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Impact of Axial Velocity and Transmembrane Pressure (TMP) on ARP Filter Performance

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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 potential method for increasing filter flux is to adjust the axial velocity and transmembrane pressure (TMP). SRR requested SRNL to conduct bench-scale filter tests to evaluate the effects of axial velocity and transmembrane pressure on crossflow filter flux. The objective of the testing was to determine whether increasing the axial velocity at the ARP could produce a significant increase in filter flux. The authors conducted the tests by preparing slurries containing 6.6 M sodium Salt Batch 6 supernate and 2.5 g MST/L, processing the slurry through a bench-scale crossflow filter unit at varying axial velocity and TMP, and measuring filter flux as a function of time.

The conclusions from this work follow.

- Filter flux increased with increasing transmembrane pressure, agreeing with the Hagen-Poiseuille equation.
- Filter flux decreased with increasing MST concentration, agreeing with the boundary layer model (and other crossflow filtration models).
- No effect of axial velocity on filter flux was observed during the testing. The likely reason for this result is that the operating conditions were not in the range at which axial velocity would have a significant effect on filter flux.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

ARP	Actinide Removal Process
CSSX	Caustic Side Solvent Extraction
MCU	Modular Caustic Side Solvent Extraction Unit
MST	Monosodium titanate
OD	Outer diameter
SRNL	Savannah River National Laboratory
SS	Stainless Steel
SS	Sum of Squares of Deviation from Mean
TMP	Transmembrane pressure

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.[1] There is a desire to understand the causes of the low filter flux and to increase ARP/MCU throughput.

One potential method for increasing filter flux is to adjust the axial velocity and transmembrane pressure (TMP). Increasing the TMP increases the driving force for liquid to move through the filter. Increasing the axial velocity increases the shear stress at the filter surface, which will reduce the resistance to flow through the filter. SRR requested SRNL to conduct bench-scale filter tests to evaluate the effects of axial velocity and transmembrane pressure on crossflow filter flux.[2] The objective of the testing was to determine whether increasing the axial velocity at the ARP could produce a significant increase in filter flux.

The authors conducted the tests by preparing slurries containing 6.6 M sodium Salt Batch 6 supernate and 2.5 g MST/L, processing the slurry through a bench-scale crossflow filter unit at varying axial velocity and TMP, and measuring filter flux as a function of time.

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 ºC. 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 gpm ± 0.1 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 ¼" SS tubing, and the filtrate lines, which are 3/8" OD and ¼" OD SS tubing.

The filtrate leaves each filter through 3/8" and ¼" 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. The filtrate can be directed to the filtrate tank or to the 100 mL ±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 μm pore size, 3/8 inch ID Mott® porous metal crossflow filter and the other is a 0.5 μm 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. For this testing, only the 0.1 μm filter was used.

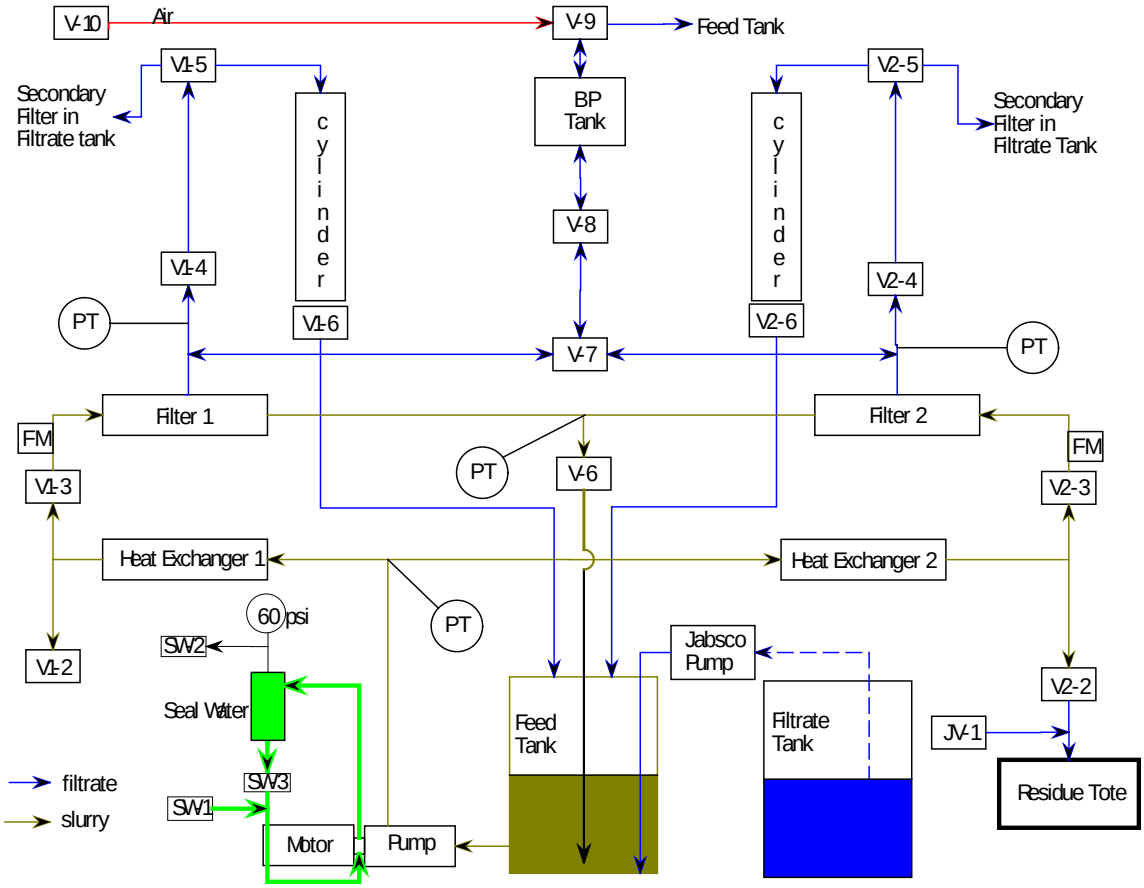

Figure 1. Schematic of Laboratory-Scale Crossflow Filter Unit

2.2 Test Protocol

The 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 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

Flux 0.1 micron filter (gpm/ft^2)	Flux 0.5 micron filter (gpm/ft^2)	TMP (psi)
0.64	2.3	40

The simulated salt solution was based on the Salt Batch 6 analysis.[3] 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 5). The salt solution was prepared by dissolving sodium hydroxide in deionized water, then adding aluminum nitrate to react with the sodium hydroxide forming sodium aluminate. The remaining components were added in order of increasing solubility. 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

Ion	Concentration (M)
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

Sufficient MST was added to the salt solution to produce a slurry containing 2.5 g MST/L slurry.[a] The slurry was mixed for 15 minutes and run through the filter at an axial velocity of 10.2 ft/s, a TMP of 30 psi, and a temperature of 25 ˚C. For the first hour, the filtrate was returned to the feed tank (recycle mode). After one hour, the filtrate was collected in a separate container and the feed slurry was concentrated (concentration mode). Once the level in the feed tank reached 3 gallons, the filtrate was recycled for the remainder of the day.

The next day, the filtrate was returned to the feed tank and the filter operated for 2 hours in recycle mode at each of the conditions in Table 3. The table includes an ARP axial velocity and a test axial velocity. The test filter has an internal diameter of 3/8 inch and the ARP filter has an internal diameter of 5/8 inch. Because the SRNL filter and ARP filter have different internal diameters, the authors accounted for these differences by matching the wall shear stress. The wall shear stress as a function of axial velocity was calculated for each filter by the Blasius equation.[2] Test velocities were selected to match the wall shear stress.

Additional tests were conducted with MST concentrations of 10 g/L and 30 g/L. Based on analysis of samples from ARP, these concentrations (2.5, 10 , and 30 g/L) would approximate the start of batch 6, the start of batch 44, and the end of batch 44.[4] MST was added to the feed tank to reach the target concentration.

[a] The MST particle size was approximately 10 micron as described in SRNL-STI-2015-00158, Revision 0

Table 3. Operating Conditions for Axial Velocity-TMP Tests

Test	ARP Velocity (ft/s)	ARP TMP (psi)	Test Velocity (ft/s)	Test TMP (psi)
1	11	30	10.2	30
2	8	25	7.4	25
3	11	22.9	10.2	22.9
4	11	30	10.2	30
5	6.8	30	6.3	30
6	14	35	13.0	35
7	11	30	10.2	30
8	8	35	7.4	35
9	11	37.1	10.2	37.1
10	11	30	10.2	30
11	14	25	13.0	25
12	15.2	30	14.2	30
13	11	30	10.2	30

3.0 Results

Figure 2 shows the filter flux as a function of time and operating conditions. The plot shows filter flux to decrease with increasing MST concentration. The effects of axial velocity and TMP are not obvious from the figure.

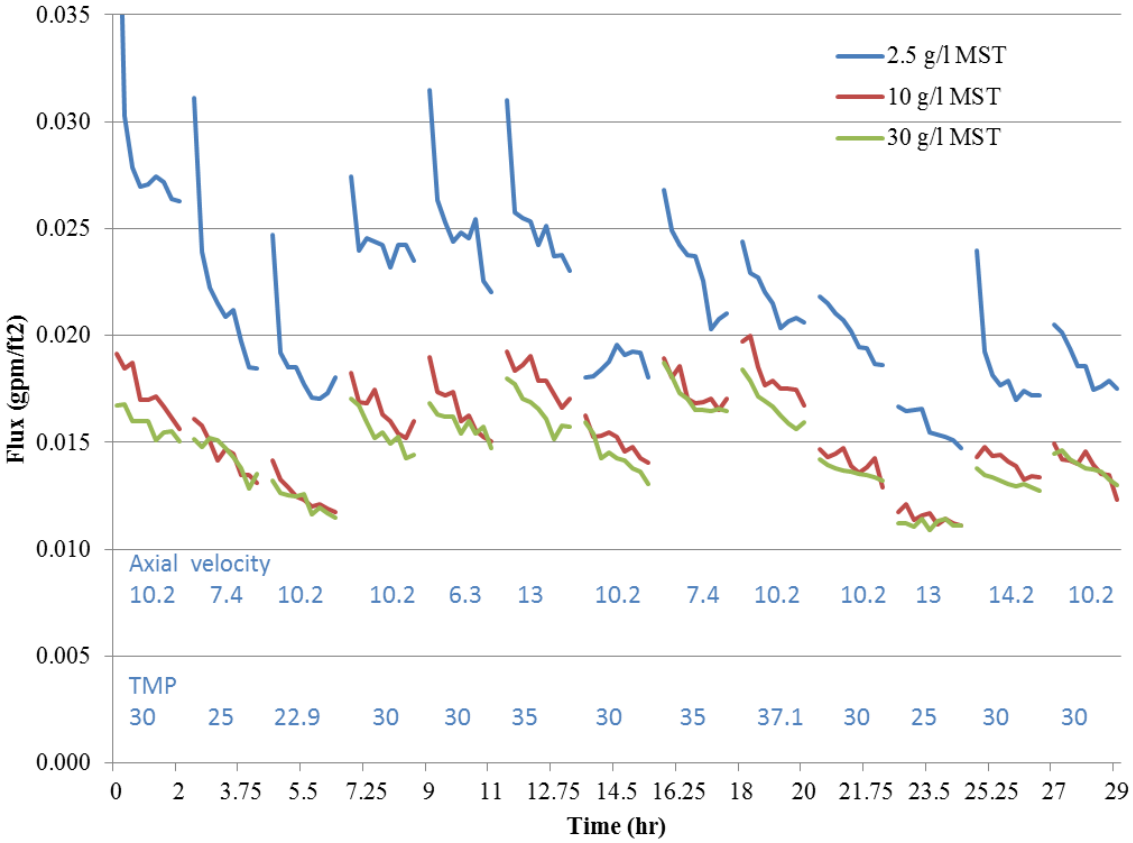

Figure 2. Filter Flux as a Function of Time and Operating Conditions

A statistical analysis of the data was conducted using the JMP® software to evaluate the impact of run number, axial velocity, TMP, and MST concentration on filter flux. In the table, SS is the sum of the squares of the deviation from the mean, F is the ratio of the mean square deviation from the model to the mean square deviation from the random error, and Prob>F is the probability that the variance is due to random errors.[8] Prob>F values less than 0.05 indicate that the effect is statistically significant with 95% confidence. According to Table 4, run number, TMP, and concentration effects are statistically significant. Axial velocity is not statistically significant.

Table 4. Statistical Analysis of Filtration Data

Source	SS	F ratio	Prob>F
Run number	**0.00006349**	**41.12**	**<0.0001**
Axial Velocity	**0.00000102**	**0.66**	**0.42**
TMP	**0.00012763**	**82.67**	**<0.0001**
1/Concentration	**0.00031309**	**202.80**	**<0.0001**

Run number having an effect on filter flux is expected. Previous testing and operating experience has shown filter flux to decrease with run time. As the filter is operating, more particles have the opportunity to build a filter cake or become trapped in the filter pores, which increases the resistance to filtration and decreases the filter flux.

The Hagen- Poiseuille equation describes laminar flow through a circular tube. Equation [1] describes the equation as it is applied to flow through porous media.

$$J = \frac{\Delta P d_p^{\,2}}{32 \mu L} = A_1 \Delta P \qquad [1]$$

where ΔP is the transmembrane pressure, d_p is pore diameter, μ is viscosity, and L is pore length.[5] The model predicts filter flux to be proportional to transmembrane pressure. The test data are consistent with this model.

Equation [2] and equation [3] describe the boundary layer model for filter flux[6]

$$J = k \ln\left(\frac{C_w}{C_b}\right) = A_8 v^{1.75} \ln\left(1/C_b\right) \qquad [2]$$

$$k = 0.078\left(\frac{d_p^{\,4}}{L}\right)^{1/3} \tau_w \qquad [3]$$

where τ_w is the wall shear stress, C_w is the concentration of solid particles at the filter surface, d_p is particle diameter, C_B is the bulk solids concentration, L is tube length, k is the mass transfer coefficient and v is axial velocity. The model predicts filter flux to increase with increasing axial velocity and to

decrease with increasing solid particle concentration. Other models predict similar results. The test data are not consistent with this model.

The Murkes-Carlsson model combines the effects of axial velocity, TMP, and solid particle concentration. Equation [4] describes the Murkes-Carlsson model[7]

$$J = \frac{\left(\frac{B\Delta P}{\mu}\right)}{\left[\frac{5.9C_b^{2/3}\left[\frac{2D}{(1-\varepsilon)\rho}\right]\Delta P}{\left(\left(\frac{21\mu}{d_p\rho}\right)v^3 + 6\left(\frac{\mu}{d_p\rho}\right)^{1/2} v^{3.5} + 0.28v^4\right)^{1/2}} + L_0\right]} = \frac{a_1\Delta P v^{1.5}}{a_2\Delta P C_b^{2/3} + v^{1.5}} \qquad [4]$$

where B is Darcy's permeability constant, ΔP is transmembrane pressure, μ is viscosity, C_b is bulk concentration, D is filter tube diameter, ε is porosity, ρ is density, d_p is particle size, v is axial velocity, and L_0 is equivalent cake thickness. The model predicts filter flux to be a function of transmembrane pressure, axial velocity and solids loading. At large axial velocity, filter flux is a function of TMP, and increases with increasing TMP. At small axial velocity, filter flux is a function of axial velocity and solid particle concentration, increasing with increasing axial velocity, and decreasing with increasing concentration. At large TMP, filter flux is a function of axial velocity and solid particle concentration, increasing with increasing axial velocity, and decreasing with increasing concentration. At small TMP, filter flux is a function of TMP, and increases with increasing TMP. At large solid particle concentration, filter flux is a function of axial velocity and solid particle concentration, increasing with increasing axial velocity, and decreasing with increasing concentration. At small particle concentration, filter flux is a function of TMP, and increases with increasing TMP.

4.0 Quality Assurance

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.

5.0 Conclusions

The conclusions from this work follow.

- Filter flux increased with increasing transmembrane pressure, agreeing with the Hagen-Poiseuille equation.
- Filter flux decreased with increasing MST concentration, agreeing with the boundary layer model (and other crossflow filtration models).
- No effect of axial velocity on filter flux was observed during the testing. The likely reason for this result is that the operating conditions were not in the range at which axial velocity would have a significant effect on filter flux.

6.0 References

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