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Pilot-Scale Testing of a 0.1 Micron Filter with SRS Simulated High Level Waste

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SUMMARY

The Savannah River Site selected caustic side solvent extraction (CSSX) as the preferred treatment technology for SRS High Level Waste. As a pretreatment step for the CSSX process, the facility will contact the incoming salt solution, which contains entrained sludge, with monosodium titanate (MST) to adsorb strontium and selected alpha-emitting radionuclides. The process then filters the resulting slurry to remove the sludge and MST. The baseline filter technology uses a 0.1 µ Mott pore-size crossflow filter based on limited data from laboratory scale experiments. The Actinide Removal Process, housed in Building 512-S and proposed for Building 241-96H, involves the identical filtration challenge. The existing equipment in these locations consists of 0.5 µ Mott pore-size filters.

Laboratory scale testing conducted by SRTC in 2002 showed that a 0.1 µ filter could produce the same, and in some cases higher, flux as a 0.5 µ filter. In addition, the baseline processes concentrate the feed slurry to 5 wt %. If the processes could concentrate the waste to a higher solids loading, a lower volume of water would transfer to the Defense Waste Processing Facility.

Savannah River Technology Center (SRTC) and University of South Carolina (USC) personnel conducted engineering-scale filtration tests using the Filtration Research Engineering Demonstration (FRED) facility. The tests used a 0.1 µ Mott crossflow filter and operated the filter with feed slurries containing up to 12 wt % insoluble solids.

The conclusions from this work follow.

- The 0.1 micron filter produced a higher flux than observed in comparable tests with the 0.5 micron filter at 0.06 wt % solids and 4.5 wt % solids.
- The average filter flux equaled 0.132 gpm/ft^2 at 0.06 wt % solids, 0.069 gpm/ft^2 at 4.5 wt % solids, and 0.026 gpm/ft^2 at 12.2 wt % solids.
- The flux for the 12.2 wt % slurry exceeded the design bases for the processes.
- Filter flux demonstrated a statistically significant correlation with axial velocity, with increasing axial velocity producing higher filter flux for 0.06, 4.5, and 12.2 wt % slurries.
- Filter flux demonstrated a statistically significant correlation with transmembrane pressure (TMP), with increasing TMP producing higher filter flux for 0.06 and 12.2 wt % slurries. We did not observe a statistically significant correlation between transmembrane pressure and filter flux for the 4.5 wt % slurry.
- The cooling rate needed to maintain 35 ± 3 ºC with 12.2 wt % slurry measured 17,600 – 20,100 BTU/hr.

INTRODUCTION

The Savannah River Site selected caustic side solvent extraction as the preferred treatment technology for SRS High Level Waste. As a pretreatment step for the CSSX process, the facility will contact the incoming salt solution, which contains entrained sludge, with MST to adsorb strontium and selected alpha-emitting radionuclides. The process then filters the resulting slurry to remove the sludge and MST. The process then removes cesium from the filtrate through the solvent extraction system. The Actinide Removal Process used for treatment of Low Curie Salt waste involves the identical sorption and filtration process.

For the SWPF, the baseline filter technology uses a 0.1 µ Mott crossflow filter, while the Actinide Removal Process uses existing 0.5 µ Mott crossflow filters. Lab scale testing conducted by SRTC in 2002 showed that a 0.1 µ filter could produce the same, and in some cases higher, flux as a 0.5 µ filter.[1] These findings led to a change in the pore size of the filter for the SWPF.[2] In addition, the baseline processes concentrate the feed slurry to 5 wt %. If the processes could concentrate the waste to a higher solids loading, a lower volume of water would transfer to the Defense Waste Processing Facility.

SRTC and USC personnel conducted engineering-scale filtration tests using the FRED facility. They conducted the filtration tests with a simulated SRS high level waste solution containing 5.6 M sodium, average salt solution, and varying concentrations of monosodium titanate and simulated sludge. The tests used a 0.1 µ Mott crossflow filter and operated the filter with feed slurries containing up to 12 wt % insoluble solids.

TESTING

Equipment

The testing occurred at the FRED facility shown in Figure 1. The FRED facility contains a filter element with seven Mott filter tubes. Each tube is made from sintered stainless steel, 0.75 inches OD, 0.625 inches ID, 10 feet long, and nominal 0.1 micron pore size. The slurry feed tank holds a maximum of 550 gallons. The filter feed pump is a long throw helical impeller pump, which has a maximum flow rate of 225 gpm with water and 175 gpm with SRS simulated waste. The filtrate can be recycled back to the feed tank or removed. The facility uses process water and a heat exchanger to control the temperature of the feed slurry to 35 ± 3 ºC. The filter can be back-pulsed with nitrogen. The backpulse pressure was set to 120 psi during this test.

Test Protocol

Table 1 describes the feed slurry used in this test. Personnel added sufficient sludge to obtain a sludge concentration of 0.0313 wt %. Personnel used sludge never used in previous filter testing to ensure presence of very fine particulates that would maximize the filtration challenge. They suspended the material in the tank, for 2 hours, with normal agitation and measured particle size using the Lasentec® probe. They added sufficient MST (lot # BSC-265-0107) to obtain an MST concentration of 0.0287 wt %. This solids loading allows comparison with previous test data. Personnel used MST never used in previous filter testing. Particle size analysis performed on material from this lot number showed a median particle size of 5 micron.[4] SRTC testing showed MST from this lot produced a strontium decontamination factor of 102 ± 23.[5] Personnel suspended the material in the tank, for 2 hours, with normal agitation and measured particle size using the Lasentec probe.

Figure 1. Filtration Research Engineering Demonstration

Table 1. Feed Composition

Species	Concentration (M)
Na (M)	5.6
K (M)	0.015
Cs (M)	0.00014
OH (M)	1.91
NO_3 (M)	2.14
NO_2 (M)	0.52
AlO_2 (M)	0.31
CO_3 (M)	0.16
SO_4 (M)	0.15
Cl (M)	0.025
F (M)	0.032
PO_4 (M)	0.01
C_2O_4 (M)	0.004
SiO_3 (M)	0.004
MoO_4 (M)	0.0002
Tributyl Phosphate (M)	0.0005
Dibutyl Phosphate (M)	0.0250
Monobutyl Phosphate (M)	0.0250
n-butanol (M)	0.0020
Sodium Formate (M)	0.022
MST (wt %)	0.0287 – 5.84
Sludge (wt %)	0.0313 – 6.36
Total Insoluble Solids (wt %)	0.06 – 12.2

Personnel performed filter tests using the conditions described in Table 2. They backpulsed the filter prior to the start of each test. At each test condition, they measured the average filter flux every two minutes. When five consecutive flux readings did not vary more than 5%, they considered the system at steady state. Once the filter flux reached steady state, personnel recorded the axial velocity, transmembrane pressure, and filtrate flow rate every two minutes over a 60 minute period. They then adjusted the filter system to the next test condition and allowed the filter to reach a new steady state.

Table 2. Filter Test Conditions

Test	Axial Velocity (ft/s)	TMP (psi)
1	9	30
2	12	40
3	4	30
4	9	15
5	12	20
6	9	30
7	6	40
8	9	45
9	14	30
10	6	20
11	9	30
12	9	40

In tests 1, 3, and 11 at the lowest solids loading (0.051 wt %), the filter flux did not reach steady state within one hour. In those tests, personnel recorded the filtrate flow rate for the next 60 minutes, and proceeded to the next test condition. The most likely cause of this long time to reach steady-state is the low solids loading in the feed. With a low solids loading, it takes longer for sufficient solids to reach the surface and form the steady-state filter cake.

After completing the test with the 0.051 wt % solids slurry, personnel added sufficient sludge to obtain a sludge concentration of 2.35 wt %. Personnel used sludge never used in previous filter testing. They also added sufficient MST to obtain an MST concentration of 2.15 wt %, reaching a total slurry concentration of 4.5 wt %. They used MST never used in previous filter testing. Personnel performed additional filter tests using the conditions described in Table 2. They backpulsed the filter prior to the start of each test.

After completing the test with 4.5 wt % solids slurry, personnel added sufficient sludge and MST (in a 6:5.5 ratio) to obtain a solids loading of 5.55 wt%. They collected filtration data for two hours at this loading. They reduced the feed volume to increase the solids loading to 9.9 wt %, and collected data for two hours. They added additional sludge and MST to increase the solids loading to 10.8 wt % and collected data for two hours. They performed a volume reduction to produce a 12.2 wt % feed slurry. Personnel continued to operate the filter for seven days, recording operating parameters during this time. The operating conditions during days 1-3 and 6-7 were 9 ft/s axial velocity and 40 psi transmembrane pressure. After seven days, personnel added filtrate to the feed tank to reduce the solids loading to 7.7 wt % and collected data for approximately one hour.

During days 4 and 5, personnel performed filter matrix tests at 12.2 wt % using the test conditions in Table 2. They backpulsed the filter prior to the start of each test condition and collected data for four hours at each condition.

At the highest solids loading (12.2 wt %), FRED personnel measured the flow rate and temperature of the cooling water to calculate the heat transfer rate needed to maintain the test slurry at 35 ± 3 ºC. This data allows one to calculate the heat transfer requirements for more concentrated slurries.

RESULTS

Filter Flux Data

Figure 2 shows the filter flux plotted as a function of time during the test. The plot also shows the solids loading during each period of operation. The initial flux with the 0.051 wt % slurry varied between 0.10 and 0.18 gpm/ft^2. The measured average flux at 0.051 wt % solids equaled 0.13 gpm/ft^2. This flux exceeds that observed in previous testing with a 0.5 micron filter (0.02 – 0.14 gpm/ft^2).[2] The flux with the 4.5 wt % slurry varied between 0.03 and 0.11 gpm/ft^2 (0.069 gpm/ft^2 average), which is higher than the flux measured in previous testing with a 0.5 micron filter (0.00 – 0.06 gpm/ft^2).[3] The average measured flux with 12.2 wt % slurry equaled 0.026 gpm/ft^2 which exceeds the baseline flux of 0.02 gpm/ft^2 for the 0.5 micron filter, which was only tested to a solids loading of 4.5 wt %.

During the test with 12.2 wt % slurry, a measurable filter flux was not detectable in several instances (< 0.0045 gpm/ft^2). Personnel restored filtrate flow in the first case by backpulsing the filter. The other not detectable values occurred at low axial velocity and transmembrane pressure test conditions (4 ft/s and 30 psi, 6 ft/s and 39 psi, and 6 ft/s and 19 psi). These three conditions had the lowest axial velocity tested.

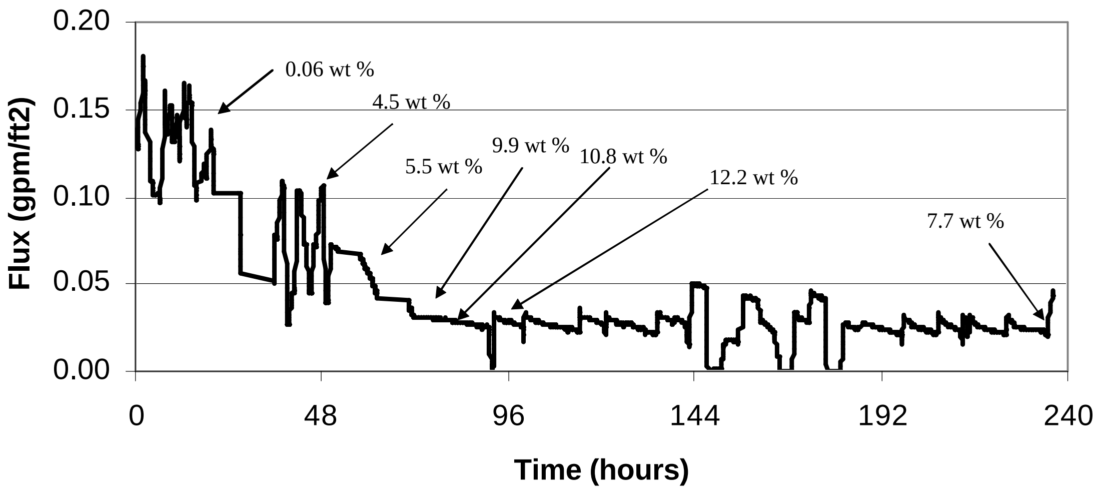

Figure 2. Filter Flux during 0.1 Micron Filter Test

Figures 3 and 4 show the filter flux as a function of transmembrane pressure and axial velocity during the matrix tests performed with 0.051, 4.5, and 12.2 wt % insoluble solids. The data points shown are the average of 30 points collected over one hour. The numbers on the plot show the order in which the tests were performed, with the numbers corresponding to test conditions in Table 2. Performing a statistical analysis of the data with the JMP software shows a statistically significant correlation exists between axial velocity and filter flux at all solids loadings (see APPENDIX A for statistical data). At 0.051 and 12.2 wt %, there is a statistical correlation between TMP and filter flux. No statistically significant correlation was observed between run order and filter flux.

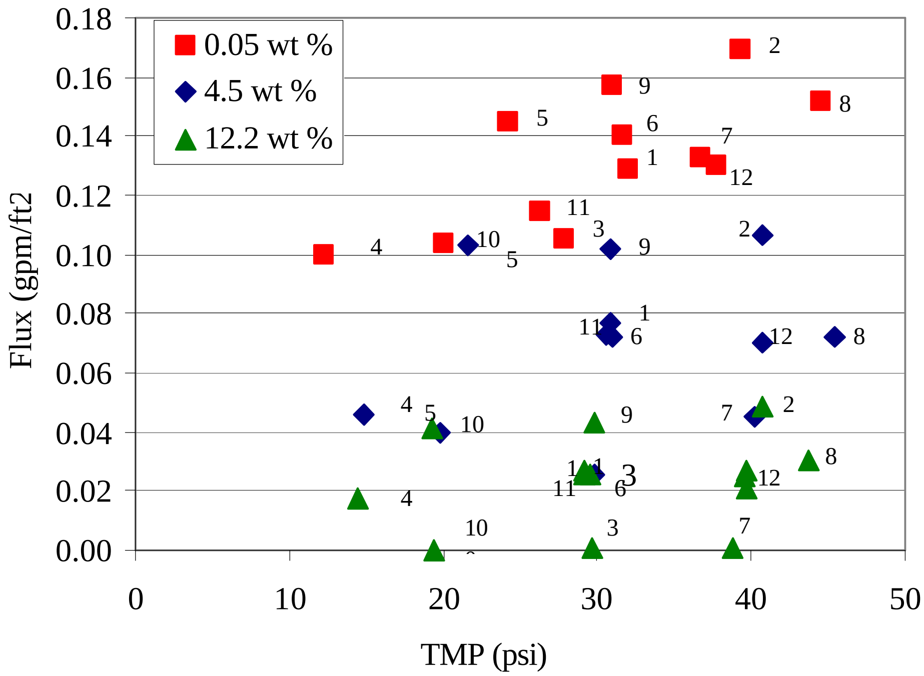

Figure 3. Filter Flux as a Function of TMP with a 0.1 Micron Filter (The numbers refer to the sequential order of the tests with operating conditions as specified in Table 2)

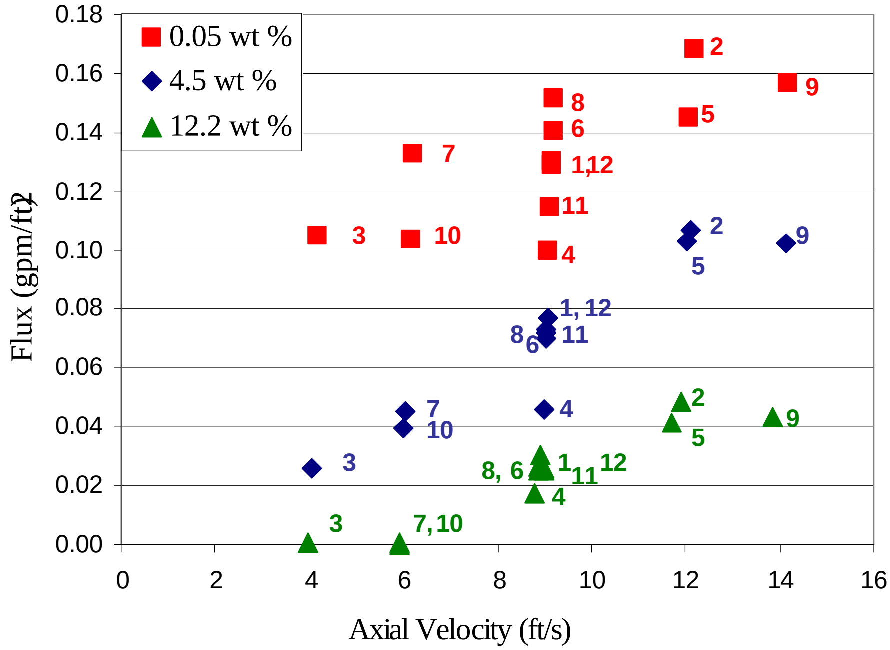

Figure 4. Filter Flux as a Function of Axial Velocity with a 0.1 Micron Filter (The numbers refer to the sequential order of the tests with operating conditions as specified in Table 2)

Figures 5 and 6 compare filter performance of the 0.1 micron filter with the 0.5 micron filter at 0.06 wt % and 4.5 wt %.[3] The 0.1 micron filter produced higher flux than the 0.5 micron filter at the same transmembrane pressure for both solids loadings. This result appears to contradict classical filtration theory, which says larger pore size filters produce higher filter flux. However, this behavior agrees with the earlier findings at lab scale using actual waste.[1] One likely reason for this result is that the smaller pore size filter allows fewer fine particles to become trapped in the pores, limiting the overall resistance to flux.

One significant difference exists between the tests with the different pore size filters. The 0.5 micron filter tests were conducted with an axial velocity of 12 – 26 ft/s, while the 0.1 micron filter tests were performed with an axial velocity of 4 – 14 ft/s.[3] Previous filtration work has shown filter flux increases or remains the same with increasing axial velocity.[6] Therefore, if the 0.5 micron filter tests had been performed at 4 – 14 ft/s axial velocity, the improvement from the 0.1 micron filter would have been the same or greater.

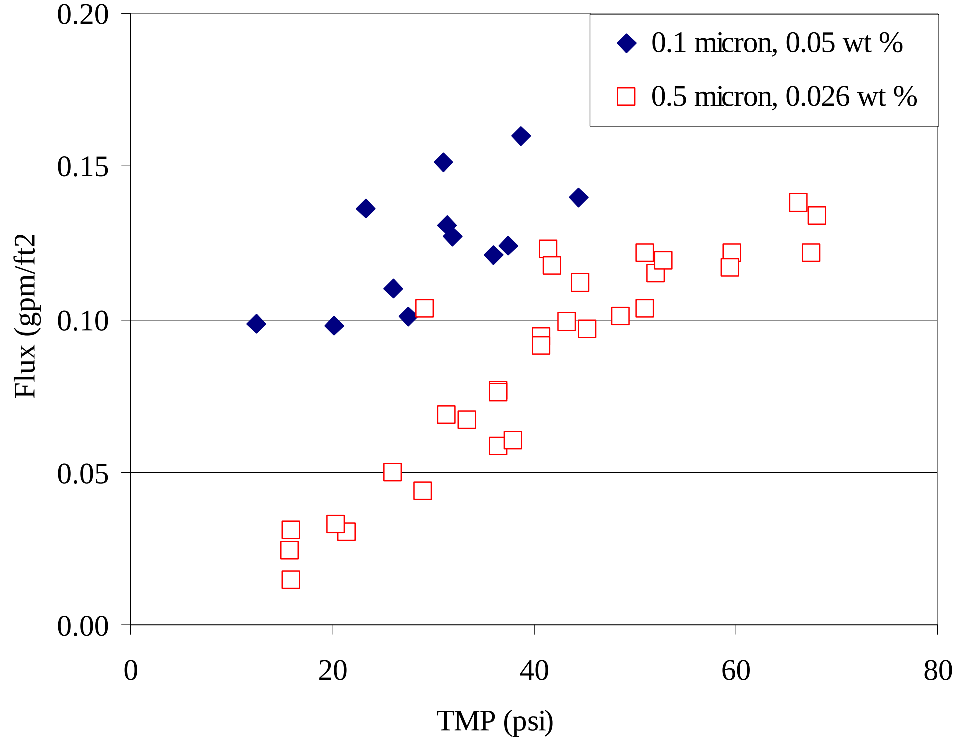

Figure 5. Filter Flux of Mott Filters at 0.06 wt % Insoluble Solids

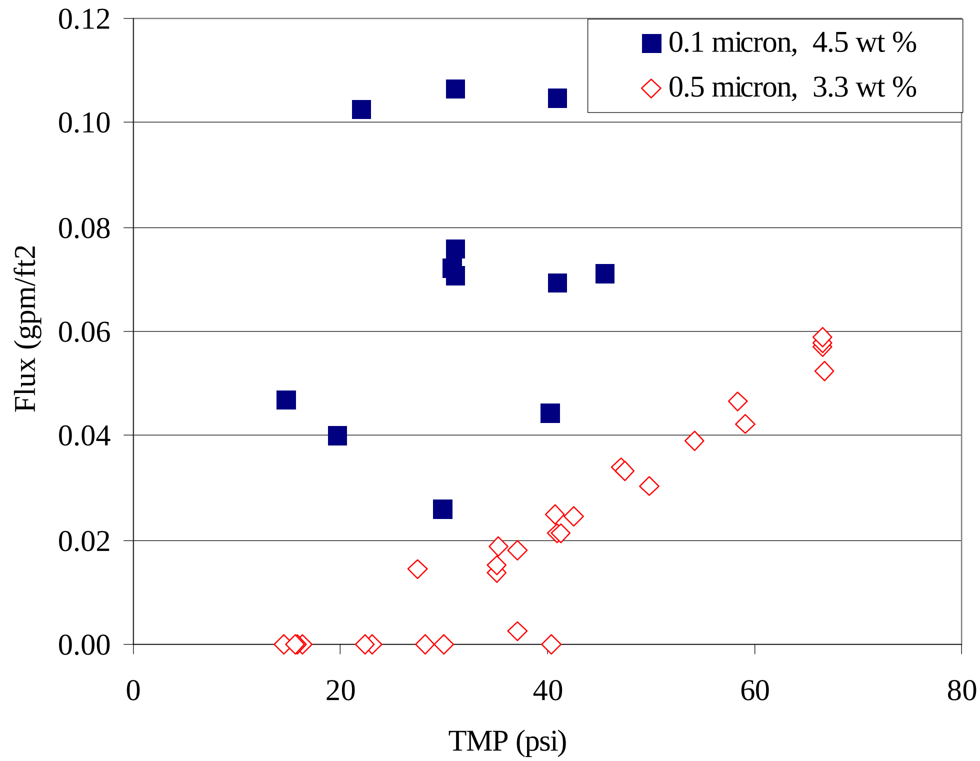

Figure 6. Filter Flux of Mott Filters at 4.5 wt % Insoluble Solids

Table 3 summarizes the filter flux measurements from this test, and compares them, where possible, with data from the 0.5 micron filter test.

Table 3. Average Filter Flux as a Function of Solids Loading

Insoluble Solids (wt %0	0.1 micron Filter	0.5 micron Filter
0.05	0.133	0.086
4.5	0.069	0.022
5.5	0.057	
9.9	0.034	
10.8	0.031	
12.2	0.026	

At the highest solids loading (12.2 wt %), FRED personnel measured the flow rate and temperature of the cooling water to calculate the heat transfer rate needed to maintain the test slurry at 35 ± 3 ºC. In one measurement, the cooling water flow rate measured 19.6 gpm, and the temperature change measured 1.0 ºC. The calculated heat transfer rate was 17,600 BTU/hr. In another measurement, the cooling water flow rate measured 20.1 gpm, and the temperature change measured 1.1 ºC. The calculated heat transfer rate was 20,100 BTU/hr.

Particle Size Data

Personnel collected particle measurements with a Focused Beam Reflectance Measurement (FBRM) probe (Lasentec®). The probe works in the following manner. Personnel installed the probe in the feed tank. The laser beam projects through the window of the FBRM probe and focuses just outside the window surface. This focused beam follows a path around the circumference of the probe window. As particles pass by the window surface, the focused beam will intersect the edge of a particle. The particle will backscatter laser light. The particle will continue to backscatter the light until the focused beam reaches the opposite edge of the particle. The instrument collects the backscattered light and converts it into an electronic signal.

The FBRM isolates the time of backscatter from one edge of an individual particle to its opposite edge. The software records the product of the time multiplied by the scan speed as a chord length. A chord length is a straight line between any two points on the edge of a particle or particle structure (agglomerate). FBRM typically measures tens of thousands of chords per second, resulting in a robust number-by-chord-length distribution.

The chord-length distribution provides a means of tracking changes in both particle dimension and particle population. The calculations do not assume a particle shape. The chord-length distribution is essentially unique for any given particle size and shape distribution. Assuming the average particle shape remains constant over millions of particles, changes to the chord-length distribution reflect solely a function of the change in particle dimension and particle number.

Figure 12 shows data collected from the FBRM. The median particle size following sludge addition (at 0.05 wt %) is 11 micron. Following MST addition (at 0.05 wt %), the median particle size increased to 13 micron. The median particle size decreased after the 2nd sludge addition (at 4.5 wt %) to 11 micron. Following the addition of additional MST, the median particle size decreased to 9 micron. This observation is somewhat surprising, since MST generally has a larger particle size than sludge. The observation could be due to the high solids loading in the test or to shearing of particles. Following slurry addition and volume reduction to reach 5.5, 9.9, and 10.8 wt % solids, the median particle size remained 9 micron. Following volume reduction to reach 12.2 wt %, the median particle size increased to 14 micron and the shape of the particle size curve changed dramatically. One plausible explanation for this observation is that the solids loading was so high that the probe became covered with particles and could not measure their cord length. A second explanation is that the yield stress of the feed slurry became very large and the tank could not be mixed. The slurry in front of the FBRM probe was stagnant. The third explanation is particle agglomeration at the highest solids loading. When the slurry was diluted to 7.7 wt %, the median particle size reduced to 10 micron, and the shape of the curve agreed with other curves from this test.

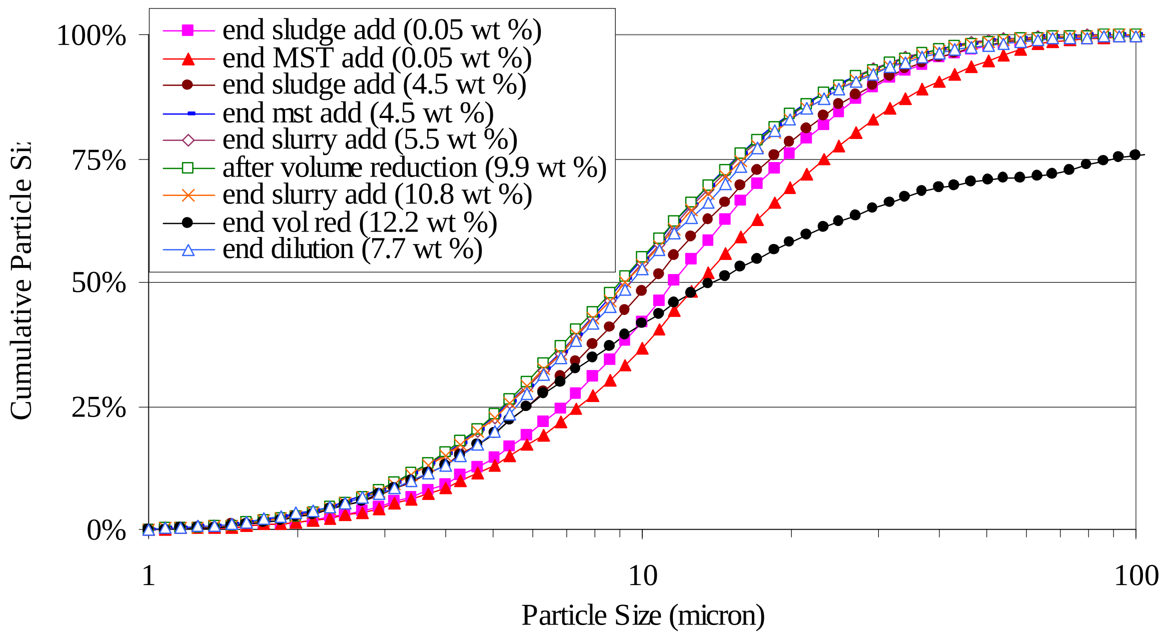

Figure 12. Particle size data from the 0.1 m Filter Test

CONCLUSIONS

The conclusions from this work follow.

- The 0.1 micron filter produced a higher flux than observed in comparable tests with the 0.5 micron filter at 0.06 wt % solids and 4.5 wt % solids.
- The average filter flux equaled 0.132 gpm/ft^2 at 0.06 wt % solids, 0.069 gpm/ft^2 at 4.5 wt % solids, and 0.026 gpm/ft^2 at 12.2 wt % solids.
- The flux for the 12.2 wt % slurry exceeded the design bases for the processes.
- Filter flux demonstrated a statistically significant correlation with axial velocity, with increasing axial velocity producing higher filter flux for 0.06, 4.5, and 12.2 wt % slurries.

- Filter flux demonstrated a statistically significant correlation with transmembrane pressure (TMP), with increasing TMP producing higher filter flux for 0.06 and 12.2 wt % slurries. We did not observe a statistically significant correlation between transmembrane pressure and filter flux for the 4.5 wt % slurry.
- The cooling rate needed to maintain 35 ± 3 ºC with 12.2 wt % slurry measured 17,600 – 20,100 BTU/hr.

REFERENCES

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3. M. R. Poirier, "FY2000 FRED Test Report,"WSRC-TR-2001-00035, Rev. 0, January 11, 2001 and included report "Final Report on the Crossflow Filter Optimization with 5.6 M Sodium Salt Solution" (V. Van Brunt, C. Stork, T. Deal, and R. Haggard, USC-FRED-PSP-RPT-09-0-015, December 20, 2000).
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5. D. T. Hobbs, Laboratory Notebook, WSRC-NB-2001-00011, pp. 104-105.
6. J. Murkes and C. G. Carlsson, Crossflow Filtration, New York: Wiley, 1988.

APPENDIX A
STATISTICAL DATA

Response Flux
Whole Model
Summary of Fit

RSquare	0.921488
RSquare Adj	0.913224
Root Mean Square Error	0.014023
Mean of Response	0.066674
Observations (or Sum Wgts)	43

Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Model	4	0.08770672	0.021927	111.5007
Error	38	0.00747273	0.000197	Prob > F
C. Total	42	0.09517944		<.0001

Parameter Estimates

Term	Estimate	Std Error	t Ratio	Prob>|t|
Intercept	0.0418187	0.011336	3.69	0.0007
Conc	-0.000826	0.000043	-19.06	<.0001
Velocity	0.0052037	0.000718	7.25	<.0001
TMP	0.0008223	0.000241	3.42	0.0015
Test order	0.0002896	0.000531	0.55	0.5889

Effect Tests

Source	Nparm	DF	Sum of Squares	F Ratio	Prob > F
Conc	1	1	0.07145394	363.3547	<.0001
Velocity	1	1	0.01033392	52.5496	<.0001
TMP	1	1	0.00229383	11.6645	0.0015
Test order	1	1	0.00005842	0.2971	0.5889

Prob > F is less than 0.05 for TMP, axial velocity, and concentration. Probability being less than 0.05 indicates the effect is statistically significant by F-test with 95% confidence.

Prob>|t| is less than 0.05 for TMP, axial velocity, and concentration. Probability being less than 0.05 indicates the effect is statistically significant by t-Test with 95% confidence.

Prob > F is greater than 0.05 for test order. Probability being greater than 0.05 indicates the effect is not statistically significant by F-test with 95% confidence.

Prob>|t| is greater than 0.05 for test order. Probability being greater than 0.05 indicates the effect is not statistically significant by t-Test with 95% confidence.