed to the crossflow filter contains sodium hydroxide and sodium aluminate. At high pH the aluminate is soluble. However, as the feed slurry is washed, the pH and free hydroxide concentration decrease. If it decreases enough, the aluminum could precipitate, forming gibbsite or boehmite. This precipitation could cause fouling of the crossflow filter or the secondary filter.

SRR requested SRNL to conduct bench-scale filter tests to evaluate whether sodium oxalate, sodium aluminosilicate, or gibbsite/boehmite could be causing excessive fouling of the crossflow or secondary filter at ARP.³ The authors conducted the tests by preparing slurries containing 6.6 M sodium Salt Batch 6 supernate, 2.5 g MST/L slurry, and varying concentrations of sodium oxalate, sodium aluminosilicate, and gibbsite/boehmite, processing the slurry through a bench-scale filter unit (containing crossflow and secondary filters), and measuring filter flux and transmembrane pressure as a function of time.⁴

2.0 Experimental

2.1 Equipment

2.1.1 *Crossflow Filter*

SRNL personnel constructed a bench-scale filtration apparatus. Figure 1 shows the layout of the bench-scale filtration apparatus. The apparatus has an approximately 10 gallon feed tank with an impeller to mix the tank contents. The mixing system was not designed to be prototypic of the ARP; it was designed to suspend the MST particles in the feed slurry. A centrifugal pump draws the slurry from the feed tank and pumps it into two parallel lines at ~ 6.0 gpm total flow rate (~ 3.0 gpm to each filter). Each line has a heat exchanger to control the temperature of the feed slurry to 25 ± 2 °C. The slurry flows past a tee where the two lines meet and the inlet pressure transducer is located. Beyond the tee there is one valve on each line which can be used to adjust the flow rate to each filter. Following each valve is a $0 - 5 \text{ gpm} \pm 0.1 \text{ gpm}$ magnetic flowmeter which is used to measure the flow of slurry into each filter. The filters are located downstream of the flowmeters. After exiting the filters, the concentrated slurry streams are combined and returned to the feed tank. The concentrate line has a manual backpressure valve and an automated backpressure valve connected in parallel. The outlet from each of these valves returns the slurry to the bottom of the feed tank. All lines are ½" stainless steel (SS) tubing except for the instrument

lines to the pressure transducers which are 1/4" SS tubing, and the filtrate lines, which are 3/8" and 1/4" SS tubing.

The filtrate leaves each filter through 3/8" and 1/4" tubing. Pressure transducers measure the filtrate pressure immediately after each filter. A three way valve is positioned even with the top of a graduated tube for each filtrate line where the filtrate can be directed to the filtrate tank or to the 100 mL \pm 1 mL graduated tube which is used to manually measure the filtrate flowrate. For these tests, the filtrate flow was measured every 15 minutes. The filtrate flow could also be sent back to the feed tank by moving the tygon tubing from the filtrate tank to the feed tank.

One of the crossflow filters is a 0.1 μ m pore size, 3/8 inch ID Mott[®] porous metal crossflow filter and the other is a 0.5 μ m pore size, 3/8 inch ID Mott[®] porous metal crossflow filter. Both filters are 24 inches in length and constructed of sintered stainless steel. A computer was used to record the pressures, feed flow rates, and feed tank temperature as well as to control the automatic backpressure valve located after the filters. A secondary filter was installed in the filter system downstream of the pressure transducer on the filtrate side of each of the crossflow filters, to look for potential fouling of the secondary filter as has been observed at ARP. The filter is a Pall 2220 filter apparatus fitted with a flat sheet 0.5 micron Mott sintered stainless steel filter. The secondary filter area in the test rig was selected to approximate the ratio of secondary filter area to crossflow filter area in the ARP. To keep the ratio the same between the ARP and the test rig, the secondary filter area would be 0.0141 ft² or 13.1 cm² (0.197 ft² * 16.5 ft²/230 ft²). This area is based on an ARP crossflow filter area of 230 ft², an ARP secondary filter area of 16.5 ft², and a bench-scale crossflow filter area of 0.197 ft². The test rig filter surface area was selected to be 9.6 cm² (0.0103 ft²). A smaller secondary filter was selected to use an off the shelf filter and to increase the accumulation of solid particles at the filter surface (i.e., make any fouling of the secondary filter more noticeable).

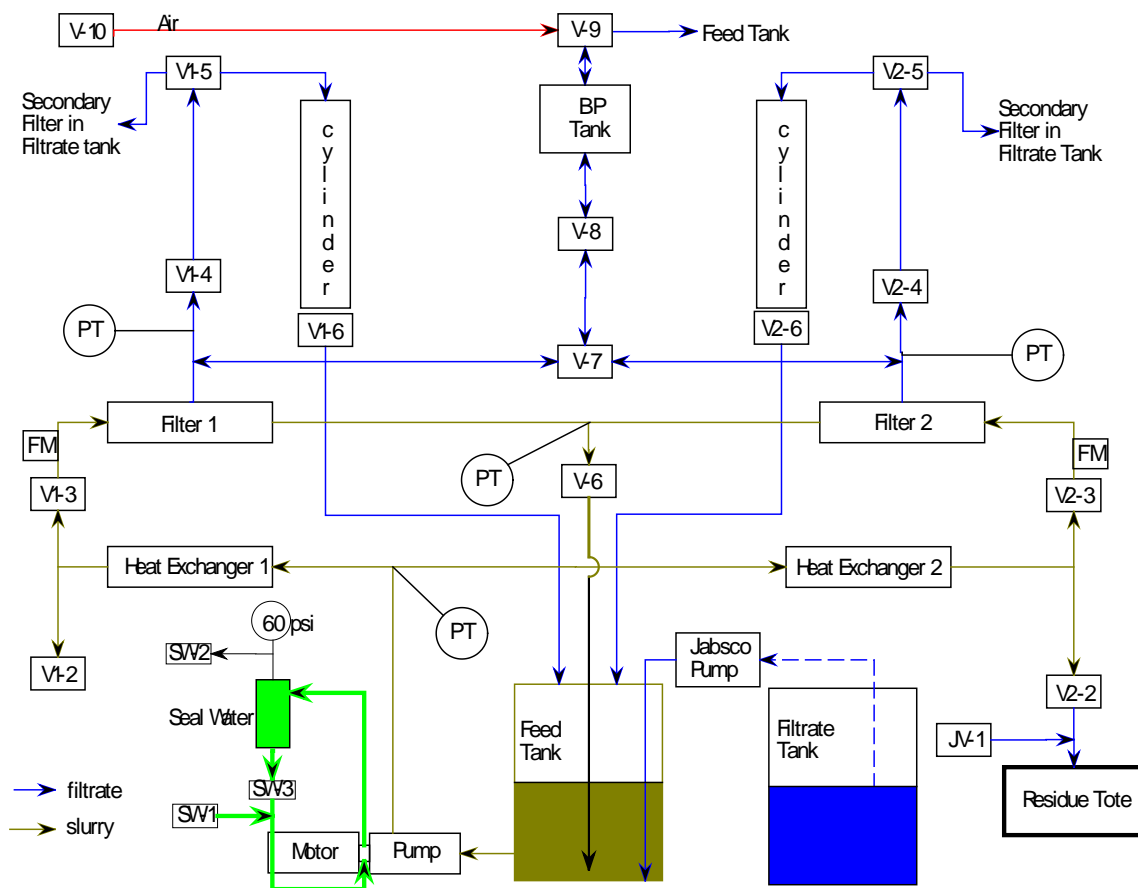


Figure 1. Schematic of Laboratory-Scale Crossflow Filter Unit

2.2 Test Protocol

The MST Filterability crossflow filter tests were conducted as follows. Prior to testing, the apparatus was chemically cleaned with 0.5 M oxalic acid and 1 M nitric acid. Rather than using a prototypic ARP cleaning method, the filters were cleaned by draining the feed slurry from the filter system into the feed tank and removing the feed slurry from the feed tank. After the feed slurry was removed from the system, approximately 3 gallons of 0.5 M oxalic acid was added to the feed tank. The oxalic acid was recirculated through the filter system (on both the feed side and the filtrate side) for at least 1 hour, drained into the feed tank, and removed. After the oxalic acid was removed, approximately 3 gallons of 1 M nitric acid was added to the feed tank. The nitric acid was recirculated through the filter system for at least 1 hour, drained into the feed tank, and removed. The filter system was flushed with deionized water until the pH was greater than 6. All filter cleaning was conducted at ambient temperature. The cleaning protocol was selected to have a clean filter and a comparable starting point for each test rather than to be prototypic. Following chemical cleaning, the filter system was run with deionized water to establish a clean water flux for each of the filters. Table 1 shows the results.

Table 1. Clean Water Flux Prior to Start of Testing

Test	Flux 0.1 micron filter (gpm/ft ²)	Flux 0.5 micron filter (gpm/ft ²)	TMP (psi)
Sodium Oxalate	0.31	0.84	40
Sodium Aluminosilicate	0.54	2.38	40
Gibbsite/Beohmite	0.35	0.80	40

The simulated salt solution was based on the Salt Batch 6 analysis.⁵ Table 2 shows the composition of the salt solution. Additional nitrate was added to balance the charges (3.03 M nitrate versus 2.5 M nitrate in reference 6). The salt solution was prepared by dissolving sodium hydroxide in deionized water, adding aluminum nitrate to react with the sodium hydroxide forming sodium aluminate, and adding the remaining components in order of increasing solubility. The salt solution contains no oxalate or soluble silica. The solution was filtered with the 0.1 and 0.5 micron crossflow filters to remove any precipitated salts prior to testing.

Table 2. Composition of Simulated Salt Solution

<u>Ion</u>	<u>Concentration (M)</u>
Na ⁺	6.6
K ⁺	0.01
OH ⁻	2.22
NO ₃ ⁻	3.03
NO ₂ ⁻	0.51
AlO ₂ ⁻	0.23
CO ₃ ⁻²	0.22
SO ₄ ⁻²	0.071
Cl ⁻	0.0085
PO ₄ ⁻³	0.0045

The slurry was run through the crossflow filters at an axial velocity of 8.7 ± 0.1 ft/s and a feed pressure of 40 ± 2 psid. The axial velocities were kept the same by measuring the volumetric flow rate through each filter and adjusting a valve upstream of each filter to keep them equal. The feed pressures were equal by design. The filter feed and concentrate lines of the two filters were connected to eliminate any differences in pressure between the two filters. Because of the pressure drop across the secondary filter, the transmembrane pressure may differ slightly between the two crossflow filters.

The first set of tests investigated the impact of sodium oxalate on filter flux. Approximately 9 gallons of salt solution was added to the filter feed tank. To this solution, sufficient MST was added to produce a feed containing 2.5 g MST/L slurry. The feed slurry was fed to the filters. For the first hour, the filtrate was recycled to the feed tank. After one hour, the filtrate was collected in a separate tank in order to concentrate the insoluble solids in the feed tank. If the volume of material in the feed tank decreased to three gallons, the filtrate was recycled to the feed tank for the remainder of the day. At the end of the day, the feed pump and agitator were turned off, and the system sat overnight. The filtrate was returned to the feed tank prior to the start of the next test.

For subsequent tests, additional sodium oxalate was added to the feed tank to increase its concentration. The sodium oxalate was dissolved in a 1 M sodium hydroxide solution (for tests 2 and 3) or deionized water (for tests 4 and 5) prior to addition to the feed tank. Table 3 shows the concentration of sodium

oxalate in the feed for each of the tests. The oxalate was a combination of precipitated solids and solution phases.

Table 3. Sodium Oxalate Concentration in Filter Feed

<u>Test</u>	<u>Sodium Oxalate Concentration (M)</u>	<u>Sodium Oxalate Concentration (g/L)</u>
1	0.000	0.000
2	0.001	0.134
3	0.004	0.536
4	0.016	2.14
5	0.064	8.58

Following the tests to investigate the impact of sodium oxalate on the filter flux, the filter system was cleaned with 0.5 M oxalic acid, followed by 1 M nitric acid. Following chemical cleaning, the filter system was flushed with deionized water, and the clean water flux was measured to verify that the filters are clean.

The next set of tests investigated the impact of sodium aluminosilicate on filter flux. The sodium aluminosilicate was prepared by making two solutions, an aluminum solution and a silicon solution. The aluminum solution contained 4 M sodium hydroxide, 0.94 M aluminum nitrate nonahydrate, 2 M sodium nitrate, and 2 M sodium nitrite. The silicon solution contained 4 M sodium hydroxide and 0.75 M metasilicate. These solutions were reacted in a Parr reactor at 80 C for 1.5 hours. The two solutions were mixed together just prior to adding them to the Parr reactor.⁶ Because of the chemistry of sodium aluminosilicate, there is the possibility that precipitation could occur after adding the solids to the filter feed tank or downstream of the crossflow filters.

Approximately 9 gallons of salt solution was added to the CUF feed tank. To this solution, sufficient MST was added to produce a feed containing 2.5 g MST/L slurry. Sodium aluminosilicate was added to the feed tank. The amount added is shown in Table 4. The feed slurry was fed to the filters. The filtration process operated for approximately eight hours. For the first hour, the filtrate was recycled to the feed tank. After one hour, the filtrate was collected in a separate tank in order to concentrate the insoluble solids in the feed tank. If the volume of material in the feed tank decreased to three gallons, the filtrate was recycled to the feed tank for the remainder of the day. At the end of the day, the feed pump and agitator were turned off, and the system sat overnight. The filtrate was returned to the feed tank prior to the start of the next test. For subsequent tests, additional sodium aluminosilicate was added to the feed tank to increase its concentration.

Table 4. Sodium Aluminosilicate Concentration in Filter Feed

<u>Test</u>	<u>Sodium Aluminosilicate (g/L)</u>
1	0.25
2	0.5
3	1.0
4	2.0
5	4.0

Following the tests to investigate the impact of sodium aluminosilicate on the filter flux, the filter system was cleaned with 0.5 M oxalic acid, followed by 1 M nitric acid. Following chemical cleaning, the filter system was flushed with deionized water, and the clean water flux was measured to verify that the filters are clean.

The next set of tests investigated the impact of aluminum solids (e.g., gibbsite and boehmite) on filter flux. Approximately 9 gallons of salt solution were added to the CUF feed tank. To this solution, sufficient MST was added to produce a feed containing 2.5 g MST/L slurry. Aluminum solids were also added to the feed tank as solid particles. The solids were approximately 50% gibbsite and 50% boehmite. The amount added is shown in Table 5. The feed slurry was fed to the filters. The filtration process operated for approximately eight hours. For the first hour, the filtrate was recycled to the feed tank. After one hour, the filtrate was collected in a separate tank in order to concentrate the insoluble solids in the feed tank. If the volume of material in the feed tank decreased to three gallons, the filtrate was recycled to the feed tank for the remainder of the day. At the end of the day, the feed pump and agitator were turned off, and the system sat overnight. The filtrate was returned to the feed tank prior to the start of the next test.

Table 5. Aluminum Solids Concentration in Filter Feed

<u>Test</u>	<u>Aluminum Solids (g/L)</u>
1	0.25
2	0.5
3	1.0
4	2.0
5	4.0

2.3 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

The plan for this testing is described in TTQAP SRNL-RP-2014-00874. The M&TE were calibrated prior to the start of testing. Data collected are recorded in Laboratory Notebooks SRNL-NB-2014-00021, SRNL-NB-2014-00006, and SRNS-NB-2015-00002.

3.0 Results

3.1 Crossflow Filter

Figure 2 shows the crossflow filter flux as a function of time for the 0.1 and 0.5 micron crossflow filters. The oxalate concentration is included on the plot. The plot also shows the filter flux with no sodium oxalate in the feed (baseline). At the start of each day/batch, the flux is higher for the baseline feed than for the oxalate-containing feed. Plausible causes are different effectiveness of cleaning prior to the start of the tests, differences in the prepared salt solutions despite being the same recipe, an effect of the oxalate in the later tests, or an impurity in the feed slurry. The clean water flux prior to the start of the sodium oxalate feed test was 0.31 gpm/ft², so ineffective cleaning is not a likely cause. It is possible that there was an impurity in the salt solution or the feed slurry, but there is no explicit evidence of this. At the end of each day, there is no significant difference in the flux for the oxalate-containing slurry compared to the baseline slurry. At the end of the 5th batch, the flux with the oxalate containing feed is higher than the flux with the baseline feed. The low starting flux was also observed for the test containing no sodium oxalate in the feed. We are not sure of the reason for this occurrence.

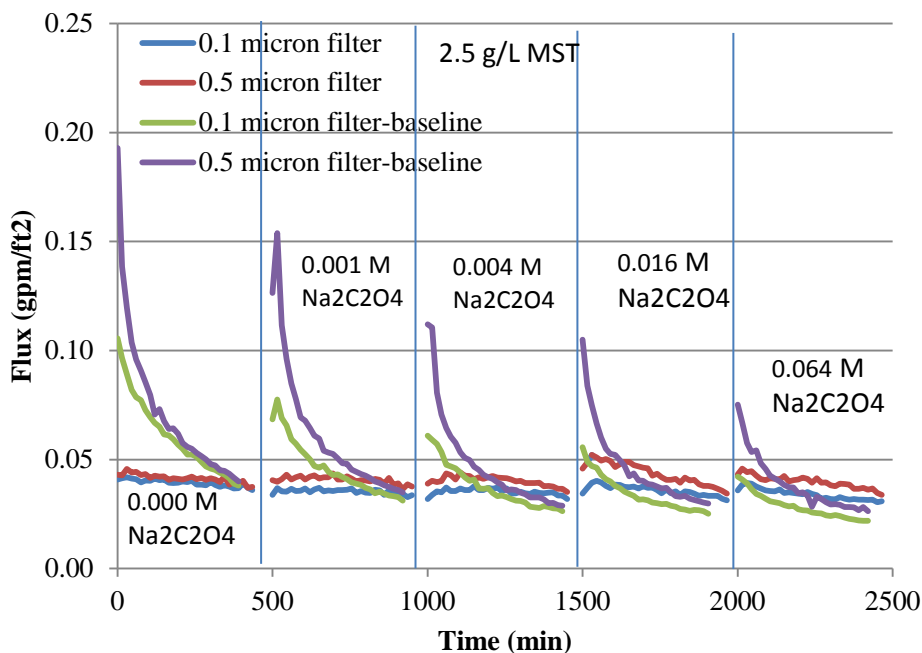


Figure 2. Comparison of filter flux with and without Sodium Oxalate

Figure 3 shows the pressure drop across the crossflow filter normalized by the filter flux. The data show an increase in the normalized pressure drop during each batch (~ 10 – 25%). This result shows some fouling of the crossflow filter, but not as much as was observed with the baseline feed. The 0.1 micron filter shows a higher pressure drop than the 0.5 micron filter.

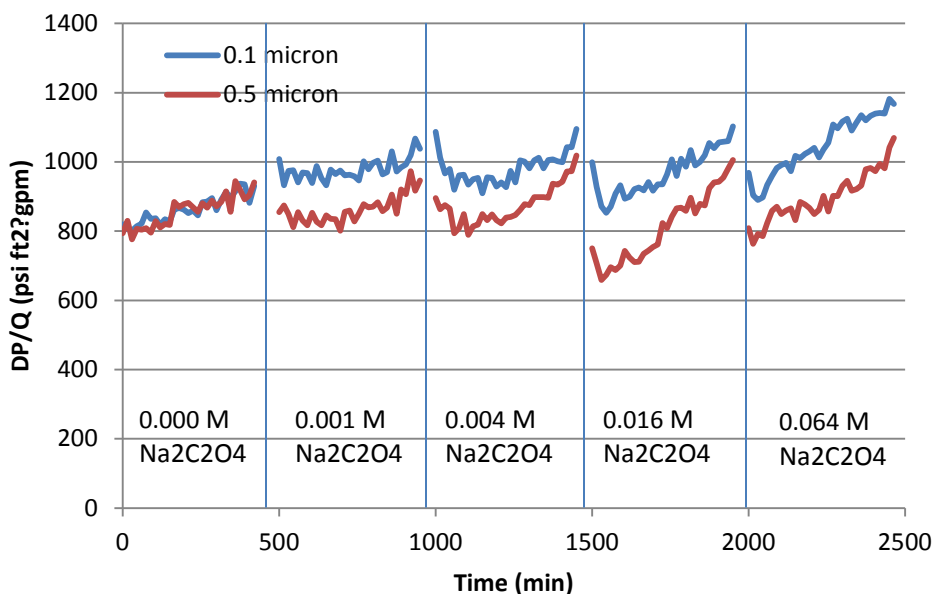


Figure 3. Pressure Drop across Crossflow Filter for Sodium Oxalate Containing Feeds

Figure 4 shows the crossflow filter flux as a function of time for the 0.1 and 0.5 micron crossflow filters for feed slurries containing sodium aluminosilicate. The sodium aluminosilicate concentration is included in the plot. The plot also shows the filter flux with no sodium aluminosilicate in the feed (baseline). There is no significant difference in the flux for the sodium aluminosilicate -containing slurry compared to the baseline slurry.

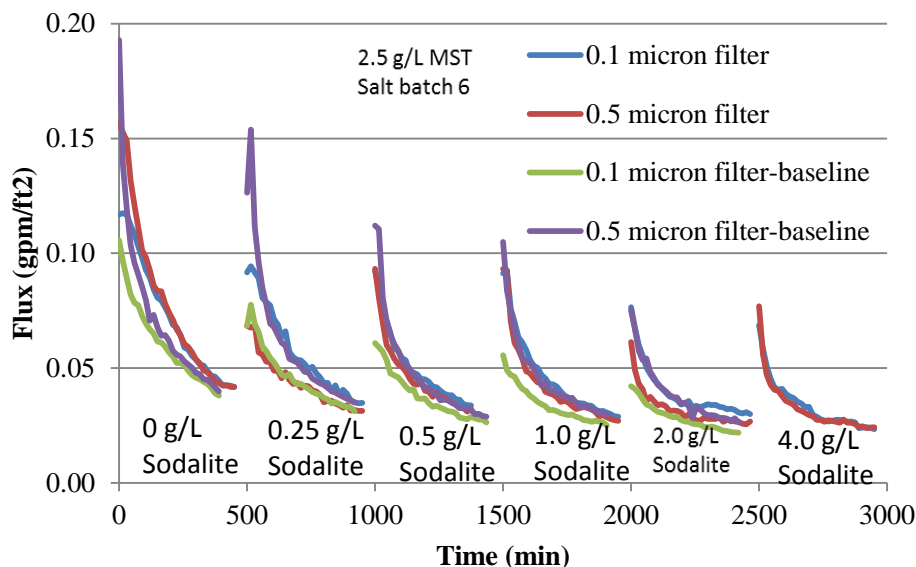


Figure 4. Comparison of filter flux with and without Sodium Aluminosilicate

Figure 5 shows the pressure drop across the crossflow filter for sodium aluminosilicate feeds normalized by the crossflow filter flux. The data show a significant increase in pressure drop during each batch (~4X). This increase is much larger than the increase observed with the sodium oxalate containing feed. In addition, the pressure drop across the 0.5 micron filter is higher than the pressure drop across the 0.1 micron filter, but the difference may not be significant. This is different from the sodium oxalate containing feed, where the normalized pressure drop was larger across the 0.1 micron filter.

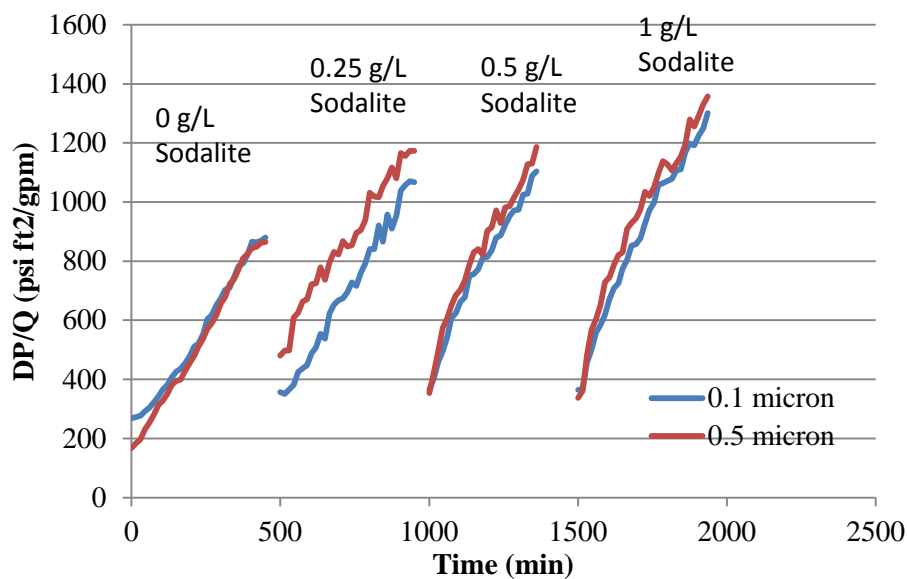


Figure 5. Pressure Drop across Crossflow Filter for Sodium Aluminosilicate Containing Feeds

Figure 6 shows the crossflow filter flux as a function of time for the 0.1 and 0.5 micron crossflow filters for feeds containing aluminum solids (gibbsite and boehmite). The aluminum concentration is included in the plot. The plot also shows the filter flux with no aluminum in the feed (baseline). There is no significant difference in the flux for the gibbsite/boehmite -containing slurry compared to the baseline slurry.

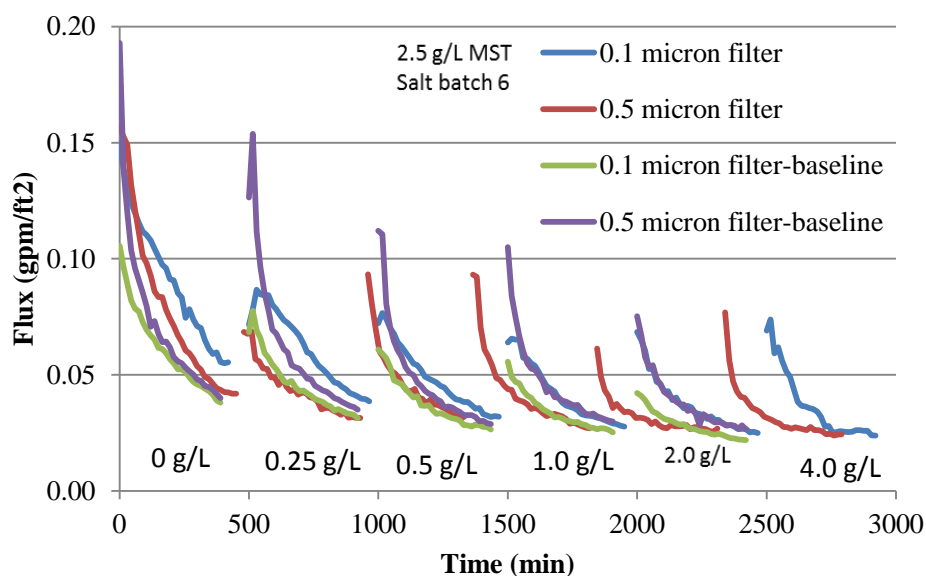


Figure 6. Comparison of filter flux with and without Gibbsite/Boehmite Solids

Figure 7 shows the normalized pressure drop across the crossflow filter with gibbsite/boehmite containing feed. The data show a significant increase in pressure drop during each batch (~ 3 - 4X). This increase is much larger than the increase observed with the sodium oxalate containing feed, but comparable to the increase observed with the sodium aluminosilicate containing feed. The 0.1 and 0.5 micron filters have approximately the same pressure drop. In addition, the slope of the normalized pressure drop appears to be increasing with each batch. The authors are not sure if this increase is statistically significant.

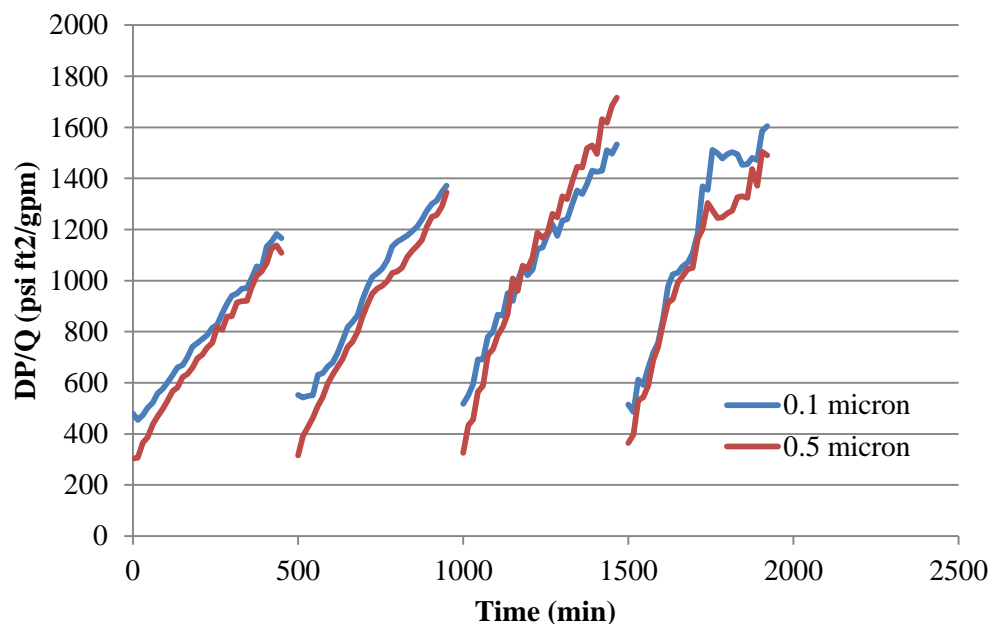


Figure 7. Pressure Drop across Crossflow Filter for Gibbsite/Boehmite Containing Feeds

3.2 Secondary Filter

Figure 8 shows the pressure drop across the secondary filter divided by the crossflow filter flux for a baseline feed containing 2.5 M NaOH, 0.5 M NaNO₂, 3.4 M NaNO₃, and varying concentrations of MST. The pressure drop divided by the crossflow filter flux ranges between 60 and 90 psi/(gpm/ft²), and is trending downward. No increase in pressure drop with increasing MST concentration is observed. The data indicate that the crossflow filters are effectively removing the MST, and no fouling of the secondary filter is occurring.

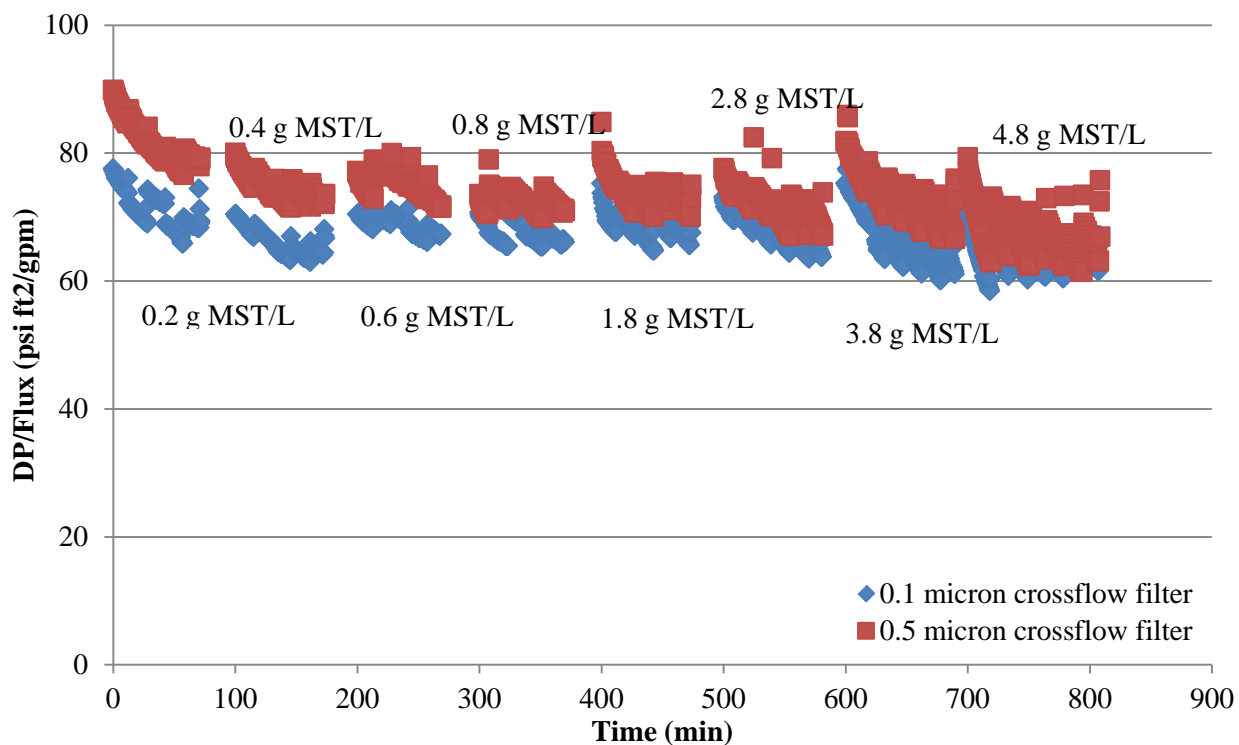


Figure 8. Pressure Drop across Secondary Filter for Baseline Feed

Figure 9 shows the pressure drop across the secondary filter divided by the crossflow filter flux for each of the secondary filters with sodium oxalate containing feed. The normalized pressure drop is larger than the pressure drop observed in the baseline test. The data for the 0.5 micron crossflow filter are very noisy on the first day. This is due to a lot of noise in the pressure data, and the authors are not sure of the cause. The curve for the 0.1 micron filter is horizontal with some small variability. There is some variability from day to day, but it is not significant. The data indicate no significant fouling of the secondary filter down stream of the 0.1 micron crossflow filter.

The curve for the 0.5 micron filter is horizontal with some small variability. There is some variability from day to day, but it is not significant. The data indicate no significant fouling of the secondary filter down stream of the 0.5 micron crossflow filter.

The data do show a larger pressure drop across the secondary filter down stream of the 0.1 micron crossflow filter than across the secondary filter downstream of the 0.5 micron crossflow filter. The difference is on the order of 20 – 25%. This difference could indicate some type of fouling of the secondary filter downstream of the 0.1 micron crossflow filter. However, it could be due to differences in the secondary filters. The secondary filters were used 0.5 micron Mott filter media previously used in rotary microfilter testing. The prior use could have created differences between the filter media.

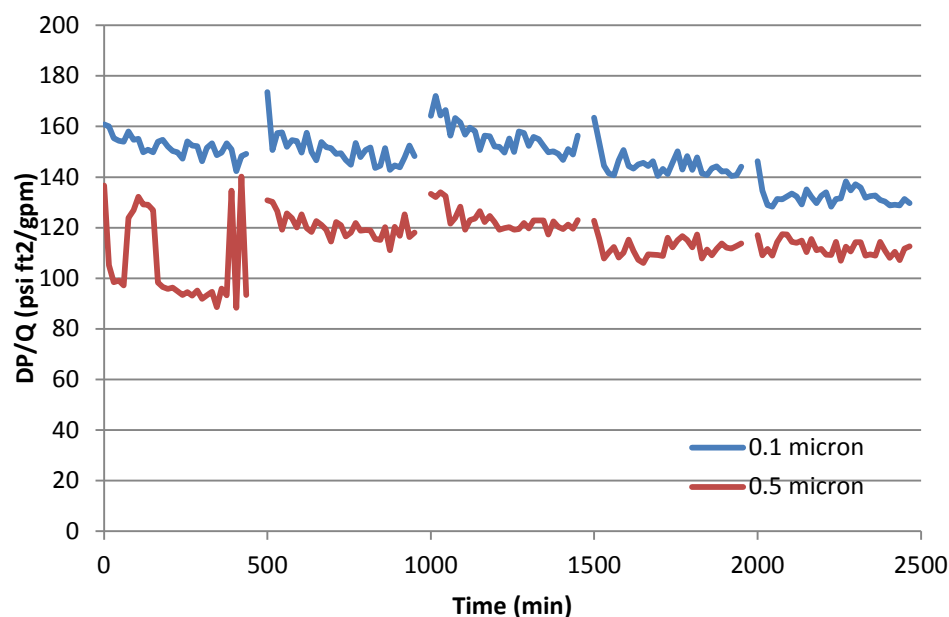


Figure 9. Secondary Filter Pressure Drop for Oxalate-Containing-Feed

Figure 10 shows the pressure drop across the secondary filter divided by the crossflow filter flux for each of the filter combinations with sodium aluminosilicate-containing feed. The pressure drop is comparable to the pressure drop in the baseline test. The data for the 0.1 micron filter show an increase in filter pressure drop as a function of time. This increase could be an indication of fouling of the secondary filter. However, the data does not show an increase from day to day.

The data for the 0.5 micron filter show an increase in filter pressure drop as a function of time. This increase could be an indication of fouling of the secondary filter. However, the data do show an increase from day 1 to day 2, but not between days 2, 3, and 4. The data show some recovery of pressure drop between batches. The recovery could be from solid particles diffusing away from the secondary filter as it sat undisturbed in filtrate overnight. This recovery of pressure drop across the secondary filter has been observed in the ARP.

The data do show a larger pressure drop across the secondary filter down stream of the 0.5 micron crossflow filter than across the secondary filter downstream of the 0.1 micron crossflow filter. This is the opposite of the oxalate-containing feed data. The difference is on the order of 20 %. This difference could indicate some type of fouling of the secondary filter downstream of the 0.5 micron crossflow filter. The 0.5 micron filter could allow fine particles to pass that are rejected by the 0.1 micron crossflow filter. However, it could be due to differences in the secondary filters. The secondary filters were used 0.5 micron Mott filter media previously used in rotary microfilter testing. The prior use could have created differences between the filter media.

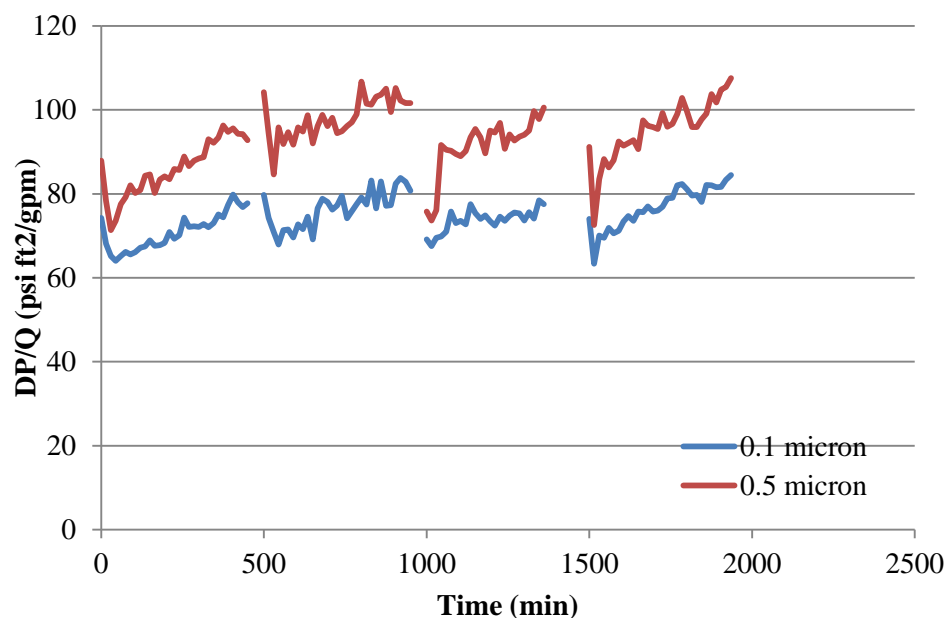


Figure 10. Secondary Filter Pressure Drop for Sodium Aluminosilicate-Containing-Feed

Figure 11 shows the pressure drop across the secondary filter divided by the crossflow filter flux for each of the filter combinations with gibbsite/boehmite-containing feed. The pressure drop is comparable to the pressure drop in the baseline test. The data for the 0.1 micron filter show an increase in filter pressure drop as a function of time. This increase could be an indication of fouling of the secondary filter. However, the data does not show an increase from day to day.

The data for the 0.5 micron filter show an increase in filter pressure drop as a function of time. This increase could be an indication of fouling of the secondary filter. However, the data does not show an increase from day to day.

The data do show a larger pressure drop across the secondary filter down stream of the 0.5 micron crossflow filter than across the secondary filter downstream of the 0.1 micron crossflow filter. This is the opposite of the oxalate-containing feed data. The difference is on the order of 15 - 20 %. This difference could indicate some type of fouling of the secondary filter downstream of the 0.5 micron crossflow filter. It could also indicate that fine particles are passing the 0.5 micron filter, but not the 0.1 micron filter. However, it could be due to differences in the secondary filters. The secondary filters were used 0.5 micron Mott filter media previously used in rotary microfilter testing. The prior use could have created differences between the filter media.

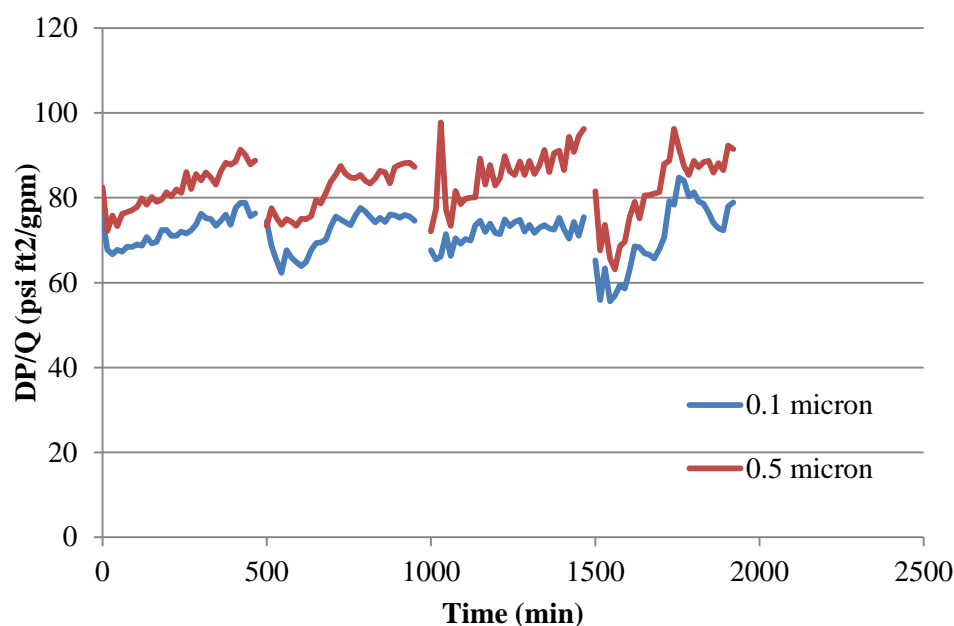


Figure 11. Secondary Filter Pressure Drop for Aluminum-Containing-Feed

4.0 Discussion

The oxalate-containing feeds show the same crossflow filter flux as the baseline feeds at the end of each day. However, at the beginning of each day, the oxalate-containing feed flux was much lower than the baseline feed flux, even in the case where there was no oxalate in the feed. This effect could be from differences in the effectiveness of filter cleaning prior to the start of the tests, differences in the prepared salt solutions (even though they were prepared to the same recipe), or an effect of the oxalate (in tests

after it was added). Given that the clean water flux was 0.31 gpm/ft² prior to the start of the test, ineffective cleaning is not a likely cause. Another possible cause is an impurity in the feed. The 0.5 micron filter showed higher flux than the 0.1 micron filter. A fine impurity could be trapped by the 0.1 micron filter, but “squeezed” through the 0.5 micron filter. There is insufficient data to draw a definitive conclusion at this time.

The feeds containing sodium aluminosilicate and gibbsite/boehmite solids show no difference in crossflow filter flux when compared to the baseline feed slurry. This result indicates no additional crossflow filter fouling occurred due to the addition of sodium aluminosilicate or aluminum solids.

The oxalate containing feeds showed no indication of secondary filter fouling. When normalizing the pressure drop across the secondary filter by dividing it by the crossflow filter flux, the normalized pressure drop is approximately constant. However, the pressure drop across the secondary filter in the tests with sodium oxalate containing feeds is larger than the pressure drop across the secondary filter in the baseline tests, indicating that some fouling may have occurred.

The sodium aluminosilicate and gibbsite/boehmite containing feeds showed an indication of secondary filter fouling. When normalizing the pressure drop across the secondary filter by dividing it by the crossflow filter flux, the normalized pressure drop increased during the tests each day. This increase could be from solid particles forming between the filters, solid particles dissolving and reprecipitating, or fine particles passing through the filter. The secondary filter pressure drop in the baseline tests showed a decrease each day. However, the secondary filter pressure drop was comparable for the baseline test, sodium aluminosilicate test, and the gibbsite/boehmite test.

The pressure drop across the secondary filter was larger downstream of the 0.5 micron crossflow filter than downstream of the 0.1 micron crossflow filter with sodium aluminosilicate containing feeds. This result suggests that fine particles may have passed through the 0.5 micron filter, but been rejected by the 0.1 micron filter.

The recovery of filter flux at the start of each batch was different when the feed contained sodium oxalate compared to when the feed contained sodium aluminosilicate or gibbsite/boehmite. One plausible reason for the difference is that sodium oxalate is an organic compound and sodium aluminosilicate, gibbsite, and boehmite are inorganic minerals.

All of the tests showed some evidence of solid particles passing the crossflow filter and fouling the secondary filter. An alternative explanation could be that soluble aluminate precipitated downstream of the crossflow filter. Visual inspection of the secondary filter following the tests did not show any evidence of solid accumulation on the secondary filter surface.

5.0 Conclusions

The conclusions from this work follow.

- Testing with feed slurries containing sodium aluminosilicate and aluminum (gibbsite and boehmite)-containing solids gave no indication of a significant change in crossflow filter fouling compared to feeds containing MST only.
- While the sodium oxalate containing feeds had the same final crossflow filter flux as the baseline feed, the initial crossflow filter flux was significantly lower than the initial baseline crossflow filter flux. This difference could be from the presence of sodium oxalate or from other causes.

- The sodium oxalate containing feed showed no change in pressure drop of the secondary filter, but the pressure drop was larger than measured in the baseline feed, indicating that some fouling may have occurred.
- The sodium aluminosilicate and aluminum solids containing feeds showed evidence of secondary filter fouling. However, the pressure drop was comparable to the pressure drop in the baseline test.
- All of the tests showed some evidence of fouling the secondary filter. This fouling could be from fine particles passing through the crossflow filter or from soluble aluminate precipitating.
- The sodium oxalate containing feeds behaved differently than the sodium aluminosilicate and gibbsite/boehmite containing feeds.

6.0 References

1. Martino, C.J., Herman, D.T., Pike, J.A., and Peters, T.B., "Actinide Removal Process Sample Analysis, Chemical Modeling, and Filtration Evaluation," Doc No. SRNL-STI-2013-00700, Rev 0, June 2014.
2. W. R. Wilmarth, S. D. Fink, D. T. Hobbs, and M. S. Hay, "Characterization and Dissolution Studies of Samples from the 242-16H Evaporator Gravity Drain Line", WSRC-TR-97-0326, October 16, 1997.
3. P. J. Bhatt, "Analyze Effect of Agitation on 512-S Crossflow Filter Performance", X-TTR-S-00021, July 29, 2014.
4. M. R. Poirier and P. R. Burket, "Task Technical and Quality Assurance Plan for SRNL Filter Testing to Support ARP", SRNL-RP-2014-00874, August 28, 2014.
5. S. E. Campbell, "Blend Evaluation for Tank 49 Feed for ISDP Salt Batch 6-D", X-ESR-H-00501, April 23, 2013.
6. J. Addai-Mensah, J. Li, M. Zbik, and S. Rosencrance, "The Chemistry, Crystallization, Physicochemical Properties and Behavior of Sodium Aluminosilicate Solid liquids: Final Report", WSRC-MS-2002-00907, November 20, 2002.

Distribution:

D. E. Dooley, 999-W
A. Fellingner, 773-42A
T. B. Brown, 773-A
M. E. Stone, 999-W
S. D. Fink, 773-A
C. C. Herman, 773-A
E. N. Hoffman, 999-W
F. M. Pennebaker, 773-42A
W. R. Wilmarth, 773-A
Records Administration (EDWS)

E. A. Brass, 249-8H
C. K. Chiu, 704-27S
E. J. Freed, 704-S
A. G. Garrison, 248-9H
B. A. Gifford, 704-56H
K. L. Lang, 707-7E
D. J. Martin, 241-152H
A. R. Shafer, 704-27S

P. R. Jackson, DOE-SR, 703-46A
J. A. Crenshaw, 703-46A