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Crossflow Filtration: Literature Review

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EXECUTIVE SUMMARY

As part of the Filtration task EM-31, WP-2.3.6, which is a joint effort between Savannah River National Laboratory (SRNL) and the Pacific Northwest National Laboratory (PNNL), tests were planned to evaluate crossflow filtration in order to the improve the use of existing hardware in the waste treatment plants at both the Department of Energy (DOE) Savannah River Site (SRS) and Hanford Site. These tests included experiments to try different operating conditions and additives, such as filter aids, in order to create a more permeable filter cake and improve the permeate flux.

To plan the SRNL tests a literature review was performed to provide information on previous experiments performed by DOE laboratories, and by academia. This report compliments PNNL report (Daniel, et al 2010), and is an attempt to try and capture crossflow filtration work performed in the past that provide a basis for future testing. However, not all sources on crossflow filtration could be reviewed due to the shear volume of information available. In this report various references were examined and a representative group was chosen to present the major factors that affect crossflow filtration. The information summarized in this review contains previous operating conditions studied and their influence on the rate of filtration. Besides operating conditions, other attempted improvements include the use of filter aids, a pre-filtration leaching process, the backpulse system, and various types of filter tubes and filter coatings. The results from past research can be used as a starting point for further experimentation that can result in the improvement in the performance of the crossflow filtration.

The literature reviewed in this report indicates how complex the crossflow issues are with the results of some studies appearing to conflict results from other studies. This complexity implies that filtration of mobilized stored waste cannot be explained in a simple generic sense; meaning an empirical model develop from one waste-filter combination will more than likely not be applicable to another combination. It appears that filtration performance varies as wide as the range of the types of slurry wastes that exist. However, conclusions can be elicited from existing information so that filter performance can be better understood, and hopefully improved. Those conclusions and recommendations for the planned tests are listed below:

- The rheology and particle size of the substance being filtered affects the rate of filtration, as shown by the various optimum operating conditions.
- Smaller particles can clog the pores or form a tight cake layer and thus significantly reduce the filtration rate.
- High solids concentrations can reduce the overall filtrate flux, as well as the time it takes for the flux to reach a pseudo-steady-state value.
- The formation of the cake is dependent on several conditions, the most important of which is either a critical filtrate flux or transmembrane pressure. Once that critical state is reached, particle deposition can begin to form the layer.
- The thickness of the cake is determined by the shearing forces of the bulk flow across the filter surface.
- Increasing the axial velocity will increase the filtrate flux.
- Reducing slurry viscosity through diluting, e.g., washing, or increasing temperature, e.g., during leaching, increases the filter flux.

- Increasing the transmembrane pressure will increase filtrate flux, but only until a critical TMP value is reached. That critical value is different for different feed materials,solids concentrations, and filter media.
- Filter aids always seem to improve filtration.
- Filter coatings were also tested and the hydrophilic polymer membranes seem to provide an improvement.
- Backpulsing produced mixed results. While it was often used successfully to clear (or partially clear) the filter cake from the surface of the membrane, it generally cannot remove the particles trapped in the pores of the filter, and worse, it seems to accelerate depth fouling.
- The volume of filtrate used by backpulsing to dislodge a filter cake and the frequency of use reduces a filters overall effectiveness.
- After irreversible filter fouling chemically cleaning with acid is necessary. Many chemicals were tested and nitric acid or oxalic acid were shown to be most effective.

Considering the finding from this review the planned crossflow filtration tests should:

- Determine an optimum set of operating conditions that will lead to a more permeable cake that will sustain a good filtrate flux for long periods of operation and result in a relatively clean filtrate stream.
- Reevaluate the need to backpulse by balancing its positive effect to maintain good filtrate flux and its negative effect of accelerating the irreversible situation of depth fouling.
- Determine if there is another method to maintain a good filtrate flux besides backpulsing.
- Evaluate the effectiveness of filter aids to improve filter performance.

TABLE OF CONTENTS

LIST OF TABLES

NOMENCLATURE

Abbreviation	Description
°C	Degree Centigrade
cP	Centipoise
CUF	Cells Ultrafiltration Facility (bench-top crossflow filter used with both radioactively cold and hot wastes)
DOE	United States Department of Energy
DW	Decladding Waste
EDL	Engineering Development Laboratory (part of SRNS/SRNL)
ft	Foot
γ	Shear Rate, sec^{-1} (Rheology Parameter)
gpm	Gallons Per Minute
hr	Hour (Time)
ID	Inside Diameter
in.	inch
L	Liter
M	Molar = Moles / Liter
NPH	Normal Paraffin Hydrocarbon
μ	Dynamic Viscosity, cP, (Rheology Parameter for a Newtonian fluid)
μ_0	Plastic Viscosity, cP, (Rheology Parameter based on the Bingham Plastic Fluid model)
nominal	The word "nominal" for a filter rating is a vague term because its meaning is manufacturer dependent. Further, a "nominal" rating does not give an exact size to a filter medium, but rather an approximation to the expected performance of a filter. In the case of Mott, a nominal rated 0.1-μm filter means that approximately 95% of particles greater than 0.1 μm will not pass the filter
OD	Outside diameter
Pa	Pascal – Unit of Pressure
PNNL	Pacific Northwest National Laboratory
PSD	Particle Size Distribution
psi	Pounds Per Square Inch
psid	Pounds Per Square Inch Differential
psig	Pounds Per Square Inch Gauge
RPP-WTP	River Protection Project – Waste Treatment and Immobilization Plant (Hanford DOE)
s	Second (Time)
SM	Spent Metathesis

SRNS	Savannah River Nuclear Solutions – Principal Contractor managing SRS
SRS	Savannah River Site
τ	Shear Stress, dynes/cm^2, (fluid property response to strain or applied stress)
τ_0	Yield Stress (Shear Stress at Shear Rate = 0), dynes/ cm^2, (Rheology Parameter for a Bingham Plastic Fluid model)
TBP	Tri-butyl Phosphate
TMP	Transmembrane Pressure (the average pressure drop across the thickness of the filter medium – perpendicular to the slurry flow.)
TPB	Tetraphenylborate
TR	(Crossflow Filtration) Test Rig
UDS	Undissolved solids
V	Axial velocity – average velocity axially through the filter tube
XF	Cross Flow

LIST OF USEFUL CONVERSION FACTORS

Symbol	Name	Conversion of Frequently Used Units of Measure						
F	Permeate Flux	1.0 gpm/ft^2	=	58.7 m/day	=	245 cm/h	=	2445 L/h•m^2
TMP	Transmembrane Pressure	1.0 psid	=	6.895 kPa	=	27.7 in.H_2O	=	2.036 in.Hg
V	Axial Velocity	1.0 ft/s	=	30.48 cm/s	=	0.3048 m/s		
μ	Dynamic Viscosity	0.672 lbm/(ft•s)	=	1.0 N•s/m^2	=	1.0 Pa•s	=	1000 cP
ν	Kinematic Viscosity	10.764 ft^2/s	=	1.0 m^2/s	=	3600 m^2/h		

1.0 Introduction

Crossflow filtration is a separation technique for the clarification of liquids and/or the concentration of undissolved particles, depending on the end product requirements. This type of filtration has many practical uses in the chemical, food, and pharmaceutical industries, but is most frequently implemented in water treatment. Unlike dead-end filtration, which runs a feed perpendicular to a filtering medium, the flow of the feed is parallel to the filter media which helps clear away the accumulated particles from the surface of the filter membrane. Though crossflow filtration is intended to be a cake-free method, filter cake can build up on the filter membrane and affect the process either beneficially or detrimentally.

In order to truly understand crossflow filtration, the formation of the filter cake on the membrane must be well understood. As a fluid flow through a pipe, a velocity profile will develop. The velocity in the center of the pipe is largest where there is nothing to hinder its movement. As one nears the walls, however, frictional forces affect the velocity profile and at the surface of the wall, one can assume the velocity is zero due to the no-slip condition. In crossflow filtration, there is another force which impacts the behavior of particles and can impact the no-slip condition, filtrate (liquid) is removed through the porous wall (filter) and this flow is perpendicular to the fluid flowing through the pipe. The flow of filtrate is called flux. As a particle nears this porous wall, its momentum slows and can deposit on the surface. In some cases, the shearing effects of the bulk of the flow can lift the particle away from the wall. However, the flow or physical properties of the substance may be such that the shearing effects are small and the particle can deposit permanently on the wall's surface. As the number of particles depositing increases, particles can begin to stick together and deposit onto one another, thus building up a cake layer, which is referred to as surface fouling. However, due to the presence of the pores, there is another way that a filter fouling can occur when the filter wall is thick enough. When the particles are sufficiently smaller than the pores, they can deposit inside the pores. More and more particles fill the pores creating what is called depth fouling (Murkes and Carlsson, 1988). In both cases, fouling affects the performance of the crossflow filter by reducing the filtrate rate.

Many experiments have been performed in academia and many models have been proposed on what forces cause the particles to deposit and how that affects the formation of the cake layer. One conclusion is that particle deposition occurs when the pressure in the system reaches a critical value and presses the particle to the wall of the tube, allowing cake formation. Based on this model, particles will initially deposit anywhere. As the cake layer thickens, the shearing affects due to flow in the tube become greater and the cake eventually reaches a uniform thickness throughout the tube. However, the cake will first reach a uniform thickness at the inlet of the system. Then that region of equilibrium thickness will slowly move down the tube until the entire cake layer is at equilibrium (Song, 1997). Another conclusion is that for a given crossflow velocity, particle deposition will not occur below a critical flux. Above that flux, particle deposition is significant and particles more easily deposit once some particles are already on the filtering surface by attaching to one another (Li, 1998).

While the thickness of the cake is affected by the shearing effects of the bulk flow, the porosity of the cake is determined by the size and shape of the particles in the feed stream. If the feed stream has a uniform particle size, the porosity of the cake will depend on the particle size. Larger particles tend to form looser cakes while finer particles pack together. However, if the feed has a wide range of particle sizes, then the porosity may vary. Assuming the larger particles deposit first, the finer particles would then deposit between the larger particles, filling the spaces in-between. In general, however, the finer particles tend to deposit more easily on the filter than the larger ones and thus coat the filter first. The fine particles usually cause depth fouling and significantly reduce the porosity and filtration rate. One academic researcher studied the affects of particle shape on cake porosity and discovered that as the shape of the particle departs from spherical, the structure of the filter cake becomes looser (Hwang et al., 1996). However, the cake formation can also be affected by the type of filter membrane employed.

There are two general types of filter-wall construction: symmetrical and asymmetrical. The symmetrical wall has a uniform surface composition throughout. That is, the material and pore structure of the filter does not change throughout the media. An asymmetrical filter wall can have many filtering media. Typically, these filters have a coating on the inside with a smaller pore size than the rest of the filter medium. This coating can help prevent the formation of a cake on the filtering surface or decrease the chances of depth fouling. However, an inner coating is not always necessary for improving filtration. A symmetrical filter can behave like an asymmetrical filter during operation. In many cases, when the filter cake forms on the surface of the filtering medium, it inadvertently creates a coating of its own, known as a precoat. The filter cake will pack down and the spaces between the particles will act like pores for the rest of the feed to filter through.

Understanding how the filter cake is formed is essential to understanding and improving the filtration rate. As the thickness of the cake membrane increases, the flux decreases. When the shear forces on the surface of the cake prevent any further build-up, the flux reaches a pseudo-steady-state value lower than the initial (Murkes and Carlsson, 1988). However, the final flux never truly holds stable at that value and will continue to decrease at a slow rate. Since the final thickness of the cake is determined by the shearing forces of the bulk flow, an increase in the shearing forces will decrease the cake layer and its resistance, thus increasing the flux. This increase in shearing forces is accomplished by increasing the axial velocity through the filter. Another operating parameter used to improve permeate flux is the transmembrane pressure (TMP), which is the pressure difference across the filter media. An increase in TMP will initially produce a greater flux. However, once the TMP reaches a certain optimum value, further increases in the TMP has diminished affects on filtrate flux. In fact, too much TMP can pack the cake down and accelerate depth fouling.

Modifications have been made to the crossflow filtering process in order to improve the permeate flux and reduce fouling to the filter. One such modification is the use of a backpulsing system, where the filtrate is momentarily pushed back through the filter, which is an attempt to remove the cake layer from the surface of the filter tube walls and increase filtrate flux. The backpulse is also used to help dislodge particles that may have settled inside the pores. However, depending on the rheology of the feed stream and the filter medium, the backpulsing system is not always effective (Duignan, 2003). In many cases, when the backpulse dislodges the cake layer, the larger particles are swept along with the rest of the slurry, leaving room for the smaller particles to take their place. This outcome creates a more compact cake and increases the chances of depth fouling. Also, the increase in filtrate flux only lasts for a short period of time. The effectiveness of the backpulsing system is determined by the frequency and length of the backpulse, as well as the amount of filtrate being pushed through the filter or pressure of the backpulse. Of course, by increasing backpulsing frequency the throughput decreases due to the loss of the filtrate that returns to the slurry.

Other adaptations are the addition of filter aids and body feeds, or the reduction of solids that are known to hinder filtration. A filter aid is used as a precoat for the filter to develop a more permeable cake. The body feed is added directly to the feed stream and acts as a flocculant, agglomerating the smaller particles into larger particles that will hopefully form a more permeable cake. Researchers from PNNL focused on washing and leaching processes with caustic solution to remove certain elements from the feed prior to or during the filtration process, e.g., aluminum.

In combination with the current studies on crossflow filtration taking place at SRNL, this report documents a literature review of previously performed experiments. The references covered include work performed within the DOE complex and in academic fields. This report summarizes the how the experiments were performed to show how crossflow filtration is performed or influenced and how the process can be improved.

2.0 Crossflow Filtration Studies

2.1 Previous DOE Crossflow Filtration Studies

Savannah River Nuclear Solutions

Cells Unit Filter

Much of the earlier work for crossflow filtration at SRNS was performed using the Cells Unit Filter (CUF), which is bench-top scale equipment specifically designed to fit inside the Hot Cells to evaluate small volumes (~1 liter) of radioactive feeds. However, non-radioactive simulants were also tested so they could be compared. The rig consisted of a single 24-inch long stainless steel crossflow filter tube with a range of diameters and pore size ratings. A frequently used filter tube has a nominal pore rating of 0.1 μm and an internal diameter of 3/8-inch and a wall thickness of 1/16-inch. The filter and operating parameters for the experiments detailed in this section can be found in Table 2-1. Most experiments using the CUF were performed to evaluate various new processes that could aid crossflow filtration and remove certain elements. One experiment with the CUF tested filtration of a non-radioactive simulant for SRS Tank 48H waste (Nash, 1995). This feed was composed mostly of tetraphenylborate (TPB) and was filtered at 1 and 8 wt% undissolved solids (UDS). For both UDS concentrations tested, filtration rates were obtained between 0.1 and 0.5 gpm/ft^2. The affect of increasing the concentration of monosodium titanate (MST) was also studied. An addition of 0.48 g/L of MST decreased the filtrate flux by 25%.

A Hanford simulant, referred to as, Envelope D, at 3 wt% UDS was also filtered through the CUF. This simulant had solids composed mostly of cadmium, iron, sodium, zirconium, and silicon. Three separate types of tests were performed: one with organics present, one without organics, and a few runs using concentrations from 25 to 2500 mg/L of a 50:50 wt% mixture of Tri-butyl Phosphate (TBP) and Normal Paraffin Hydrocarbon (NPH). The purpose of these experiments was to determine the effects of trace quantities of separable organics such as NPH and TBP (Zamecnik and Baich, 2002). The presence of the NPH/TBP mixture had no effect on flux at any of the tested concentrations. There was a noticeable decline in the filtrate flux initially which was either due to particle degradation or to irreparable changes to the filter membrane that occur over time. Water and nitric acid were used to clean the filters, but cleaning proved difficult due to the presence of the organics. However, certain influences on the process of crossflow filtration itself were also observed. Those effects included operating conditions, the effectiveness of the backpulsing system, and type of cleaning acid to be used.

The effect of operating conditions on the rate of filtration has been a major objective to understand in many experiments. One CUF experiment was done to evaluate a new process called Strontium-Transuranic Precipitation using a radioactive feed (Nash et al., 2000). The process precipitated strontium and permanganate before entering the crossflow filter where the chamber of the CUF was pressurized to 45 psid. The new process performed before filtration allowed the removal of up to 80% iron. A washing process using inhibited water (0.1 M NaOH) caused the filtrate flux to increase with increasing number of washing cycles. The major observation for this experiment was that the filtration rate improved under operating conditions with high TMP and high axial velocity, specifically for the runs at low UDS concentrations. However, the effect of TMP on the filtrate flux was smaller than that of velocity. At high UDS levels, a low TMP and high axial velocity produced better filtration rates.

Table 2-1. Summary of Parameters for all CUF Experiments Performed at SRNS

Reference	Filter	Geometry (type of inner coating)	Length x ID x thickness	Pore Rating (µm)	Axial Velocity Range (ft/s)	TMP Range (psi)	Notes
Nash, 1995	Mott	Symmetrical	24 x 3/8 x 1/16	0.1	3 - 4	5 - 25	MST addition
Nash et al., 2000	Mott	Symmetrical	24 x 3/8 x 1/16	0.1	9 - 15	30 - 70	Strontium-Transuranic Precipitation
Zamecnik and Baich, 2002	Mott	Symmetrical	24 x 3/8 x 1/16	0.1	7 - 15	20 - 60	Effects of trace organics
Poirier et al., 2003	Mott	Symmetrical	24 x 3/8 x 1/16	0.1	-	-	Washing and leaching removal of soluble species
Zamecnik et al., 2003	Mott	Symmetrical	24 x 3/8 x 1/16	0.1	12	50	T = 50ºC
Zamecnik et al., 2004b	Mott	Symmetrical	24 x 3/8 x 1/16	0.1	12	40	Washing and leaching

Those observations were made again in an experiment using actual Hanford waste, AY-102/C-106, and the slurry was initially at 11 wt%. This simulant contained large amounts of Pu-238 and -239, Cs-137, Sr-90, Am-241, Tc-99, Co-60, sodium, iron, and aluminum. The particles had an average diameter between 2.5 and 97.2 µm. It was filtered in the CUF to observe the removal of soluble species during washing and caustic leaching, which was able to remove 40% of the aluminum, 11% of the iron, 60% sodium, 30% Sr-90, 30 % Am-241, approximately 23 % of the Cs-137, and 20 % of Pu-239/240. Only 1 % of the Co-60 was removed but more than 77% of the technicium-99 was removed. The leaching process was performed with 6M NaOH at 85ºC. During filtration, a higher permeate flux was achieved with a higher axial velocity and TMP (Poirier et al., 2003). Other observations included a flux between 0.5 and 1 gpm/ft^2 throughout the runs.

In other experiments, the effects of TMP varied. Two more simulated wastes were tested in the CUF, termed Envelope C AN-102 R1 and R2. The R1 simulant is bimodal with peaks at 4.5 and 13.5 µm. The R2 simulant is also bimodal but with peaks at 2.5 and 10.5 µm. The filtrate flux increased with an increase in axial velocity but was independent of transmembrane pressure. As the UDS concentration increases, the filtrate flux decreased. Also, the flux decreased with time. The project was made to gather data on the performance of the single-tube unit to de-water the simulant precipitate derived from a specific tank (Zamecnik et al., 2003). The temperature was set at 25 and 50ºC. The minimum flux for the R1 simulant was 0.02 gpm/ft^2 but the R2 simulant had and even lower flux. The system was cleaned with a backpulse at 75 psi between runs and 2 M nitric acid.

Proper cleaning of the filters was also studied and involved the testing of various acids or the backpulsing system. One such experiment was used specifically to gather data for a particular tank simulant. This project had to demonstrate operating requirements, throughput for the ultrafiltration system and effectiveness and extent of sludge washing and leaching achievable (Zamecnik et al., 2004b). One run used a pre-evaporated simulant, the other ran the simulant without evaporation. The simulants consisted mostly of iron, sodium, silicon, lead, aluminum and zirconium with large amounts of phosphate, oxalate, sulfate, and carbonate. The mean diameter of the particles is 3.7 μm. The pre-evaporated simulant formed sodium carbonate decahydrate crystals and constantly plugged the tubing. Its filtrate flux was lower than that of the un-evaporated simulant. Various cleaning acids were tried in this experiment, including 1 M nitric acid, 0.5 M oxalic acid, 0.5 M glycolic acid, 1 M nitric acid plus 0.1 M oxalic acid, 1 M nitric acid plus 0.1 M glycolic acid, 1 M nitric acid plus 0.5 M glycolic acid. The preferred cleaning acid combination was oxalic acid followed by nitric acid and inhibited water. The oxalic acid had greatest effect but still required nitric acid to return most of the filtrate flux. Also, the backpulsing system employed had no effect on the evaporated simulant. In the experiment that used the caustic washing and leaching, a solution of 2 M nitric acid was tried but proved insufficient for cleaning in these tests. The 0.5 M oxalic acid, however, had a significant improvement (Poirier et al., 2003).

Pilot Scale Crossflow Filtration Rig (Table 2-2)
Much work has been done with the pilot scale rig consisting of seven parallel tube filters made of 316L stainless steel. Like the experiments done with a CUF, certain conclusions about crossflow filtration proved similar throughout the projects.

Table 2-2. Summary of Parameters for all Pilot Scale Experiments Performed at SRNS

Reference	Filter	Filter Design	Length x ID x wall thickness (in.)	Pore Rating (μm)	Axial Velocity Range (ft/s)	TMP Range (psi)	Notes
Duignan, 2000a	Mott	Symmetrical	40 x 3/8 x 1/16	0.1	-	15 - 70	Flow rate: 10-50 gpm; backpulsing
Duignan, 2000b	Mott	Symmetrical	40 x 3/8 x 1/16	0.1	-	15 - 70	Flow rate: 10-50 gpm; backpulsing
Duignan et al., 2001	Mott	Symmetrical	40 x 3/8 x 1/16	0.1	3..3 – 15	17 – 70	Backpulsing
Duignan, 2003	Mott	Symmetrical	90 x ½ x 1/16	0.1	7 - 15	20 – 60	Backpulsing
Duignan et al., 2004a	Mott	Symmetrical	90 x ½ x 1/16	0.1	9 – 13	10 – 50	Backpulsing
Duignan et al., 2004b	Mott	Symmetrical	90 x ½ x 1/16	0.1	11 – 12	10 – 40	-
Duignan et al., 2004c	Mott	Symmetrical	90 x ½ x 1/16	0.1	11 – 12	10 – 40	-
Duignan et al., 2005	Mott	Symmetrical	90 x ½ x 1/16	0.1	9 - 15	10 – 50	High Temp. T = 45ºC

One experiment was performed on a non-radioactive simulant of Envelope A with insoluble solids between 0.5 and 5 μm in particle size at a concentration of 0.5 wt%. Envelope A simulant is characterized by the presence of mostly aluminum, sodium, nitrites, and nitrates. The entrained solids in the slurry are mostly composed of chromium oxide and sodium oxalate (Duignan, 2000a). In general, it was observed that the filtrate flux was strongly affected by velocity but only weakly by TMP. However, an increase in either results in an increase of filtrate flux. The results indicated an axial velocity should be maintained at 12 ft/s or higher and the TMP kept between 40 and 55 psid. The use of backpulsing was also tested at a frequency of 30 minutes. While there was a momentary increase in filtrate flux after the pulse the effect was ultimately ineffective. Cleaning with 1M nitric acid was necessary to restore performance of the filter.

Another experiment was performed the same year with almost identical operating conditions. However, a different non-radioactive simulant for Envelope C was used (Duignan, 2000b). The undissolved solids with this slurry are mostly sodium oxalate and sodium carbonate monohydrate. The slurry was prepared at 0.5 wt% UDS and the size of the particles ranged between 1-2 μm for smaller particles and between 5 and 10 μm for larger particles. This simulant also behaves differently, having properties of a Bingham fluid at higher solids loadings. However, the general results and recommendations were the same. That is, higher axial velocities and transmembrane pressures lead to higher filtrate fluxes, but velocity has more effect on the flux than TMP. The engineers suggested operating at velocities of 12 ft/s or higher and at transmembrane pressures of 30 to 55 psid. The use of the backpulsing system was again ineffective for improving the filtration rate. The filtrate flux, however, could only be maintained between 0.01 and 0.02 gpm/ft^2 for this simulant, which was an order of magnitude lower than for the previous simulant. Using 1M nitric acid was effective to clean the filter.

A third experiment was performed using the same seven-tube pilot test rig, but with a third type of simulant and different operating conditions. The simulant was non-radioactive and contained insoluble solids between 0.5 and 50 μm in size that consisted mostly of sodium oxalate and chromium oxide (Duignan et al., 2001). The slurry had a density of 1.23 g/mL and a viscosity close to 3 cP when prepared at a solids loading of 5 wt%. Even with these changes, the results were still similar. The effects of velocity and TMP on filtrate flux, as well as the lack of effectiveness of backpulsing, are the same as for the other two experiments. The optimum flow conditions obtained during these experiments occurred at a velocity of 12.1 ft/s and a TMP of 42 psi for a low concentration of insoluble solids. These three experiments show that while specific operating conditions are dependent on the type of stimulant, the influence of the TMP and axial velocity on the filtrate flux is the same.

In a fourth experiment, the rig was modified but the results reflected those observed in the previous experiments. The non-radioactive simulant was mostly made of sodium carbonate, sodium oxalate, and sodium phosphate (Duignan, 2003). This simulant was prepared between 0.8 and 1.6 wt% UDS and contained particles ranging in size from 1 and 10 μm. At a solids loading of 1 wt%, the slurry was measured to have a density of 1.29 g/mL and a viscosity of 4 cP. An increase in velocity by a factor of 2 increased the flux by 20%. The TMP, once again, had no significant influence on the filtrate flux. It was recommended that the velocity should not fall below 11 ft/s or the TMP below 30 psid. The use of backpulsing was only effective up to 5 wt%, after which it seemed to increase the rate of depth fouling. Nitric acid at a concentration of 2 M was used and restored the performance of the filter.

The first campaign of a four-campaign project termed Semi-Integrated Pilot Plant was proposed to collect data on the performance of an ultrafilter to dewater simulants and to estimate the removal efficiency for soluble species to minimize waste oxides. The test rig for these campaigns is the same as that used in the previous experiment but seemed to produce conflicting results on the effectiveness of the backpulsing system and cleaning acid (Duignan et al., 2004a). The slurry used for this campaign had strong concentrations of aluminum, iron, manganese, sodium, phosphorus and silicon, with large amounts of nitrates, nitrites, carbonates, phosphates, sulfates, and some alumina silicates. The particle size distribution was bi-modal with peaks at 0.55 and 1.5 µm. The most effective axial velocity was 12 ft/s, but even still the average permeate for during filtration was only 0.008 gpm/ft^2. When the filter became plugged with highly concentrated solids, recovery was partially successful with backpulsing. An initial cleaning with 2 M nitric acid was insufficient to return the filter to pre-filtration conditions and the acid cleaning had to be repeated two more times to return recover a clean condition.

The second campaign included studies on all four WTP Pretreatment Facility unit operations, including the crossflow filter, and carried similar results to the previous campaign. The recycle from the first campaign was used for all tests in this campaign (Duignan et al., 2004b). This recycle feed has a similar composition to the original slurry, but contains more potassium and less phosphates, sulfates and alumina silicates. Particles sizes average between 5 and 6 µm, and the slurry is initially concentrated to 3 wt% UDS. The average permeate flux for this campaign was 0.011 gpm/ft^2. The turbidity of the permeate increased with time (days), indicating that solids were precipitating out of solution, but filtering reduced turbidity to acceptable levels. Cleaning with 2 M nitric acid and caustic rinses increases the flux by one order of magnitude but could not return the filter to pre-filtration conditions. The third campaign used the recycle from the second campaign as well as the same operating conditions (Duignan et al., 2004c). The feed stream for this campaign is similar to that in the second campaign, starting at 3 wt% UDS, but contains free hydroxides. The average permeate flux was 0.011 gpm/ft^2. The same cleaning solution was used and provided the same results. However, it was suggested that submerging the filter in 0.1 M NaOH solution over an extended period of time may improve filtrate flux.

In the fourth and final campaign, the Mott filters were replaced with 90-inch GKN filter tubes because WTP decided to purchase filters from GKN. Even though the filters tubes came from a different manufacturer the specification was the same as for the Mott tube and they indeed appeared to look the same. Because of this the filter performance was expected to be the same, but this fact had to be demonstrated. Besides testing the new filters to compare to the Mott performance WTP decided to see the effect of a higher temperature; therefore, some tests were conducted at 45ºC and 12 ft/s (Duignan et al., 2005). The GKN filters did indeed performed similar to that of the Mott filters under steady state conditions. The average, temperature-adjusted permeate flux was 0.011 gpm/ft^2. For the higher temperature trials the permeate flux improved. At 19 wt% UDS, 40 psid TMP, and a slurry velocity of 12 ft/s, the permeate flux improved almost 75% at the higher temperature. Finally, during the filter cleaning the filter performance between the second and third acid cleaning steps did not result in a significant improvement as was realized during the Campaign I filtration. The difference in Campaign I was that the waste stream did not contain the recycle streams, which may have helped removed or diluted some of the iron and aluminum compounds that are more difficult to dissolve. The amount of acid and length of contact time is definitely dependent on the waste being treated.

Miscellaneous Crossflow Filter Tests (Table 2-3)
The In-Tank Precipitation facility at SRS has performed several filter tests at different scales with both simulated and radioactive wastes (McCabe, 1995). Among the various pore sizes studied, the 0.5 µm filter tube was determined to produce sufficiently decontaminated filtrate while allowing for a higher flow rate than the 0.2 µm filter. An axial velocity was determined with an axial velocity of 3 to 3.5 ft/s with a backpulse frequency of five minutes. The pump type also had an effect on the performance of the filter. The best filtration results came with the use of a low-shear pump, such as a diaphragm pump. When the filter tube bundles were placed in series, the first bundle had the highest flux. The change in filtrate flux throughout these bundles indicates that a lower differential pressure and higher slurry concentration found in downstream bundles will result in lower filtrate flux. Oxalic acid was successful in cleaning the filters, and has been

proven to clean filters fouled by tetraphenylborate precipitates, sodium titanate, and simulated sludge solids.

Table 2-3. Summary of Parameters for Other Experiments Performed at SRNS

Reference	Filter	Geometry (type of inner coating)	Length x ID x thickness	Pore Rating (µm)	Axial Velocity Range (ft/s)	TMP Range (psi)	Notes
McCabe, 1995	Several	Symmetrical	varied	0.2, 0.5, 2	varied	varied	-
Morrissey and Dworjanyn, 1992	-	Symmetrical	-	-	2 - 10	14 - 45	-
Nash and Siler, 1997	Mott	Symmetrical	-	0.1	2 - 10	15 - 45	Filtrate flux maintained at 12 ft/s
	Graver						
Poirier et al., 2008	Mott	Symmetrical bench scale	24 x 3/8 x 1/8	0.1 and 0.5	6 - 14	15 - 41	Actinide Removal Process
	Mott	Symmetrical pilot scale	120 x 5/8 x 1/8	0.1 and 0.5	6 - 26	12 - 64	
Steimke, 2004	Du Pont	Asymmetrical (sintered titanium oxide)	- x 5/8 x -	0.6	6 - 14	20 - 40	T = 50ºC; backpulsing
Walker, 1997a	Mott	Symmetrical	48 x ½ x -	0.5	1.8 – 5.4	10 - 40	-
	Graver	Asymmetrical (titania)	30 x 5/8 x -	0.1			
Walker, 1997b	Mott	Symmetrical	- x ½ x -	0.5	1.6 – 3.6	15 - 25	Filter aid
Zamecnik et al., 2004a	Mott	Symmetrical bench scale	24 x 3/8 x -	0.1 and 0.5	11 - 12	40 - 50	-
	Mott	Symmetrical pilot scale	90 x ½ x -	0.1 and 0.5			
Freeman, 2008	GE AG, Dow XLE, LE RO	Asymmentric	varied	varied	varied	150	Filter coatings
Siler et al., 1991	Norton Ceraflo™	Asymmetrical (aluminum oxide ceramic coating)	- x – x 0.0006	0.2	-	19 - 30	Inorganic and biological fouling

The Cladding Removal Waste tests were performed on several different separation techniques, but the best option was the crossflow filter (McCabe, 1995). Three non-radioactive and uranium-spiked Zirflex stream simulants were chosen for these tests, including Decladding Waste (DW) and Spent Metathesis (SM). Mott filter tubes with different dimensions were also tested: an 18 x 0.25 inch single tube, 8 of those tubes in series, and a 60 x 0.25 inch seven-tube bundle. Using the single-element filter, the non-radioactive DW gained a filtrate flow rate of 0.1 gpm/ft^2 at a velocity of 12 ft/s and a backpulse frequency of 5 minutes. The TMP was not specified, but given a range between 15 and >45 psi. The non-radioactive SM achieved the same filter flux but produced poor filtrate quality. The radioactive DW achieved a filtrate flux between 0.18 and 0.26 gpm/ft^2 wit ha velocity of 14 ft/s and a backpulse frequency of 5 minutes. Using the seven tube bundle, the non-radioactive DW gained a flux of 0.18 gpm/ft^2 with a feed pressure of 35 psi and a velocity of 5 ft/s.

Neutralized Current Acid Waste tests were performed on simulated wastes that were first centrifuged twice before entering the crossflow filter in order to prevent rapid filter plugging. Mott filters with 0.2 and 0.5 μm pore ratings were used with either a centrifugal pump or a diaphragm pump. In general, both filters produced poor filtrate clarity, but the 0.5 μm filter tube produced filtrate fluxes 20% higher than the 0.2 μm filter (McCabe, 1995). Tests with the diaphragm pump produced higher flux and greater filtrate clarity, but reduced the crossflow velocity and increased the TMP. The flux rate with the 0.5 μm filter and the centrifugal pump reached 0.13 to 0.22 gpm/ft^2.

Various filter tubes from different manufacturers were compared and tested for improved filtration rates, but the crossflow results were similar with no significant improvement. One experiment compared the performance of two filters, manufactured by Mott and by Graver (Nash and Siler, 1997). Two simulants were also compared: Strontium/Transuranic (Sr/TRU) and Envelope A at low solids loading of 0.15 and 0.65 wt% UDS. Using the Sr/TRU, no filtrate was produced with the Mott filter when velocity was below 7.5 ft/s. The same thing occurred with the Graver filter for the velocity was below 7 ft/s. During the runs with the 0.15 wt% Envelope A, flux was determined to be a function of TMP until approximately 20 psid while cross flow velocity continued to have insignificant effects. This stayed true for the runs with higher concentration. In all test runs, no depth fouling occurred. It was concluded that a higher axial velocity prevented cake formation and improved flux. Also, the permeability of the simulants decreased with TMP for a given velocity.

In another experiment, crossflow filtration was evaluated as effective solid/liquid separation technology for treating Hanford wastes. Both a stainless steel Mott filter and a stainless steel Graver filter with a layer of titania were used in the evaluations (Walker, 1997a). The simulant used for both filter tubes was prepared at 0.05 and 8 wt% UDS and contained boehmite and gibbsite. The Graver filter had a higher flux for the 0.05 wt% slurry but the Mott filter had a higher flux for the 8 wt% slurry. At 8 wt% UDS, the Mott filter produced a filter cake indicating that the flux was dependent on axial velocity. In all cases, the author claimed that filtrate flux was a function of TMP due to the presence of depth fouling. Cleaning of the filter to 80% of its original filtrate flux was possible using 2 wt% NaOH and 2 wt% oxalic acid.

Four Du Pont crossflow tube filters, each with a 30 μm thick layer of sintered titanium oxide on the inside surface, were tested using backpulsing with a simulant of SRS Tank 49H waste. The solids loading used was 10 wt% UDS and the slurry was composed mostly of sodium tetraphenylborate and sodium hydroxide (Steimke et al., 1994). During some of the initial runs, bacterial growth was observed that caused filter blockage. However, it was suggested that ionizing radiation from actual waste may prevent growth. In general, the filtrate flux seemed dependent on TMP until a threshold pressure was reached. Above that pressure, the flux then becomes dependent on axial velocity and increases with increasing axial velocity. Results implied that with a higher filtrate fluxes or TMP, or lower axial velocities, a thicker cake layers. Backpulsing was included in the cleaning operations, but it was not always effective in restoring filter performance by itself. The researchers recommended that higher axial velocities be used to reduce the need for backpulsing. However, cleaning was only effective with acid and a solution of 1 wt% oxalic acid followed by 1 wt% sodium hydroxide was used. The most successful cleaning came from using 1 wt% hydrogen peroxide and sodium hydroxide.

One project, funded by DOE but performed at a university (Freeman, 2008), explored the possibility of using specialized hydrophilic coatings on crossflow filters to reduced fouling. Acrylic acid, 2-hydroxyethyl acrylate or poly(ethylene glycol) acrylate was copolymerized with poly(ethylene glycol) diacrylate to form highly hydrophilic, crosslinked hydrogel coatings. These materials were placed on the filter media by either coating or grafting. Grafting proved to be the better method; however, the membrane was not fully coated due to shrinkage of the polymer. The feed slurry tested was composed of oil (n-decane or n-dodecane) and a surfactant (DTAB, SDS) in a 9 to 1 ratio. The results stated that water sorption increased with increasing co-monomer chain length of the coatings. The new copolymers performed better due to greater affinity to water and low concentrations of PEG diepoxide maximized the water flux. Salt rejection was found to be a strong function of pH between values of 3 and 10. Also, a higher flow through the filter reduced the formation of filter cake and increased the salt rejection.

Some projects sought to compare results between bench and pilot scale experiments. Crossflow ultrafiltration tests were conducted at SRNL to support the design of the Hanford Waste Treatment Plant and to validate the use of a simulant. The validation was done by comparing actual radioactive waste to simulated waste filtration test data using a CUF. Once a simulant was validated it was used in both bench-scale CUF and the SRNL pilot-scale facilty (Zamecnik et al., 2004a). All experiments used the same simulant prepared at 1.5 wt% UDS. The particles were mostly around 2 or 3 μm in size and were composed mostly of nitrides, sulfides, sodium and carbonate. The pilot scale tests used seven filter tubes 90-inches in length, while the CUF had only a single 24-inch tube. An unexpected observation was that the slurries with a smaller average particle size filtered better. In general, a higher axial velocity gave a higher flux but the flux was not dependent on TMP, but it was inversely proportional to the slurry viscosity. The side-by-side comparison showed that pilot scale filtrate flux was always lower than that of the CUF. The difference fluxes between the CUF and pilot scale data were attributed to differences in wall shear stress, entrance and exit effects due to the different length tubes, and the different pressure boundary conditions from the multitude arrangement to the single tube.

A new process called the Actinide Removal Process was used to remove strontium and select actinides from radioactive liquid waste. Test data from the bench scale was compared with operating experience at the pilot scale using seven parallel filter tubes. The bench scale tests used actual waste containing a sludge-to-MST ratio of 0.6 to 0.55. The pilot scale tests used a simulant with a sludge-to-MST ratio of 6:5.5, as well as actinides and strontium (Poirier et al., 2008). The simulant was comparable to the actual waste. In all tests, the 0.1 μm rated filter had better filtrate flux values than the 0.5 μm rated filter. The flux increased with TMP and axial velocity but decreased with increasing concentration. For the bench scale experiments at 1.4 wt% UDS, the flux averaged 0.135 gpm/ft^2 (0.1 um filter) and 0.082 gpm/ft^2 (0.5 um filter). For the pilot scale at 4.5 wt% UDS, filtrate flux averaged 0.133 gpm/ft^2 (0.1 um filter) and 0.086 gpm/ft^2 (0.5 um filter).

An entire experiment was dedicated to determine the best cleaning method for filters. Slow flushing and high velocity axial recirculation were tested with various chemicals, such as oxalic acid with caustic and M-Pyrol with KOH/Triton. A single, short filter tube was used to filter a simulated slurry that contained sodium tetraphenylborate and a surfactant. All cleaning runs were then performed at 6 ft/s and 30 psi. Slow flushing technique with oxalic acid and caustic proved to be less effective than recirculation with the same chemicals. The least effective cleaning was performed with M-Pyrol and KOH/Triton recirculation. The best filtrate flux recovery after cleaning was obtained using oxalic acid and caustic recirculation (Morrissey and Dworjanyn, 1992).

Filter aids were also tested for their ability to improve crossflow filtration rates in the separation of ORNL wastes. A filter aid of bentonite clay, also known as thixogel, was added to some of the 6.8 wt% UDS waste sludge (Walker, 1997b). A filtrate flux between 0.01 and 0.06 gpm/ft^2 was obtained for runs with and without filter aid. Without the filter aid, the filtrate flux is a function of axial velocity and TMP, indicating the presence of a filter cake and depth fouling. With the filter aid, the flux was independent of axial velocity and TMP, indicating no filter cake or fouling. In cleaning three solutions were tried, including 2 wt% NaOH and 2 wt% oxalic acid. However, the most effective cleaning solution was 2 wt% nitric acid.

Fouling of the crossflow filters due to inorganic colloidal precipitates and, more importantly, biofouling has been of major concern in operation of the filters (Siler et al., 1991). It has been suggested that the presence of Al(III), Fe(III), and Si(IV) in the feed results in the formation of colloidal precipitates as small as 15 µm in the neutralized filter feed. Experiments were performed at the laboratory-scale, 2 gpm pilot-scale, and 40 gpm semiworks scale with both single lumen and multiple lumen (19 single-lumen feed channels) filters. A feed was prepared with 5.7 mg/L Fe(IlI), 5.0 mg/L Si(IV), 2.7 mg/L Al(III), and 1500 mg/L NANO3 to test fouling by inorganic precipitates. Bacterial fouling tests were run with an addition of *Enterobacter cloacae* and *Pseudomonas sp*., which were added to the standard simulant concentrations of 105 to 107 bacteria/mL. The addition of aluminum nitrate at concentrations of 15 to 30 mg/L to the feed prior to neutralization improved filtration by controlling the adsorption of the precipitates. The addition also controlled to adsorption of bacteria. Tests to kill the bacteria contaminants were successful with the addition of 0.03 M nitric acid or 0.25 M NaOH, boiling in water, or exposing to UV-ozone, but those techniques were ineffective at alleviating the fouling problem. The process of cleaning the filters was improved by the addition of the aluminum nitrate but also by efficient backpulsing. The backpulse system was operated at 40 to 90 psi every 2 to 3 minutes, but the pressure used was more important than frequency for improving the cleaning. Other methods of cleaning included using 2% oxalic acid at 40 to 60ºC, but preferably 2% NaOH with 100 mg/L OCl^-.

Flocculant Testing with a Dead-End Filter

Various flocculating agents were tested for their ability to improve crossflow filtration by agglomerating feed particles and creating a more permeable cake. Flocculants made by Cytex, HX-200, -300, -400, and -2000, were used to prepare slurries ranging from 0.15 to 2.1 wt% flocculated solids. The agglomerated particle sizes were between 1 and 30 mm. Flocculants called Alclar® 600, 662 and W23 were used to make between 0.1 to 2 wt% flocculated solids. The resulted flocculated particle sizes were greater than or equal to 2 mm. The simulant was a mixture of 0.6 g/L sludge, 0.55 g/L MST and 5.6 M supernate with a density of 1.25 g/mL and 1.15 g/L UDS (Martino et al., 2001). A Mott dead-end filter with a pore rating of 0.5 µm was used but the overall purpose of these tests was to determine which flocculant was best suited for later use in a crossflow filter. The filtration rate was maintained at 0.25 gpm/ft^2. Overall, the Cytex HX series performed better than the Alcar® flocculants and provided a lower TMP increase. HX-200, -300, and -400 had similar results but the HX-400 was more consistent. The HX-2000 was most effective at lower doses while HX-400 was better at higher doses. However, the HX-2000 had the narrowest flocculant concentration range and did not flocculate as many solids as the others. Backpulsing was used during the experiment when the TMP reached 30 psi and proved effective for the given tests.

Similar flocculating agents were used in another series of tests using Tank 40H simulated sludge with MST. Several hydroxamated amines (Cytex HX series) and Alclar® were first tested in dead-end filters to determine which flocculants would be used in the following crossflow filter runs. The 115 mL dead-end filters had a pore rating of 0.45 µm and the crossflow filter had a pore rating of 0.5 µm. During preparation of the slurries with flocculating agents, the agglomerates formed using the hydroxamated amines were larger and settled faster than those produced with the Alclar® agents (Poirier, 2001). The best agglomerates were produced with HX-200 and HX-2000. Filtration was significantly improved in the dead-end filter tests but only by 30% at most in the crossflow tests using the best flocculants. This decrease in improvement from the dead-end to crossflow filters was attributed to the breakdown of flocculants by the shearing of the pump.

Hanford – Pacific Northwestern National Laboratory

<u>Bench-scale Tests Using a Cell Unit Filter (Table 2-4)</u>
A series of experiments with a Cells Unit Filter (CUF) similar to the one used at SRNS, was performed to evaluate filtration on different feeds subject to a leaching process. One slurry consisted mostly of tributyl phosphate and was initially at a solids loading of 10 wt% UDS (Edwards et al., 2009). The leaching process in this experiment was carried out at temperatures ranging from 40 to 80ºC with sodium concentrations between 0.25 and 3 molar. The leached slurry was also fed through the filter at 60ºC and 20 wt% UDS. The particle sizes of the feed were between 0.2 and 750 µm. The resulting filtrate flux was between 0.01 and 0.028 gpm/ft^2. During the leaching process, between 50 and 80% of the phosphorus was removed before heating. The sodium concentration and temperature had little effect on P dissolution but largely affected gibbsite dissolution. Aluminum dissolution was best at the higher temperatures. During the filtration of the feed, increases in TMP or axial velocity resulted in increases in filter flux only when the concentration of UDS was above 20wt%. Ultimately, changes in permeate viscosity during leaching had larger affects on the filtrate flux than the undissolved solids concentration.

Table 2-4. Summary of Parameters for all Experiments Performed at PNNL

Reference	Filter	Geometry (type of inner coating)	Length x ID x thickness	Pore Rating (µm)	Axial Velocity Range (ft/s)	TMP Range (psi)	Notes
Edwards et al., 2009	Mott	Symmetrical	24 x ½ x 1/8	0.1	8 - 16	20 - 63	Leaching: 0.25 – 3M NaOH and T= 40 – 80ºC
Lumetta et al., 2009	Mott	Symmetrical	24 x ½ x 1/8	0.1	8 - 18	20 - 60	Leaching: 1- 5M NaOH and T= 40-100ºC
Fiskum et al., 2009	Mott	Symmetrical	24 x ½ x 1/8	0.1	-	-	Leaching: 3.7M NaOH and T= 60ºC
Russell et al., 2009	Mott	Symmetrical	24 x ½ x 1/8	0.1	9 - 17	20 - 60	Leaching and Oxidative Leaching; T= 45ºC
Shimskey et al., 2009	Mott	Symmetrical	24 x ½ x 1/8	0.1	9 - 15	20 - 60	Oxidative Leaching
Brooks, 2000	Mott	Symmetrical	24 x ½ x 1/8	0.1	9 - 15	30 - 70	Leaching and washing

The leaching process was also performed on bismuth phosphate sludge and bismuth phosphate saltcake (Lumetta et al., 2009). Both feeds contained phosphate, aluminum, chromium, fluorine, oxalate, iron and sulfate, but the saltcake had higher concentrations of all elements except the iron. The sludge particles were between 0.3 and 100 µm with a mean size around 6 µm. The saltcake particles were between 0.3 and 20 µm. During the filtration runs, the filtrate flux was found to be dependent on the TMP but not the axial velocity for both high and low solids concentrations for both slurries.

The leaching technique was repeated for a feed of ferrocyanide tank sludge at 5 wt% UDS composed mostly of phosphate and iron. The particle size of the feed was between 0.2 and 170 μm. However, for this experiment, the leaching was only performed with a 3.7M free hydroxide matrix at 60ºC (Fiskum et al., 2009). It was observed that an increase in TMP proportionally increases the filter flux. The impact of axial velocity on flux decreases at concentrations greater than 10 wt%. Conversely, the undissolved solids concentrations began to have an affect above 15wt%. Chromium and aluminum in the form of gibbsite dissolved before leached slurry reached 40ºC and phosphorus in insoluble solids was not dissolved by caustic leaching. Ultimately, the conclusion was that caustic leaching of this sludge may not be considered very effective to improve the filtration rate.

The leaching process was modified in another experiment. This time, the solution was filtered initially, and then again after and addition of 0.01 M NaOH. Then an oxidative leaching process was tried using a 1:1 molar ratio solution of 1 M permanganate at a temperature of 100ºC, followed by a an addition of 0.01 M NaOH (Russell et al., 2009). The simulant being used contained boehmite, gibbsite, oxalate, chromium and inter solids. Particles sizes for the slurry ranged from 0.5 to 60 μm. The crossflow filter was operated at temperatures of 25 and 45ºC. Small amounts of inert fines less than 1 micron in size were added for a fouling test but did not have a significant impact on filtrate flux. In general, the filter flux increased with temperature during the initial filtration. The leaching process increased the flux while the oxidative leaching had little effect.

One more variation of the leaching process was tried on actual REDOX sludge and S-saltcake waste. REDOX simulant was mostly composed of aluminum with a little chromium while the saltcake was mostly chromium and oxalate. Both simulants had particles sizes between 2 and 200 μm. The oxidative leaching used a 1:7 molar ratio of Mn:Cr and a solution of 0.09 M NaOH (Shimskey et al., 2009). The REDOX sludge's filtration flux was dependent on solids concentrations, axial velocity and TMP, but only at solids concentrations above 12 wt%. The leaching initially decreased the flux, but then significantly increased it once slurry washing began. The S-saltcake had lower flux values and faster flux decay over time. The researchers decided to add REDOX sludge to increase undissolved solids concentration to increase the filtrate flux. Again, caustic leaching initially decreased the flux but significantly increased flux after washing. Oxidative leaching had no significant effects on the filter flux but after washing the axial velocity effects became more significant and flux no longer decayed with time. The leaching was effective to remove most of the chromium from slurry but did not improve filter flux. Flux improvement was best realized from a decrease in slurry viscosity through washing.

Another leaching process was performed on a radioactive simulant composed mostly of aluminum, iron, silicon, sodium, thorium, and zirconium, with nitrates, chromates and nitrites. The radioactivity was mostly from the presence of cesium, with some uranium and plutonium also present. The particles are initially between 0.2 and 50 μm in size. The simulant was washed seven times with 0.01 M NaOH and then leached with 3 M NaOH at 85ºC (Brooks, 2000). The washing steps removed almost all soluble anions and 91% of the sodium. The first caustic leaching removed 91% aluminum so more leaching was not required. In general, the total mass of solids was reduced by 50% during the washing and leaching steps. For highly concentrated slurries, the filtrate flux was measured between 0.0095 and 0.0172 gpm/ft^2. A higher axial velocity improved the flux but the changes in TMP had no effect. The optimum operating conditions were 70 psi and 15 ft/s. For low concentrations, the flux was between 0.38 and 0.83 gpm/ft^2. A higher axial velocity again improved filtrate flux but higher TMP increase the rate at which the filter fouling occurred. The optimum filter operating conditions were determined to be

30 psi and 15 ft/s. When the crossflow filter was operated at 85ºC during leaching, the flux was three to four times higher than those at 25ºC, which was probably due to the change in slurry viscosity. The author noted that a solution of 1 M nitric acid was needed for sufficient cleaning.

Other Experiments
Experiments were performed at the bench scale level to determine the ability of the crossflow filtration system to handle slurries at different solids loadings (Geetings et al., 1996). Various models were studied to determine which best predicted the filtration of the different loadings. A six inch long, 0.5 μm Mott filter with an inside diameter of ½ inch was used. The temperature of the system was maintained between 28 and 38ºC. Axial velocity ranged from 3 to 9 ft/s and TMP went from 5 to 35 psig. Two simulants, Hanford Tank S-107 sludge and Hanford Tank C-107 supernatant, contained boehmite and gibbsite and were tested with solids loading of 0.05, 1.5 and 8 wt% UDS. The mean particle diameter was about 0.4 μm. The flux for the higher solids loadings was observed to be a linear function of axial velocity but independent of TMP above 20 psid. The lower solids loadings were independent of axial velocity but varied linearly with TMP. The use of a smaller pore size filter media for future experiments was suggested to prevent depth fouling. A frequency of 30 minutes for backpulsing was not enough to restore filtrate flux and 2 wt% oxalic acid was necessary for cleaning.

Other Laboratories

Oak Ridge National Laboratory
An experiment was created to develop, design, and deploy solid/liquid separation (SLS) technology (Kent et al., 2001). Two 5-ft long Mott filters with a pore rating of 0.5 μm and a ¾-inch outer diameter were placed in series. The filter tubes were operated at axial velocities between 6 and 10 ft/s with TMP from 10 to 35 psig. The filtrate flux obtained was between 0.02 and 0.1 gpm/ft^2. No information was provided as to the type of slurry being used in this experiment. Filtrate production was significantly affected by TMP and less so by velocity, indicating no filter cake production. With the presence of a filter cake, axial velocity had significant affects but not the TMP. Backpulsing at a frequency of once per hour was employed and had no significant affect other than a momentary increase of filtrate flux. Dilute nitric acid and sodium hydroxide seemed sufficient for cleaning.

Simulated and radioactive wastes from the Melton Valley Storage Tanks were tested using three filter tubes models: 0.2 and 0.5 μm Mott stainless steel filters, a 0.5 μm CARRE with ZOSS membrane and a CARRE filter with proprietary membrane (McCabe, 1995). The simulant feed contained insoluble solids prepared at 11.2 wt% and composed of various metal hydroxides and bentonite clay. During filtration, the Mott filters produced higher fluxes than the CARRE filters. The filter with the proprietary membrane actually degraded during filtration. The 0.5 Mott filter gave the best flux of 0.039 gpm/ft^2 under operating conditions with a TMP of 19 psi and an axial velocity of 4.5 ft/s. In fact, an axial velocity of 4.5 ft/s produced a higher filtrate flux than using a velocity of 9 ft/s. A backpulse frequency of 10 minutes was also used to aid filtration. The radioactive waste obtained an optimum flux of 0.11 gpm/ft^2 with an axial velocity of 4.5 ft/s and a TMP of 19 psi while using a 0.5 μm Mott filter. The filtration flow rate decreased over time until the TMP was increased to 41 psi, and then remained constant for the rest of the test. The CARRE-ZOSS filter was also tested on the radioactive simulant but produced a filtration rate less than 10% of that observed with the Mott filter.

Idaho National Engineering and Environmental Laboratory
An experiment focused investigation into alternative, state-of-the art filtration technologies to facilitate the strontium and actinide removal process, which involved comparing various filters

(Mann et al., 2004). The types of filters and their dimensions can be found in Table 2-5. A simulated sludge with MST was prepared for all filters at solids loadings of 0.06, 0.29 and 4.5 wt%. The operating conditions were the same for all filters. Feed flow was maintained at approximately 9 gpm, with axial velocities between 6 and 9 ft/s and TMP from 30 to 40 psig. The 0.1 µm Pall and 0.07 µm Graver achieved the highest average steady state fluxes for all solids loadings, nearly 13 to 21 percent higher at solids loadings of 0.29 and 4.5 wt%. They also had the lowest variability, possibly due to the fact that small particles present in the solution were unable to penetrate the ceramic layer, producing surface filtration. Filters without the asymmetrical coating are susceptible to depth filtration. In general, insoluble solids concentration and axial velocity had the strongest effect on filter flux. A solution of 0.5 M oxalic acid was used to clean the filters but the 1 M nitric acid solution and waters fluxes cleaned more. There was still an overall reduction in flux due to fouling.

Table 2-5. Summary of Filter Tube Parameters used in Mann et al., 2004

Filter Manufacturer	Geometry (type of inner coating)	Wall Thickness (inches)	Pore Rating (µm)
Mott	Symmetrical	0.062	0.1
Mott	Symmetrical	0.062	0.5
GKN	Asymmetrical (sintered metal)	0.079	0.1
Graver	Asymmetrical (sintered titania)	0.055	0.07
Pall	Symmetrical	0.035	0.8
Pall	Asymmetrical (sintered zirconia)	0.035	0.1

2.2 Previous non-DOE Crossflow Filtration Studies (Table 2-6)

Academic investigations on crossflow filtration focus primarily on theory and model development; some that include experiments. To date none of these investigations give a comprehensive description that can be used to accurately predict filter performance for a specific filter situation (Ripperger and Altmann,. 2002; Daniel et al., 2010) However, these models and experiments that may help to better understand this technology and therefore a few of the many studies are included below.

Experiments were conducted to observe profiles of fluid velocity and particle concentration in turbulent flow in order to calculate the rate of particle deposition. Two parallel plates, with only the stainless steel lower plate containing pores, were used as the filtering system. The feed for this system was 0.5 wt% light calcium carbonate slurry with particles ranging in size from 1 to 20 µm and having an average diameter of 7.2 µm (Lu et al., 1993). The flow profile was determined to have a turbulent core with turbulent eddies on either side, and laminar sublayers at the wall of the tube. Particle concentration on the filter increased along the filter length until the axial velocity profile was fully developed. Then, the concentration profile also becomes constant. An increase in velocity decreased the rate of particle deposition and generated higher shear stress on the cake surface. For a given velocity, the rate of particle deposition decreased with decreasing permeation rate. An increase in shear stress on the cake surface had more significant effects in improving rate of crossflow filtration than increases in the bulk velocity.

Table 2-6. Summary of Parameters for all Experiments Performed outside of the DOE

Reference	Geometry and Composition	Dimensions of Filter (inch)	Filtration Area (in^2)	Flow Profile	Axial Velocity Range (ft/s)	TMP Range (psi)	Notes
Lu et al., 1993	2 stainless steel parallel plates	0.276 x 56.69 x 2.76	7.75	turbulent	1.9 – 3.7	-	-
Chang et al., 1995	Durapore membrane with 9 channels	15.2 x 1.5 x (0.091-0.249)	-	laminar	1.6 - 6	5.8	Electrolytic concentration = 0 – 0.1M; T=30ºC; pH=5.4
Wang and Song, 1999	polyolefin	6.7 x 1.5 x 0.59	1.24	-	-	0.1 – 0.2	shear stress = 70, 140, and 280 s^{-1}
Hwang and Wang, 2001	two-parallel plate microfilter	0.018 x 3.94 x 0.19	0.62	laminar	0.66 - 2.6	-	Computer simulation
Xiao-Zhang, 2002	40 x 40 mm square Twilled Butch Weave	39.4 x 2.9 x 0.63	10 µm (pore rating)	-	0.52 - 3.8	-	Filter aid
Hwang et al., 2005	2-parallel plate microfilter made of mixed cellulose ester	0.039 x 2.2	0.1 µm (pore rating)	laminar	0.3 - 2	-	-
Li, 1998	Anopore membrane of anodized aluminum	1.57 x 4.3 x 0.08	0.02 µm (pore rating)	-	0.16 - 3.28	-	-
Gésan-Guiziou, 2002	Tubular Kerasep membrane	72 long with 0.18 ID (7 channels)	0.1 µm (pore rating)	-	1.6 - 1.9	0 - 23	pH=7 T=50ºC

A mathematical model was developed to predict quasi-steady-state permeate flux after investigating steady-state permeate flux under various operating conditions. The feed was composed of suspended spherical polystyrene latex at 50 ppm with particle diameters of 0.303, 1.02 and 2.967 µm. The pore sizes were varied throughout experiments, being 0.1, 0.2, 0.45 and 0.65 µm. The mathematical model assumes the presence of only certain forces acting on the particle: the tangential force, normal drag force, lateral lift, shear-induced force, double layer force, gravity, and the Brownian diffusion force. The model accurately predicted quasi-steady-state flux under various operating conditions. The quasi-steady-state is defined as when the filtrate flux starts "exhibiting linear decrease in time and cake thickness does not change" (Chang et al., 1995). That quasi-steady-state permeate flux increased with increasing wall shear rate and membrane pore size but decreased with increasing electrolyte concentration. At low shear rates, the flux was larger for smaller particles. The opposite effect proved true at high shear rates.

In an effort to study flux decline, membrane fouling was assumed to be a dynamic process from a state of non-equilibrium to one of equilibrium (Song, 1997). Using this perception, a mathematical model was developed for this process. It was decided that cake formation occurs when applied pressure is greater than a critical pressure in the non-equilibrium operation of the filter. The pores would be blocked and a cake layer will form to absorb the excess pressure. The cake forms until it reaches an equilibrium thickness and then the filtrate flux will also reach an equilibrium state. A mechanistic model was developed to describe the change from a state of non-equilibrium to equilibrium. The model suggests that when an applied pressure is higher than a certain critical pressure, the whole channel will be at non-equilibrium and particles will deposit everywhere on the filter tube. An equilibrium region forms at inlet first when the cake layer reaches an equilibrium thickness. Then the front of the equilibrium region will move downstream as filtration continues. In the beginning, the average flux in the entire filter is mainly determined by the non-equilibrium flux because that state is present in most of the filter. As the front reaches the end of filter, the fraction of the filter in the non-equilibrium phase becomes small. Thus the average flux is determined by the equilibrium region. Basically, the flux curve follows the flux in the non-equilibrium region before reaching the constant equilibrium flux. These effects were physically observed experimentally (Wakeman, 1994). The mathematical model was developed and used to predict the time it takes to reach steady-state as well as the steady-state flux.

The theory of fouling dynamics was compared and verified with experiments under various conditions (Wang and Song, 1999). An ultrafiltration membrane was made with pores determined to be small enough not to let any feed particles through. The feed was a suspension of spherical MP-1040 silica colloids with a mean size of 0.12 μm diluted in ultra pure water. It was prepared at concentrations of 0.2, 0.5 0.8 and 1 wt% solids. The pressure of the slurry throughout the experiments was held at 3 psi. It was determined that a higher feed concentration results in a higher fouling rate and thus a smaller filtrate flux. Cake formation occurred when applied pressure was greater than a critical pressure. Cake continues to form until it reaches an equilibrium thickness and then the filtrate flux also reaches an equilibrium state. A previously developed mechanistic model was used to describe the change from non-equilibrium to equilibrium. The model suggests that when an applied pressure is higher than critical pressure, the whole channel is at a non-equilibrium state with particles depositing everywhere. The equilibrium region forms at the inlet first when the cake reaches equilibrium thickness. Then the front of the equilibrium region moves downstream as filtration continues. The researchers hypothesize that the flux in the equilibrium region is a function of shear rate while the non-equilibrium region is independent of shear stress.

The migration and deposition of binary particles was simulated by a computer. An analysis of the forces exerted on the particles was performed and the effects of operating conditions on packing structure and cake porosity were discussed (Hwang and Wang, 2001). The feed consisted of polymethyl methacrylate as spherical particles with mean diameters of 0.25 and 0.8 μm. It was observed through the simulation that packing of large particles resulted in a more compact filter cake than that of small particles due to larger normal drag forces exerted on the large particles. An even more compact cake was observed when the small particles filled in spaces between the larger particles. The packing porosity increased with increasing crossflow velocity due to increasing friction angle between particles. A larger filtration rate was associated with a lower packing porosity. For a given mixing fraction of particles, a looser packing was observed with a larger crossflow velocity or a smaller filtration rate.

The effect of particle size distribution on cake properties and performance of crossflow microfiltration was examined. The affects of various operating conditions on size and filtration rates at pseudo-steady state was also discussed (Hwang et al., 2005). The feed was a solution of 0.1 wt% polymethyl methacrylate at a pH of 7 and with a density of 1210 kg/m^3. The feed particles were spherical in shape, ranging from 0.2 to 20 μm in size with an average diameter of 6.2 μm. The forces affecting particle deposition were found to be inter-particle forces for submicron particles and inertial lift force, drag force, and gravitational force for micron particles. Furthermore, the authors found that particle size plays a major role in particle deposition. An increase in crossflow velocity reduces the probability of particle deposition while decreasing particle size distribution. Pseudo-steady state was achieved when the cake formation was completed. The cake mass increased and porosity decreased with increasing filtration pressure.

Another theory for the cause of particle deposition was proposed (Li, 1998). For a given velocity, there is a critical flux below which particle deposition is negligible but above which deposition is significant. An Anopore membrane was used to directly observe the particle deposition. The feed streams solids were either yeast cells with a mean diameter of 5 µm or latex beads with mean diameters of 3, 6.4 and 11.9 µm. Both feeds were prepared at concentrations between 0.05 and 0.057 vol%. The critical flux increased with increasing velocity and particle size. At higher axial velocities, the smaller particles deposited more easily on the membrane and in doing so, reduced the flux. At or above the critical flux, a slower velocity had depositing particles that appeared stagnant and smaller particles that were more mobile. A higher velocity showed more rapid deposition and release from the membrane, producing what looked like a flowing cake layer. It was also observed that particles deposited more easily when there were other particles already on the membrane. Specifically, particles can more easily attach to one another than on the surface.

The theory of the critical flux value was later revisited and more parameters were explored. A new Kerasep membrane was used for these experiments (Gésan-Guiziou, 2002). The feed was a suspension of latex stabilized by surfactants with an average particle diameter of 0.19 µm and a narrow size distribution. The testing focused on what happens above the critical flux. Below that condition, the deposited mass of particles and cake thickness increased with TMP until the limiting flux was reached. The limiting flux is a steady-state flux that occurs when the cake reaches a uniform thickness. The porosity varied slightly with TMP but was always low. This low porosity indicated a possible compression of the deposited particles and a high packing density. A higher wall shear stress had a higher critical flux because less deposited on the membrane surface. The critical flux decreased with increasing latex concentration up to a point then remained stable for further increases in concentration. An increase in suspension conductivity for given concentration lowered the critical flux. Throughout the experiments, the surfactants affected the deposition characteristics and the critical flux

Various filter aids were also tested for their ability to improve filtration rates. One study examined the effects of operating parameters on precoat layer build-up under crossflow conditions (Xiao-Zhang et al., 2002). A model was also made to characterize structure and filtration resistance of precoat layer built up under different operating conditions. A filtration pressure of 14.5 psi was maintained. The feed was not identified but the filter aid was diatomite. It was prepared at concentrations between 200 and 5000 g/m^3. Filtration rates decreased with increasing crossflow velocity. The precoat layer of diatomite also decreased while the total resistance increased. Though the precoat was less thick at higher velocities, it consisted of finer particles that increased resistance. The filtration rate decreased with increasing filter aid concentration and the size of deposited particles decreased with decreasing particle concentration.

Suspension removal and ultrafiltration processes were both performed to optimize the operating conditions for ultrafiltration, specifically, the pumping rate (Liu and Wu, 1998). The suspension removal system used a cellulose acetate with a 0.2 µm pore rating as the filtering media while the ultrafilter used a hollow-fiber membrane of polysulphone with a filtration area of 775 in^2. The feed was erythromycin with a pH of 8. The feed was aided by a filter aid of 3 to 5% (w/v) $ZnSO_4$. The pumping rate to be used was based on three methods: minimum cost, maximum flux and optimum operating pressure. In order to achieve minimum cost, the preferred pumping rate was 23.2 ft/s. For maximum flux, the preferred operating conditions are an inlet pressure of 22 psi and an outlet pressure of 13 psi. The optimum operating pressure is obtained with a TMP of 17.4

psi and pumping rate of 12.6 ft/s. An increased temperature proved to be beneficial to filtration because it reduced the viscosity of the feed. The chosen temperature in this experiment was 40ºC.

Various body feeds were experimented with to assess the applicability of different flocculants as filter aids in MBR operation and explore how filter aids affect mixed liquor properties to alleviate membrane fouling (Ji et al., 2008). As such, a MBR system along with a dead-end microfiltration cell was decided upon for the test rig. The membrane for the system was a flat sheet of polyvinylidene. The feed had a viscosity between 1.35 and 1.54 cP and was composed of glucose, NH_4Cl peptone, yeast extract, KH_2PO_4, and $NaHCO_3$. The particles of the feed stream were between 143 and 167 µm. TMP was varied from 0.73 to 10 psi while the filtrate flux was maintained at 20 and 40 L/m^2h. The body feeds were polymeric ferric sulfate (PFS), aluminum sulfate and a natural polycationic polysaccharide called Chitosan. The fouling rates of PFS and Chitosan were 7 times less than those measured without filter aid addition. This is most likely due to the fact that the filter aids promoted macromolecules in supernatant to coalesce into large aggregate and alleviate membrane fouling. That fact, in turn, lead to reduced pore blocking and gel layer resistance. The specific cake resistance was reduced by increasing the mean flocculant size and decreasing the fractal dimension of flocculants. The concentration and molecular weight distribution of macromolecules in supernatant was determined to be important in gel layer formation and loss of membrane porosity. In general, the addition of Chitosan and PFS significantly increased the total porosity of the cake layer. Cleaning with milli-Q water was sufficient to restore the filtration system.

3.0 Conclusions

Based on the limited review of the references described herein, certain conclusions can be made about the current experiences of crossflow filtration performance and filter cake formation:

- The rheology and particle size of the substance being filtered affects the rate of filtration, as shown by the various optimum operating conditions.
- Smaller particles can clog the pores or form a tight cake layer and thus significantly reduce the filtration rate.
- High solids concentrations can reduce the overall filtrate flux, as well as the time it takes for the flux to reach a pseudo-steady-state value.
- The formation of the cake is dependent on several conditions, the most important of which is either a critical filtrate flux or transmembrane pressure. Once that critical state is reached, particle deposition can begin to form the layer.
- The thickness of the cake is determined by the shearing forces of the bulk flow across the filter surface.
- Increasing the axial velocity will increase the filtrate flux.
- Reducing slurry viscosity through diluting, e.g., washing, or increasing temperature, e.g., during leaching, increases the filter flux.
- Increasing the transmembrane pressure will increase filtrate flux, but only until a critical TMP value is reached. That critical value is different for different feed materials, solids concentrations, and filter media.
- Filter aids always seem to improve filtration.
- Filter coatings were also tested and the hydrophilic polymer membranes seem to provide an improvement.

- Backpulsing produced mixed results. While it was often used successfully to clear (or partially clear) the filter cake from the surface of the membrane, it generally cannot remove the particles trapped in the pores of the filter, and worse, it seems to accelerate depth fouling.
- The volume of filtrate used by backpulsing to dislodge a filter cake and the frequency of use reduces a filters overall effectiveness.
- After irreversible filter fouling chemically cleaning with acid is necessary. Many chemicals were tested and nitric acid or oxalic acid were shown to be most effective.

Considering the finding from this review the planned crossflow filtration tests should:

- Determine an optimum set of operating conditions that will lead to a more permeable cake that will sustain a good filtrate flux for long periods of operation and result in a relatively clean filtrate stream.
- Reevaluate the need to backpulse by balancing its positive effect to maintain good filtrate flux and its negative effect of accelerating the irreversible situation of depth fouling.
- Determine if there is another method to maintain a good filtrate flux besides backpulsing.
- Evaluate the effectiveness of filter aids to improve filter performance.

References

Brooks, K. P. et al., 2000. "Characterization, Washing, Leaching, and Filtration of C-104 Sludge." Technical Report PNWD-3024, Battelle, Richland, Washington.

Chang, D.J., F.C. Hsu, and S.J. Hwang, 1995. "Steady-state permeate flux of cross-flow microfiltration." *Journal of Membrane Science. Vol. 98*, pp. 97-106.

Daniel, R.C., R.W. Shimskey, P.P. Schonewill, and R.A. Peterson, 2010, "A brief review of filtration studies for waste treatment at the Hanford Site," Technical Report PNNL-20023.

Duignan, M.R., 2000a, "Final Report: Pilot-Scale Cross-flow Filtration Test -Envelope A + Entrained Solids," Technical Report BNF-003-98-0221, Savannah River Site, Aiken, SC.

Duignan, M.R., 2000b, "Pilot-Scale X-Flow Filtration Test-Env C + Entrained Solids+Sr/TRU," Technical Report BNF-003-98-0226, Savannah River Site, Aiken, SC.

Duignan, M., 2003, "Final Report: Pilot-scale Cross-flow Ultrafiltration Test using a Hanford Site Tank 241-AN-102 Waste Simulant," Technical Report WSRC-TR-2003-00204, Savannah River Technology Center, Aiken, SC.

Duignan, M.R., C.A. Nash, and P.S. Townson, 2001, "Pilot-scale cross-flow filtration test for a radioactive waste stimulant with high solids loading and sodium molarity," *4th International Conference on Multiphase Flow*, New Orleans, LA.

Duignan, M.R. et al., 2004a, "Interim Report: RPP-WTP Semi-Integrated Pilot Plant – Campaign I," Technical Report WSRC-TR-2004-000201, Savannah River Site, Aiken, SC.

Duignan, M.R. et al., 2004b, "Interim Report: RPP-WTP Semi-Integrated Pilot Plant – Campaign II," Technical Report WSRC-TR-2004-000478, Savannah River Site, Aiken, SC.

Duignan, M.R. et al., 2004c, "Interim Report: RPP-WTP Semi-Integrated Pilot Plant – Campaign III," Technical Report WSRC-TR-2004-000565, Savannah River Site, Aiken, SC.

Duignan, M.R., J.R. Zamecnik, and M.R. Williams, 2005, "Final Report: RPP-WTP Semi-Integrated Pilot Plant," Technical Report WSRC-TR-2005-00105, Savannah River Site, Aiken, SC.

Edwards, M. K. et al., 2009, "Characterization, Leaching, and Filtration Testing for Tributyl Phosphate (TBP, Group 7) Actual Waste Sample Composites," Technical Report PNNL-18119, Pacific Northwestern National Laboratory, Richland, Washington.

Fiskum, S. K. et al., 2009, "Characterization, Leaching, and Filtration Testing for Ferrocyanide Tank Sludge (Group 8) Actual Waste Sample Composites," Technical Report PNNL-18120, Pacific Northwestern National Laboratory, Richland, Washington.

Freeman, B. D., 2008, "Novel Fouling-Reducing Coatings for Ultrafiltration, Nanofiltration, and Reverse Osmosis Membranes," Technical Report DE-FC26-04NT15547, University of Texas at Austin, Austin, TX.

Geeting, J.G.H., and B.A. Reynolds, 1996, "Bench-Scale Cross Flow Filtration of Tank S-107 Sludge Slurries and Tank C-107 Supernatant," Technical Report PNNL-1137, Pacific Northwest National Laboratory, Washington.

Hwang, K.J., Y.S. Wu, and W.M. Lu, 1996. “The surface structure of cake formed by uniform-sized rigid spheroids in cake filtration.” *Powder Technology, Vol. 87,* pp.161-168.

Hwang, K. J. and Y.S. Wang, 2001. “Numerical Simulation of Particle Deposition in Cross-flow Microfiltration of Binary Particles.” *Tamkang Journal of Science and Engineering. Vol. 4*, pp. 119-125.

Hwang, K. J., Y.L. Hsu, and K.L. Tung, 2005. “Effect of Particle Size on the performance of cross-flow microfiltration.” *Advanced Powder Technol. Vol. 17,* pp. 189-206.

Ji, J., *et al*, 2008. “Enhancement of filterability in MBR achieved by improvement of supernatant and floc characteristics via filter aids addition.” *Water Research, Vol. 42,* pp.3611-3622.

Kent, T.E., J.H. Wilson, and J.R. Farmer, 2001, “Development and Development of a full-scale cross-flow filtration system for treatment of liquid low-level waste at Oak Ridge National Laboratory," Technical Report ORNL/TM-2000/27, Oak Ridge National Laboratory, Oak Ridge, TN.

Liu, C. and X. Wu. 1998. “Optimization of Operation Parameters in Ultrafiltration Process.” *J. Biotech., 66*, pp. 195-202.

Lu, W.M., K.J. Hwang, and S.C. Ju, 1993, “Studies on the Mechanism of Cross-flow Filtration,” *Chemical Engineering Science, Vol. 48*, pp. 863-872.

Lumetta, G.J. et al., 2009, “Characterization, Leaching, and Filtration Testing for Bismuth Phosphate Sludge (Group 1) and Bismuth Phosphate Saltcake (Group 2) Actual Waste Sample Composites,” Technical Report PNNL-17992, Pacific Northwestern National Laboratory, Richland, Washington.

Mann, N.R., R.S. Herbst, et al., 2004, "Alternative Ultrafiltration Membrane Testing for the SRS Baseline Process," Technical Report INEEL/EXT-04-01933, Idaho National Engineering and Environmental Laboratory Idaho Falls, Idaho.

Martino, C.J. et al., 2001, “Flocculating, Settling, and Decanting for the Removal of Monosodium Titanate and Simulated High-Level Waste Sludge from Simulated Salt Supernate.” Technical Report WSRC-TR-2001-00413, Savannah River Technology Center, Aiken, SC.

McCabe, D.J., 1995. “Evaluation and Ranking of the Tank Focus Area Solid Liquid Separation Needs.” Technical Report WSRC-TR-95-0337, Savannah River Site, Aiken, SC.

Morrissey, M.F. and L.O. Dworjanyn, 1992, "Initial Technical Basis for Late Washing Filter Cleaning," Technical Report WSRC-RP-92-925, Savannah River Technology Center, Aiken, SC.

Murkes, J. and C.G. Carlsson, "Crossflow Filtration." Johns Wiley & Sons Ltd: New York, 1988.

Nash, C.A., 1995, "Filtration of Tank 48H Contents with a Cells Unit Filter," Technical Report WSRC-TR-95-0483, Savannah River Technology Center, Aiken SC.

Nash, C.A., H.H. Saito, and W.R. Wilmarth, 2000, "Strontium-Transuranic Precipitation and Crossflow Filtration of 241-AN-201 Large C," Technical Report WSRC-TR-2000-00506, Savannah River Technology Center, Aiken, SC.

Nash, C.A. and J. Siler, 1997, "Filtration of Nonradioactive Hanford Waste Simulants," Technical Report SRTC-BNFL-016, Savannah River Technology Center, Aiken, SC.

Poirier, M., 2001, "Improving the Filtration of Sludge/Monosodium Titanate Slurries by the addition of Flocculants," Technical Report WSRC-TR-2001-00175, Savannah River Technology Center, Aiken, SC.

Poirier, M.R. et al., 2003, "Filtration, Washing, and Leaching of a Hanford AY-102/C-106 Sample," Technical Report WSRC-TR-2003-000240, Savannah River Site, Aiken, SC.

Poirier, M.R. et al., 2008, "Development of a Crossflow Filter to Remove Solids from Radioactive Liquid Waste: Comparison of Test Data with Operating Experience – 9119," Technical Report SRNL-STI-2008-00492, Savannah River National Laboratory, Aiken, SC.

Ripperger, S. and J. Altmann, 2002, "Crossflow microfiltration – state of the art," Separation and Purification Technology, 26, pp. 19-31.

Russell, R.L. et al., 2009, "Development and Demonstration of Ultrafiltration Simulants," Technical Report PNNL-18090, Pacific Northwestern National Laboratory, Richland, Washington.

Shimskey, R.W. et al., 2009, "Filtration and Leach Testing for REDOX Sludge and S-Saltcake Actual Waste Sample Composites," Technical Report PNNL-17965, Pacific Northwest National Laboratory, Richland, Washington.

Siler, J.L., M.R. Poirier, D.J. McCabe, and T.C. Hazen, 1991, "Fouling of Ceramic Filters and Thin-Film Composite Reverse Osmosis Membranes by Inorganic and Bacteriological Constituents," Technical Report WSRC-MS-91-350, Savannah River Laboratory, Aiken, SC.

Song, L. 1997. "Flux Decline in Crossflow Microfiltration and Ultrafiltration: Mechanisms and Modeling of Membrane Fouling." *J. Mem. Sci., 139*, pp. 183-200.

Steimke, J.L., C.M. Hart, and S.E. Sponseller, 1994, "Results from Tests of Du Pont Crossflow Filter," Technical Report WSRC-TR-94-0332, Savannah River Technology Center, Aiken, SC.

Wakeman, R.J., 1994, "Visualization of cake formation in crossflow microfiltration: Part I: Effects of particle and pore size," Trans. I. Chemical Engineering 72, pp. 530-540.

Walker, W., 1997a, "Hanford Underground Storage Tank Waste Filtration Process Evaluation," Technical Report WSRC·TR·97·00353, Savannah River Site, Aiken, SC.

Walker, W., 1997b, "Oak Ridge National Laboratory Melton Valley Storage Tanks Waste Filtration Process Evaluation," Technical Report WSRC-TR-97-00354, Savannah River Technology Center, Aiken, SC.

Wang, L. and L. Song. 1999. “Flux Decline in Crossflow Microfiltration and Ultrafiltration: Experimental Verification of Fouling Dynamics.” J*ournal of Membrane Science, Vol. 160*, pp. 41-50.

Xiao-Zhong Li, H. Hess, and W. Höflinger, 2002, “Influence of operating parameters on precoat layers built up under crossflow condition.” *Separation and Purification Technology. Vol. 31*, pp. 269-280.

Zamecnik, J. and Baich, M.A., 2002, “Evaluating the Effects of Tri-butyl Phosphate and Normal Paraffin Hydrocarbon in Simulated Low-Activity Waste Solution on Ultrafiltration.” Technical Report WSRC-TR-2002-00108, Savannah River Site, Aiken, SC.

Zamecnik, J. et al., 2003, “AN-102 Simulant Sr/TRU Precipitation and Ultrafiltration.” Technical Report WSRC-TR-2003-00056, Savannah River Site, Aiken, SC.

Zamecnik, J. et al., 2004a, “Filtration of Hanford SiteTank 241-AN-102 Waste Sample with Alternate Sr/TRU Precipitation Conditions at Bench and Pilot Scales," Technical Report WSRC-MS-2003-00756, Savannah River Site, Aiken, SC.

Zamecnik, J. et al., 2004b, “Tank 241-AY-102 Simulant Development, Ultrafiltration, and Washing.” Technical Report WSRC-TR-2003-00547, Savannah River Site, Aiken, SC.

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