Development of a Crossflow Filter to Remove Solids from Radioactive Liquid Waste: Comparison of Test Data with Operating Experience -- 9119

M. R. Poirier, D. T. Herman, and S. D. Fink
Savannah River National Laboratory
Aiken, SC 29808

J. U. Lacerna
Washington Savannah River Company
Aiken, SC 29808

ABSTRACT

In 2008, the Savannah River Site (SRS) began treatment of liquid radioactive waste from its Tank Farms. To treat waste streams containing Cs-137, Sr-90, and actinides, SRS developed the Actinide Removal Process (ARP) and the Modular Caustic Side Solvent Extraction Unit (MCU). The Actinide Removal Process contacts the waste with monosodium titanate (MST) to sorb strontium and select actinides. After MST contact, the process filters the resulting slurry to remove the MST (with sorbed strontium and actinides) and any entrained sludge. The filtrate is transported to the MCU to remove cesium. The solid particles removed by the filter are concentrated to ~ 5 wt %, washed to reduce the concentration of dissolved sodium, and transported to the Defense Waste Processing Facility (DWPF) for vitrification.

The authors conducted tests with 0.5 μ and 0.1 μ Mott sintered stainless steel crossflow filters at bench-scale (0.018 m^2 or 0.19 ft^2 surface area) and pilot-scale (1.04 m^2 or 11.2 ft^2). The collected data supported design of the filter for the process and identified preferred operating conditions for the full-scale process (21.4 m^2 or 230 ft^2). The testing investigated the influence of operating parameters, such as filter pore size, axial velocity, transmembrane pressure (TMP), and solids loading, on filter flux, and validated the simulant used for pilot-scale testing.

The conclusions from this work follow.

- The 0.1 μ Mott sintered stainless steel filter produced higher flux than the 0.5 μ filter. The likely cause of this result is the smaller pore size prevented submicron particles becoming trapped in the pores, which significantly increases filter resistance and decreases filter flux.
- Filtrate samples showed no visible solids.
- The filter flux with actual waste is comparable to the filter flux with simulated waste, with the simulated waste being conservative. This result shows the simulated sludge is an acceptable representative of the actual sludge.
- When the data is adjusted for differences in transmembrane pressure and temperature, the filter flux in the Actinide Removal Process is comparable to the filter flux in the bench-scale and pilot-scale testing.
- Filter flux increased with transmembrane pressure, increased with axial velocity, and decreased with concentration in agreement with classical crossflow filtration theories.

INTRODUCTION

The Savannah River Site has developed a treatment process, called the Actinide Removal Process (ARP), to remove strontium and select actinides from radioactive liquid waste. In this process, liquid waste containing nominally 0.6 g/L of sludge particles is transported to a tank. MST (0.4 g/L) is added to the tank, which is mixed for 24 hours. After 24 hours, the slurry is filtered to remove the insoluble sludge and MST particles. Filtration continues until the feed slurry is concentrated to 5 wt % insoluble solids. The filtrate is transported to a solvent extraction process to remove cesium or, depending on radionuclide content, to the feed tank for a grout process to stabilize the waste. The solids are washed to reduce the sodium concentration. The concentrated, washed solids are transported to the Defense Waste Processing Facility (DWPF) for vitrification.

The filtration process is one of the rate limiting steps for this process. To size the filtration equipment and maximize throughput, the authors conducted pilot-scale filtration testing with simulated waste streams and bench-scale filtration testing with actual waste. This work defined the final design for the operating facility. The ARP started processing waste in May 2008.

This paper describes the testing conducted and compares the observed behavior with the performance of the filter after startup of the ARP.

TEST DESIGN

Equipment

This report discusses four sets of scale-up tests to support the design of the ARP. The four tests are bench-scale actual waste tests with a 0.5 μ filter, bench-scale actual waste tests with a 0.1 μ filter, pilot-scale simulant tests with a 0.5 μ filter, and pilot-scale simulant tests with a 0.1 μ filter. All testing used filter tubes fabricated by Mott Corporation.

The 0.5 μ filter was selected for evaluation, because it had been previously selected for the In Tank Precipitation and Late Wash processes at SRS. In addition, a full-scale 21.4 m^2 (230 ft^2) unit existed in the building where the ARP was to be located. Because the Hanford Waste Treatment Plant selected a 0.1 μ filter for their solid liquid separation process, the authors evaluated that pore size as well.

The bench-scale testing occurred in the Savannah River National Laboratory (SRNL) Shielded Cells. The equipment contained a sintered stainless steel 0.5 μ pore-size cross-flow filter of 1.27 cm (0.5 inches) outer diameter (OD), 0.95 cm (0.375 inches) inner diameter (ID), and 0.61 m (2 ft) length. The equipment included a feed vessel, a feed pump, a heat exchanger to control solution temperature, a magnetic flow meter to measure the filter feed rate, and pressure gauges to measure feed, concentrate, and filtrate pressure. A graduated glass cylinder located down stream of the filter measured the filtrate flow rate, or flux. Personnel determined the flux, by measuring the time to collect a known volume of filtrate. The working volume of the equipment is approximately 800 mL. A 0.1 μ pore size filter with the same dimensions and materials was used for some tests.

The pilot-scale filtration testing occurred at the Filtration Research Engineering Demonstration (FRED) located at the University of South Carolina. The FRED facility contains a filter element with seven filter tubes. Each tube is constructed of sintered stainless steel, 1.91 cm (0.75 inches) OD, 1.59 cm (0.625 inches) ID, and 3.05 m (10 feet) long. The filter elements tested had pore sizes of 0.1 and 0.5 μ. The FRED facility uses identical, but fewer, filter tubes compared to the ARP. The equipment contains flow meters to measure feed and filtrate flow rates, pressure gauges to measure feed, concentrate, and filtrate pressure, and thermocouples to measure temperature.

The filter in the ARP contains 144 filter tubes. Each tube is 3.05 m (10 feet) long, 1.91 cm (0.75 inches) OD, and 1.59 (0.625 inches) ID. Based on the laboratory and pilot-scale testing, the tubes are constructed of sintered stainless steel with a pore size of 0.1 μ.

Slurries Tested and Operating Conditions

In the 0.5 μ pore-size bench-scale test with actual waste test [1], the feed slurry contained 5.6 M sodium salt solution from two tanks, sludge composite from three tanks, and MST. The sludge and supernate composition are described in the test report [1]. The ratio of sludge to MST was 0.6:0.55. The sludge MST ratio was 6:5.5 based on nominal feed assumptions of 0.6 g/L entrained sludge and 0.55 g/L MST. The axial velocity varied from 1.8 – 3.7 m/s (6 – 12 ft/s),the transmembrane pressure (TMP) varied from 103 - 282 kPa (15 – 41 psi), and the temperature was ~ 33 °C. The insoluble solids concentration varied from 0.06 – 4.5 wt %. We collected filtration data at varying axial velocity, TMP, and solids loading in a statistically designed test. Following each change in test conditions, the filter was backpulsed to remove the filter cake. Each test condition was held for at least 30 minutes to obtain steady-state.

Subsequent bench-scale testing examined both 0.1 μ and 0.5 μ pore-size filters using actual waste test, the feed slurry contained 5.6 M sodium salt solution collected from four tanks, a sludge sample from a feed tank to DWPF, and MST [2]. The sludge and supernate composition are described in the test report [2]. The ratio of sludge to MST was 0.6:0.55. The axial velocity varied from 1.8 – 4.2 m/s (6 – 14 ft/s), the TMP varied from 103 - 282 kPa (15 – 41 psi), and the temperature was ~ 33 °C. The insoluble solids concentration varied from 0.06 – 4.5wt %. The filter pore size was 0.5 and 0.1 μ. We collected filtration data at varying axial velocity, TMP, and solids loading in a statistically designed test. Following each change in test conditions, the filter was backpulsed to remove the filter cake. Each test condition was held for at least 30 minutes to obtain steady-state.

In the 0.5 μ pore-size pilot-scale simulant filtration tests [3], the feed slurry contained 5.6 M sodium salt solution, simulated SRS sludge, and MST. The sludge and supernate composition are described in the test report [3]. The sludge MST ratio was 6:5.5. The axial velocity varied from 3.7 – 8.0 m/s (12 – 26 ft/s), the TMP varied from 82 – 470 kPa (12 – 68 psi), and the temperature was ~ 35 °C. The insoluble solids concentration varied from 0.03 – 3.3 wt %. We collected filtration data at varying axial velocity, TMP, and solids loading in a statistically designed test. Following each change in test conditions, the filter was backpulsed to remove the filter cake. Each test condition was held for at least one hour to obtain steady-state.

In the 0.1 μ pore-size pilot-scale simulant filtration tests [4], the feed slurry contained 5.6 M sodium salt solution, simulated SRS sludge, and MST. The sludge and supernate composition are described in the test report [4]. The sludge MST ratio was 6:5.5. The axial velocity varied from 1.8 – 4.2 m/s (6 – 14 ft/s), the transmembrane pressure (TMP) varied from 103 – 310 kPa (15 – 45 psi), and the temperature was ~ 33 °C. The insoluble solids concentration varied from 0.06 – 12 wt %. We collected filtration data at varying axial velocity, TMP, and solids loading in a statistically designed test. Following each change in test conditions, the filter was backpulsed to remove the filter cake. Each test condition was held for at least one hour to obtain steady-state.

RESULTS

Bench-Scale Actual Waste Test with 0.5 μ Filter

Figure 1 shows the filter flux from bench-scale actual waste tests using feed slurries containing 0.29 wt % solids [1]. The figure shows a correlation between filter flux and TMP. The data also show good agreement between filter flux with actual waste and filter flux with simulated waste. Similar results occurred at other concentrations of solids. The filtrate showed no visible solids. In general, the filter flux proved higher with actual waste than with simulant, suggesting the simulated sludge is a conservative source of solids for filter testing. Possible reasons for this occurrence are the smaller tube diameter and shorter tube length.

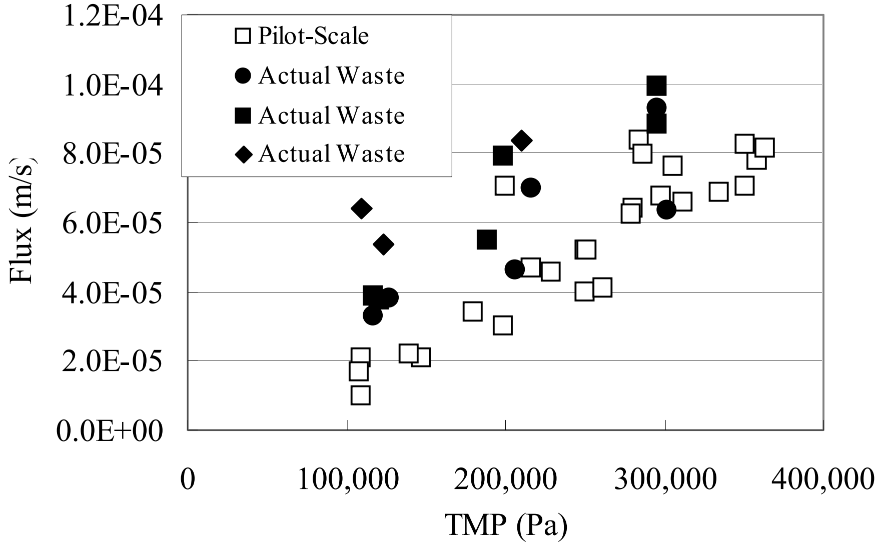

Figure 1. Bench-Scale Actual Waste Filter Flux with 0.29 wt % solids.

The bench-scale filter tube is 0.375 inches in ID, while the pilot-scale filter tubes are 0.625 inches in ID. Given the same axial velocity, the smaller diameter tube will have a higher wall shear stress. This higher wall shear stress will reduce the filter cake thickness and increase filter flux.

The shorter tube length has the following effect on the filtration rate. When the feed slurry enters the filter tube, it develops and builds up a boundary layer in the entrance region. In the entrance region, the mass transfer rate is higher. The higher mass transfer rates help remove solid particles from the filter cake. The filter flux is higher in the entrance region than in the remainder of the filter tube. With a shorter tube, the entrance region occupies a larger fraction of the filter and the average mass transfer rate (and filter flux) will be higher.

Bench-Scale Actual Waste Test with 0.1 □ Filter

In subsequent testing with both 0.1 μ and 0.5 μ pore-size filters, the data shows higher filter flux with a 0.1 □ filter than with a 0.5 □ filter. The filter flux averaged 9.2 x 10^{-5} m/s (0.135 gpm/ft^2) with the 0.1 □ filter versus 5.6 x 10^{-5} m/s (0.082 gpm/ft^2) with the 0.5 □ filter for slurry containing 1.4 wt % solids.

The result is not intuitively obvious, since a 0.5 □ filter has less resistance to flow than a 0.1 □ filter, everything else being the same. However, the smaller particles in the feed slurry (< 1 □) are more likely to become trapped in the pores of the 0.5 □ filter than the 0.1 □ filter. When the particles are trapped in the 0.5 μ filter pores, the filtrate resistance increases dramatically greatly reducing the effective porosity of the filter. When the particles remain in the slurry cake on the surface of the 0.1 μ filter, we suspect they form a filter cake with higher porosity than the fouled 0.5 μ filter which does not increase the resistance as much and this filter cake readily removed by backpulsing.

The filter pore size is a nominal pore size. A 0.5 □ filter rejects 50% of the particles greater than 0.5 □, 90% of the particles greater than 1 □, 99% of the particles greater than 1.7 □, and 99.9% of the particles greater than 2.2 □.[5] A 0.1 □ filter rejects 50% of the particles greater than 0.1 □ and 99.9% of the particles greater than 0.7 □.

Analysis of SRS actual sludge shows 0.3 – 6 vol % of the particles are less than 0.7 □ and 5 – 40 vol % of the particles are between 0.7 and 2.2 □ [6]. Therefore, a significant fraction of the sludge particles are smaller than 2.2 □, and within the range to become trapped in the pores of the 0.5 □ filter, but not the 0.1 □ filter.

The filtrate showed no visible solids during the test.

Pilot-Scale Simulant Test with 0.5 □ Filter

Figure 2 shows the filter flux as a function of TMP and solids loading for the pilot-scale testing using 0.5 μ pore-size filters and simulated waste. The data shows strong correlation with TMP and solids loading. One reason for the high correlation between the flux and TMP is the high axial velocity in this test. The corresponding plot of filter flux as a function of axial velocity (not shown) does not indicate a correlation for these variables. The testing also showed backpulsing the filter is an effective means for recovering filter flux. The filtrate contained no visible solids, indicating that the filter removed the solid particles from the feed slurry. The turbidity of filtrate samples collected during the test measured less than 10 NTU. Given the ARP filter area of 21.4 m^2 (230 ft^2), the data suggests the ARP filtration rate will be 0.017 – 0.10 m^3/min (4.6 – 27 gpm) at 275 kPa (40 psi) TMP.

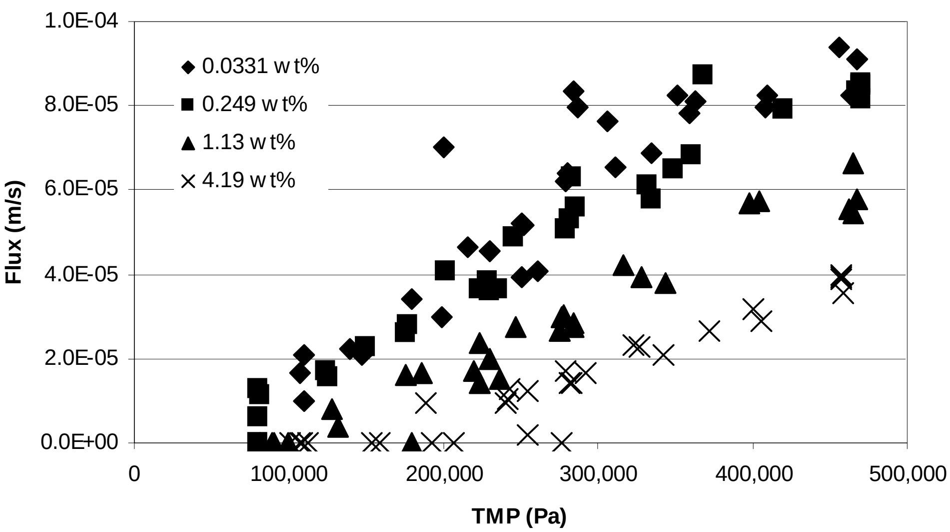

Figure 2. Filter Flux for Pilot-Scale Simulant Test with a 0.5 □ Filter

A statistical analysis of the data resulted in the following model:

$$J \text{ (m/s)} = 22.1 + 19{,}000 \, P \text{ (Pa)} - 27.9 \ln[C \text{ (g/L)}] + 0.0048 \, v(\text{m/s})^2/C(\text{g/L})^2 \quad [1a]$$

$$[J \text{ (gpm/ft}^2) = 0.015 + 0.0019 \, P \text{ (psi)} - 0.019 \ln[C \text{ (g/L)}] + 0.000035 \, v(\text{ft/s})^2/C(\text{g/L})^2] \quad [1b]$$

where J is filter flux, P is transmembrane pressure, C is solids concentration, and v is axial velocity. The model predicts filter flux will increase with higher transmembrane pressure, increase with greater axial velocity, and decrease with elevated solids concentration in agreement with classical crossflow filtration theories.

Pilot-Scale Simulant Test with 0.1 □ Filter

Figure 3 shows the filter flux from the pilot-scale filter test using simulated waste and 0.1 □ pore-size filters. The data shows higher filter flux with a 0.1 □ filter than with a 0.5 □ filter. At nominal 0.05 wt % solids, the filter flux averaged 9.0 x 10^{-5} m/s (0.133 gpm/ft^2) with the 0.1 □ filter versus 5.8 x 10^{-5} m/s (0.086 gpm/ft^2) with the 0.5 □ filter. At nominal 4.5 wt % solids, the filter flux averaged 4.7 x 10^{-5} m/s (0.069 gpm/ft^2) for the 0.1 □ filter versus 1.5 x 10^{-5} m/s (0.022 gpm/ft^2) with the 0.5 □ filter. This increase in filter flux with the 0.1 □ filter is consistent with results from the bench-scale testing.

The filtrate contained no visible solids, indicating that the filter removed the solid particles from the feed slurry. The turbidity of filtrate samples collected during the test measured less than 10 NTU.

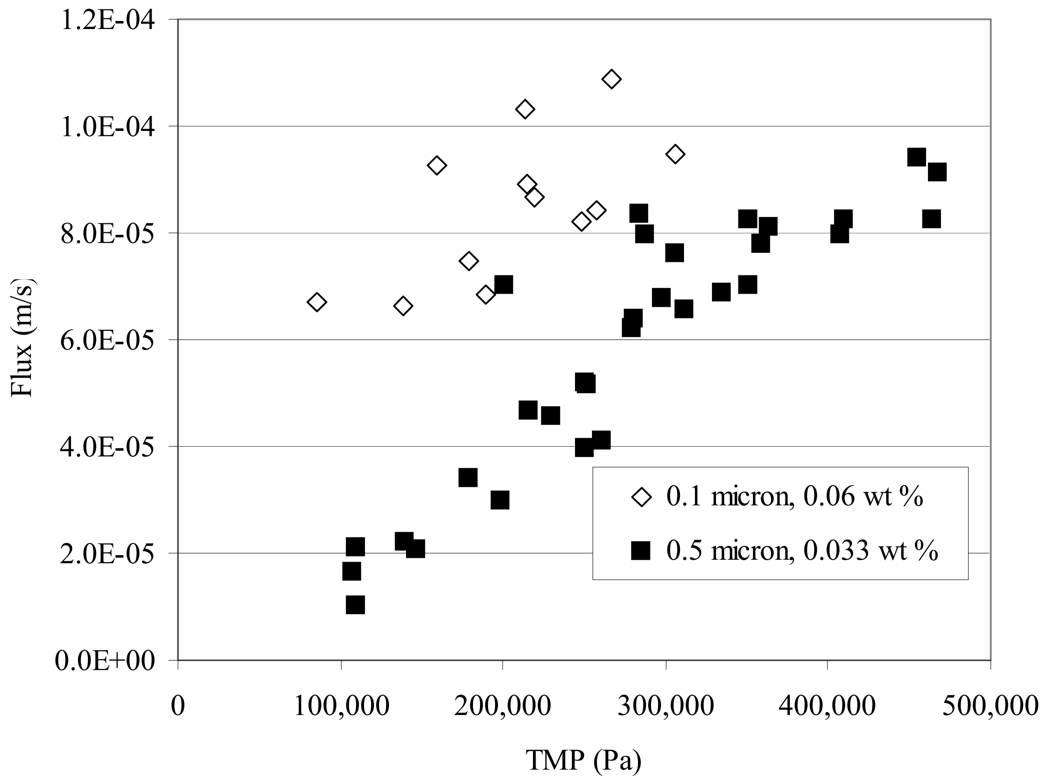

Figure 3. 2003 Pilot-Scale Simulant Filter Test with a 0.1 □ Filter

Actinide Removal Process (ARP) Operations

The ARP started radiological operations in May 2008. The design operating conditions are 3.0 – 3.4 m/s (10 – 11 ft/s) axial velocity and 275 kPa (40 psi) TMP. The ARP contains a 0.5 □ sintered stainless steel dead end guard filter down stream of the crossflow filter. The project installed this filter to protect downstream facilities (i.e., the MCU) in case of particle breakthrough of the crossflow filter. Since startup, the guard filter has experienced high pressure drops, which have reduced the flow rate through the crossflow filter.

Figure 4 shows the crossflow filter data from the ARP, along with data from the bench-scale and pilot-scale tests with a 0.1 □ filter. The ARP data was collected at lower TMP and temperature than the test data. The lower transmembrane pressure occurred because the secondary, or guard, had a much higher pressure drop than expected. This high resistance through the guard filter decreased the overall filtration rate. The ARP filtration occurred at lower temperature than the bench-scale and pilot-scale tests (18 – 25 °C versus ~ 35 °C). The reason for the lower temperature is to prevent post-filtration precipitation in the

MCU, which operates at 23 °C. The authors corrected the measured ARP filter flux to 275 kPa using equation [1a] and to 35 °C using equation [2]

$$J\ (35\ °C) = J\ (T)\ \exp\{2500[(1/(273+T))-(1/308)]\} \quad [2]$$

where J is filter flux and T is temperature in °C.

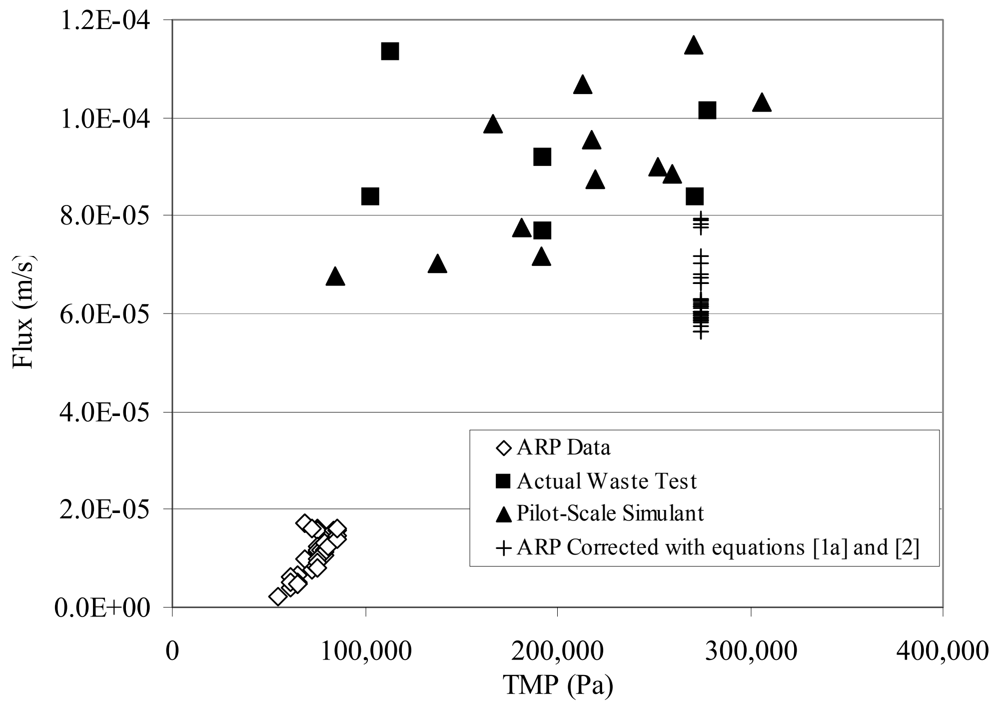

Figure 4. ARP Filtration Data

After correcting for transmembrane pressure and temperature, the corrected ARP filtration rates are ~ 20% lower than the rates in the actual waste test, and ~ 30% lower than the rates in pilot-scale simulant test. Possible causes of the difference in filter flux between the ARP process and the bench-scale and pilot-scale tests are differences in feed solids, recycling the feed slurry, and differences in operation time.

Each of the tests used a different source of sludge. The actual waste test used an archived sample of Tank 40H sludge, the pilot-scale test used simulated Tank 8F and Tank 40H sludge, and the ARP feed contains Tank 49H entrained sludge. While sludge components of these feed slurries are expected to have similar particle size distributions, the particle size distribution were not measured on the actual samples. In addition, each test used a different batch of MST. Table 1 shows the fraction of the MST particles less than 1 □ in each test and for the ARP operations. The MST added to the ARP feed contains a higher fraction of submicron particles than the MST used in the testing. These submicron particles could become trapped in the filter pores, increasing the filter resistance and decreasing the filter flux.

Table 1. MST Characteristics

Test	Bench-Scale	Pilot-Scale	ARP
Fraction < 1 □	1.7%	1.7%	5.86 - 7.83%

During testing, the feed solution was recycled to minimize the amount needed for testing. Because of the recycling, once the fine particles became trapped in the filter pores they would be removed from the feed.

Because the ARP has a continuous fresh feed, a small amount of fine particles would become significant after processing thousands of gallons of feed. Measurements of the feed showed the presence of aluminum-containing fines [7, 8].

The differences in operating time may explain some of the differences in flux. The bench-scale tests operated for 30 minutes, the pilot-scale tests operated for 60 minutes, and the ARP operated for 600 – 4000 hours. A slow gradual decrease in filter flux would not be observable over 30 – 60 minutes, but it would be observable and significant over the long duration of an ARP filtration cycle.

CONCLUSIONS

The conclusions from this work follow.

- The 0.1 □ Mott sintered stainless steel filter produced higher flux than the 0.5 □ filter. The likely cause of this result is the smaller pore size prevented submicron particles becoming trapped in the pores, which significantly increases filter resistance and decreases filter flux.
- Filtrate samples showed no visible solids.
- The filter flux with actual waste was comparable to the filter flux with simulated waste, with the simulated waste being conservative. This result shows the simulated sludge is an acceptable representative of the actual sludge.
- When the data is adjusted for differences in transmembrane pressure and temperature, the filter flux in the Actinide Removal Process proved comparable to the filter flux in the testing.
- Filter flux increased with transmembrane pressure, increased with axial velocity, and decreased with concentration in agreement with classical crossflow filtration theories.

REFERENCES

1. M. R. Poirier, F. F. Fondeur, T. L. Fellinger, and S. D. Fink "Cross-Flow Filtration Demonstration for Slurries Containing High Level Waste Sludge and Monosodium Titanate", WSRC-TR-2001-00212, April 11, 2001.
2. M. R. Poirier, J. L. Siler, and S. D. Fink, "Filtration of Actual Savannah River Site Waste Treated with Permanganate or Monosodium Titanate", WSRC-TR-2002-00134, March 14, 2002.
3. M. R. Poirier, "FY2000 FRED Test Report", WSRC-TR-2001-00035, January 11, 2001.
4. Michael R. Poirier, Jeffrey L. Siler, Samuel D. Fink, Ralph Haggard, Travis Deal, Carol Stork, and Vincent Van Brunt, "Pilot-Scale Testing of a 0.1 Micron Filter with SRS Simulated High Level Waste", WSRC-TR-2003-00469, October 8, 2003.
5. M. R. Poirier, T. M. Jones, S. D. Fink, R. Haggard, and V. Van Brunt, "Evaluation of Mott Filter Performance", WSRC-TR-2002-00256, May 30, 2002.
6. M. R. Poirier, F. F. Fondeur, D. P. Lambert, David T. Hobbs, and S. D. Fink, "Particle Size of Simulated SRS Sludge, Actual SRS Sludge, and Monosodium Titanate", WSRC-TR-2003-00221, May 20, 2003.
7. C. A. Nash, T. B. Peters, S. D. Fink, "Tank 49H Salt Batch Supernate Qualification for ARP/MCU", WSRC-STI-2008-00117, Rev. 0, August 25, 2008.
8. T.B. Peters and S.D.Fink, "ISDP Salt Batch #2 Supernate Qualification", SRNL-STI-2008-00446, Rev. 1, January 5, 2009.