EVALUATION OF SPIRAL WOUND REVERSE OSMOSIS FOR FOUR RADIOACTIVE WASTE PROCESSING APPLICATIONS

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ABSTRACT

A pilot-scale spiral wound reverse osmosis rig was used to treat four significantly different radioactive waste streams, three of which were generated at the Chalk River Laboratories at AECL. These streams included:

1. A chemical/decontamination (CD/DC) waste stream which is routinely treated by the plant-scale membrane system at CRL;
2. Reactor waste which is a dilute radioactive waste stream (containing primarily tritium and organic acids), and is an effluent from the operating reactors at AECL;
3. An ion exchange regenerant waste stream which contains a mixture of stream {1} (CD/DC), blended with secondary waste from ion exchange regeneration;
4. Boric acid simulated waste which is a by-product waste of the PWR reactors. This was the only stream treated that was not generated as a waste liquid at AECL.

For the first three streams specified above, reverse osmosis was used to remove chemical and radiochemical impurities from the water with efficiencies usually exceeding 99%. In these three cases the "permeate" or clean water was the product of the process. In the case of stream {4}, reverse osmosis was used in a recovery application for the purpose of recycling boric acid back to the reactor, with the concentrate being the "product ". Reverse osmosis technology was successfully demonstrated for the treatment of all four streams.

Prefiltration and oxidation (with photocatalytic continuous oxidation technology) were evaluated as pretreatment alternatives for streams {1}, {2}, and {3}. The results indicated that the effective crossflow velocity through the membrane vessel was more important in determining the extent of membrane fouling than the specific pretreatment strategy employed.

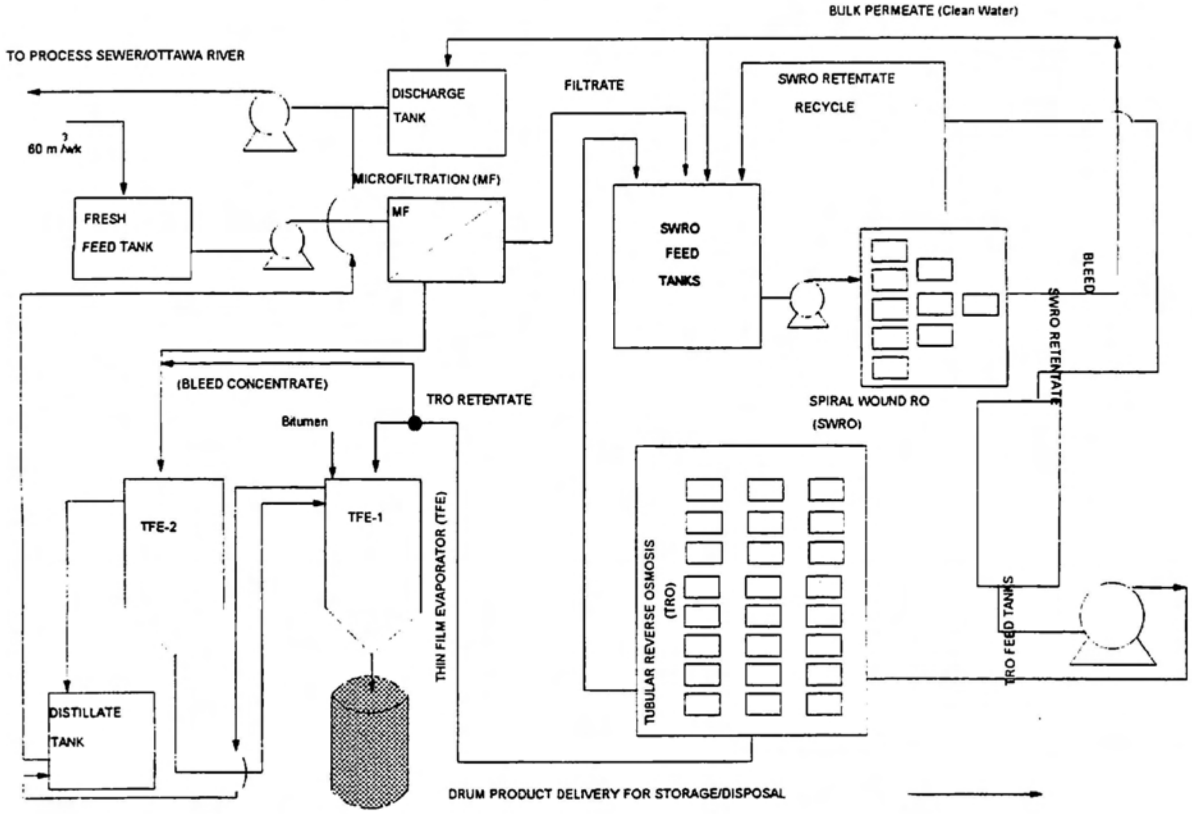
LIQUID RADWASTE PROCESSING

Treatment processes traditionally used to purify low-level radioactive waste (LLRW) liquids that result from operation of nuclear power plants have not utilized membrane separation techniques and have considered them to be novel (1). There have been few applications of reverse osmosis systems in nuclear power plants, although the technology has improved dramatically. Feed characterization, system chemistry control, system configuration and membrane selection are key to establishing successful reverse osmosis treatment. Data to permit appropriate and satisfactory design were not available to predict with confidence that a membrane-based technology is appropriate for nuclear-power-plant effluent treatment.

This paper examines the application of reverse osmosis technology for the processing of four significantly different aqueous radioactive waste streams. Comparisons will be made between the performance of the plant-scale reverse osmosis system used for routine liquid waste processing at AECL (2), with a smaller pilot-scale system, whose primary function is for experimental development work and for optimization studies.

Fig. 1

Integrated Plant-scale System for Aqueous Radwaste Processing.



The plant-scale system (Figure 1) is set up in a tapered 5-3-1 vessel configuration, where each vessel contains the equivalent of 5.5 forty inch diameter vessels. In the 5-3-1 configuration there are five vessels in parallel in the first stage, followed by three vessels in parallel in the second stage, and a single vessel in the third stage. Retentate (or waste concentrate) from each preceding vessel is the feed to the next stage. There are no inter-stage booster pumps in the system which means each successive stage operates at lower pressure than the preceding one. The crossflow through the plant-scale system is maintained at 40 L/min/vessel. The pilot-scale system contains two forty inch vessels and operates at a variable crossflow ranging from 30 to 60 L/min/vessel. In this study the crossflow velocity was set at 55 L/min/vessel.

Another important difference between the two systems is the mode of operation employed. In the plant-scale system a "feed and bleed" type processing strategy is used, where retentate is continuously recirculated back to the feed tank, while a bleed stream is purged away. Fresh filtrate from the MF system is mixed in with the recirculated concentrate stream. This mode of operation leads to a feed tank concentration which approaches an equilibrium value close to the target volumetric recovery of 85% fairly rapidly. In contrast, the pilot-scale system operates in the batch volume reduction mode, where the tank is volume reduced to a final volumetric recovery of 85%. In this processing strategy the membranes are not exposed to a concentrated solution until near the very end of the processing campaign, representing about 20% of the total processing time. Observations in this report will show that this latter mode of operation may decrease the likelihood of premature membrane fouling, although more rigorous experimentation would be required to confirm this.

SPIRAL WOUND REVERSE OSMOSIS FOR WASTE PROCESSING

Reverse osmosis (RO) is a technology that is well established for the production of potable water from brackish water or seawater. It has been successfully used to produce high-purity water for the electronics, pharmaceutical, and power industries. Because of its ability to remove ionic impurities, particulates and colloids, organics, microorganisms and pyrogenic material from water, RO has attained a prominent role in water purification (3).

In RO treatment, feedwater containing dissolved and suspended solids is pumped into the system at a desired feed pressure greater than the osmotic pressure. The feed stream is pumped into a pressure vessel containing one or more membrane elements connected in series. The feedwater then flows into the brine channels between the membrane sheets. These feed channels are composed of a plastic netting which breaks up the flow into small turbulent areas above the membrane surface.

Immediately above the membrane surface a concentration boundary layer forms whose thickness depends on feedwater ionic strength, particulate level, and flow in these small turbulent areas. The water and ions are transported by a solubility-diffusion process to the permeate water carrier. The purified water or permeate is recovered at atmospheric pressure. The remaining water, dissolved solids and particulates form the reject stream. The pressurized concentrate or retentate is dropped to atmospheric pressure through a back pressure regulating valve, immediately downstream of the system.

The performance of an RO membrane is usually described in terms of permeate flow, or "flux" and contaminant rejection efficiency. Permeate flux refers to the amount of flow across the membrane per unit area, at a particular operating pressure and dissolved solids concentration. The flow of water across the membrane is proportional to the effective pressure (applied pressure minus the osmotic pressure of the solution). Increasing the applied pressure will increase the permeate flow without increasing the solute flow.

Rejection is the relative change in contaminant concentration from the feed stream to the permeate stream. RO membranes are not absolute barriers, and some small percentage of the solute (typically about 0.5%) does pass through the membrane. The amount of solute transport is a function of the membrane type and is proportional to the differential concentration across the membrane.

RO membrane types can be broadly classified as cellulosic or noncellulosic. Cellulose acetate membranes are still widely used because of their resistance to fouling and their low cost. They are, however, easily damaged by bacterial attack and have relatively low rejection efficiencies. Noncellulosic membranes, such as the Filmtec SW30HR membranes used at CRL, have a wider pH range and exhibit high solute rejection efficiencies.

There are four types of RO membrane module configurations: spiral wound, plate and frame, hollow fibre, and tubular. In each design, maximum turbulence is important to minimize the phenomenon of concentration polarization. Spiral wound or hollow fibre membranes are the most economical, if large volumes of water are to be treated. The SWRO configuration achieves a large specific surface area per unit volume, which is typically 1000 m^2/m^3 . This can be compared to 165 m^2/m^3 for plate modules and 335 m^2/m^3 for tubular modules. In the design of a SWRO system, a flat membrane is formed around fabric spacers and is closed on three sides. The open side terminates at a perforated product water tube. The membrane edge, with an external spacer for the feedwater stream, is rolled spirally around the product tube and is then installed in a pressure vessel. The feed stream flows axially through the channels between the spiral windings. Water permeates through the membrane and flows radially inside the membrane envelope towards the product tube.

The ratio of permeate volume to feed volume in a RO system is referred to as volumetric recovery. To achieve high volumetric recoveries (up to 85%), it is often necessary to stage the concentrate stream or recycle the concentrate for reprocessing.

Membrane Fouling

Membrane fouling is regarded as the most critical issue affecting the successful implementation of RO for the treatment of aqueous waste streams containing substantial quantities of both chemical and radioactive contaminants. It is necessary for the process engineer to develop an appreciation for the root causes and symptoms of membrane fouling before effective pretreatment strategies can be implemented in the field. Adequate pretreatment technologies are critical to minimize the loss of throughput due to membrane fouling.

The phenomenon of RO membrane fouling can best be described as any mechanism that reduces the water transport per unit area. This is usually thought to occur by accumulation of materials on the membrane surface or in the membrane material by diffusion. Reduction of flow through and over the RO membrane can also be affected by compaction of the membrane and fouling of the materials of construction used in the brine channel and permeate water carrier.

Rationale for Pretreatment

The RO membranes that are commercially available are generally not robust enough to operate directly on typical feedwater streams. For instance, the Filmtec guidelines for elements suggest a maximum feed turbidity of 1 NTU and a maximum SDI of 5. This has been confirmed by four years of operating experience at AECL. Feedwaters usually contain components that can adversely affect the performance and lifetime of the membrane system. Therefore, the performance of an RO system will only be as good as the system used to pretreat the water before it enters the system.

Every RO system includes some level of pretreatment designed to:

(1) extend the lifetime of the membranes;
(2) prevent fouling of the membranes and;
(3) maintain the performance (rejection and recovery).

The extent of pretreatment required will depend on several factors including:

(1) the type of membrane modules;

(2) the composition of the feed stream, and;
(3) the desired performance of the system.

Feedwater pretreatment is essential to avoid membrane fouling. Fouling refers to the irreversible flux decline with time from the trapping or deposition of some material within the RO device or on the membrane surface.

Role of Crossflow MF System

Fouling of an RO system can be reduced with an effective upstream filtration plant. In crossflow filtration (also called tangential flow or inertial filtration), pressure drives only part of the feed through the medium; the remaining feed flows tangentially along the surface of the medium, continuously sweeping particles from the medium's surface back into the feed. Generally, crossflow filters are operated as surface filters and have pores that are smaller than the particles to be removed. By using a microfiltration membrane as the separation medium, particles in the 0.1 to 10 mm range can be removed. The primary role of the crossflow MF system is to remove suspended solids from the fresh feed, and is thus a pretreatment process for the SWRO system. The MF system should process heavily contaminated feed without loss of filtration efficiency or physical (flow and differential pressure) performance.

Turbidity Removal

One of the primary performance indicators of MF as a pretreatment scheme for SWRO, is the turbidity reduction across the membrane surface. In the MF system at AECL turbidities of up to 3000 NTU (in the concentrated cross-flow stream) have been reduced to less than 0.5 NTU. The removal efficiency of the MF system for turbidity ranges between 98% and 99.9% depending upon the feed quality (2).

Pilot-Scale RO System Description

The pilot-scale RO system is typically operated in a batch volume reduction mode. Feed is pumped through a 5 µm cartridge pre-filter to the suction side of one of two positive displacement high pressure feed pumps. The pressurized feed flowing at about 55 L/min is directed to one of two membrane pressure vessels, each containing two 10 cm diameter by 100 cm long Filmtec SW30HR RO membrane elements. These are the same elements used in the WTC plant-scale system (Figure 1). The purified permeate stream exits the vessel, then passes through a flowmeter, and can be rejected from the system or recycled back to the feed tank.

The retentate stream containing 99% of the contaminants exits the vessel and splits into two streams. The majority of the retentate recirculates back to the suction side of the high pressure pump to maintain the high crossflow rate. The remainder of the retentate is directed back to the feed tank. The operating pressure of the system is manually controlled by adjusting the backpressure control valves on both retentate streams leaving the vessel. For the experiments reported here, the system was first operated in volume reduction mode to achieve the desired volumetric recovery. The permeate was rejected from the system while the retentate was returned to the feed tank. When the desired volumetric recovery was achieved the permeate stream was directed back to the feed tank with the retentate stream. The system was then operated in full recirculation for several hours. A system was provided to flush the vessel and piping with clean water after an experiment. A skid-mounted cleaning tank was also provided if chemical cleaning of the membranes was required.

Samples were taken of the feed and filtrate from the MF system and from the feed, retentate, and permeate streams from the pilot-scale RO system. The samples were analyzed for pH, conductivity, total solids, total $\beta\gamma$ activity, total α activity, various cations and anions, and gamma emitting isotopes by gamma spectroscopy. No pretreatment chemicals, such as antiscalants, were added to the MF filtrate and there were no pH adjustments made after MF filtration.

LIQUID WASTE PROCESSING OF FOUR WASTE STREAMS

Chemical Composition of Stream {1}

The Decontamination Centre (DC) stream has radioactivity concentrations generally below 40 kBq/L, but has substantial concentrations of suspended and soluble solids, typically in the range of 1 to 3 g/L. The Chemical Drains (CD) stream has a total solids loading of roughly 0.1 g/L, but has radioactive concentrations ranging from 400 to 100 000 kBq/L. Both solutions are acidic and are generated at approximately the same weekly rate of 30 000 L.

The average radioactivity concentration of the blended CD/DC waste stream {1} is about 15 000 kBq/L. Stream {1} is processed in batches first, with the microfiltration system, to remove the suspended solids and to provide a feed with a turbidity of less than 1 NTU and a silt density index of less than 3. The filtered feed is then batch processed through the spiral wound reverse osmosis system. After an operating period of about 200 hours, the average flux at a crossflow of 55 L/min (and at 85% recovery) was 1.85 L/min/element, compared to 0.55 L/min/element at 40 L/min crossflow and at 85% recovery in the plant-scale system.

A proportional fraction of the identical MF-filtered waste (about 1200 L per batch) that was processed by the plant-scale system, was simultaneously treated in parallel by the pilot-scale plant. Over 50 different batches of stream {1} waste were processed by the pilot-scale system. Each run took approximately 8 hours to complete. By employing the same membranes and vessel configuration, and processing the same feed stream at the same operating conditions, a direct comparison between the two system configurations could be made.

Osmotic Pressure and Conductivity for Streams {1} and {2}

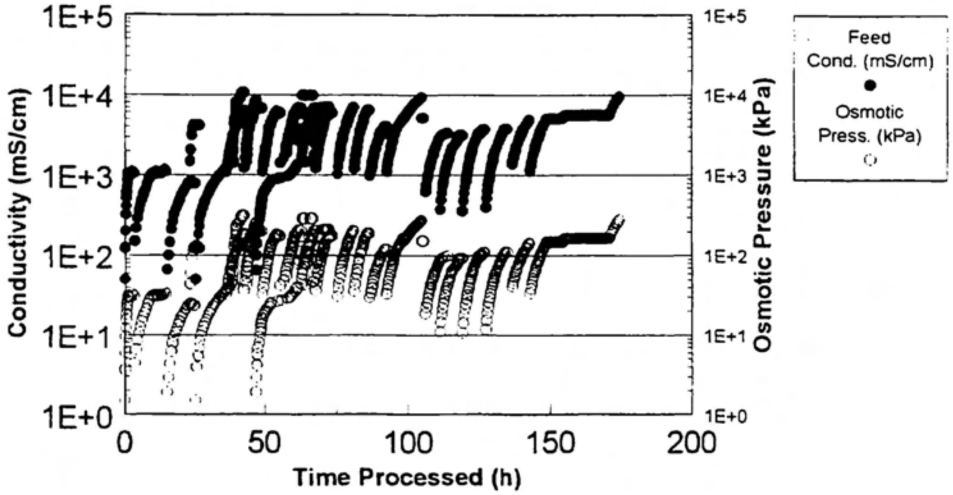
Two parameters were used to characterise the variability of the feed of {stream 1} for the pilot-scale system: osmotic pressure and conductivity, both of which are affected primarily by the non-radioactive chemical contaminants. The osmotic pressure of the feed stream was computed using equation 1 (4):

$$\pi(kPa) = 7.72\mu[T + 273] \qquad (1)$$

where : P = osmotic pressure (kPa),
T = temperature (°C), and
μ = total ionic strength (mol/L).

Figure 2 shows the increase of the osmotic pressure and conductivity for {stream 1} during the runs carried out over the first 200 hours of pilot-scale operation. The conductivity of the water increases during a run due to the rejection of conductive ions in the retentate. Typically, the conductivity of the feed solution increases by a factor of about 10 which agrees with the overall volumetric recovery of about 90% that is achieved. The osmotic pressure also increases by a factor of about 10.

Fig. 2
Conductivity and Osmotic Pressure
Processing of Streams {1} and {2} with Pilot System



Permeation Flux Performance for Streams {1} and {2}

The permeation flux from an RO system is a function of several variables including temperature, pressure, and pH. All permeation flux data obtained were normalised to an applied pressure of 2.76 MPa and a feed temperature of 25°C using a method given by Bukay (5). The observed permeation flux (OBF) was normalised for temperature using a temperature correction factor (TCF) obtained from the membrane manufacturer, an effective pressure, and for the total number of elements using equation (2).

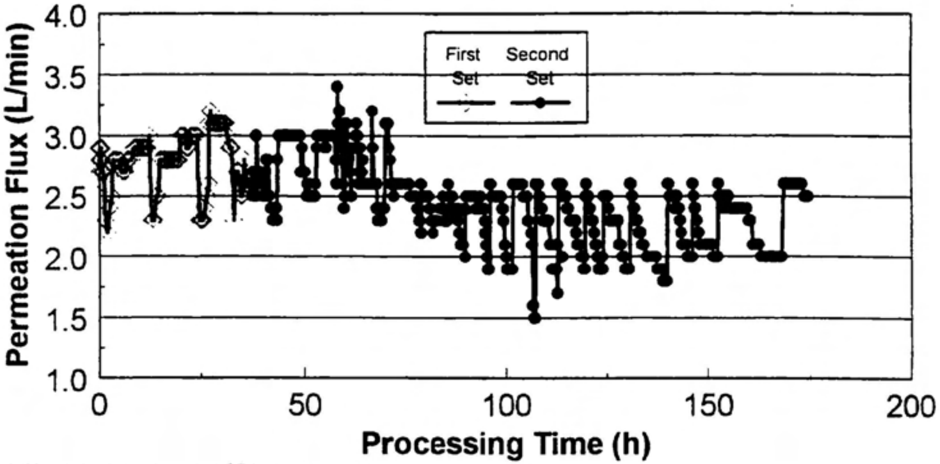
$$NPF = \frac{1}{TCF}\left[\frac{2.76(MPa)}{FEED\ PRESSURE\ -\ PERMEATE\ BACK\ -\ OSMOTIC}\right] * \frac{OBF}{No.\ Elements} \quad (2)$$

The instantaneous permeation flux per element for the pilot-scale system for {stream 1} is shown in Figure 3. During the initial 75 hours of processing there was no *net decline* of permeation flux per element. This is, in part, due to the fact that the majority of the waste stream processed during this time was "reactor drains waste", which is essentially clean water {stream 2} containing tritium from the operating reactors at AECL. Although there was also a flux decline during the processing of a given batch of CD/DC waste {stream 1} during the initial 75 hours of processing through the unit, this was recoverable to the original flux after a water flush at the end of the volume reduction step. On average, the permeation rate decreased from about 3 L/min to 2.2 L/min at the end of a run. At this point an overall volumetric recovery of 85 to 90% was achieved.

Fig. 3

Permeation Flux vs. Processing Time [1]

Processing of Streams {1} and {2} with Pilot System



1. Membranes replaced at 35 h due to rupture.

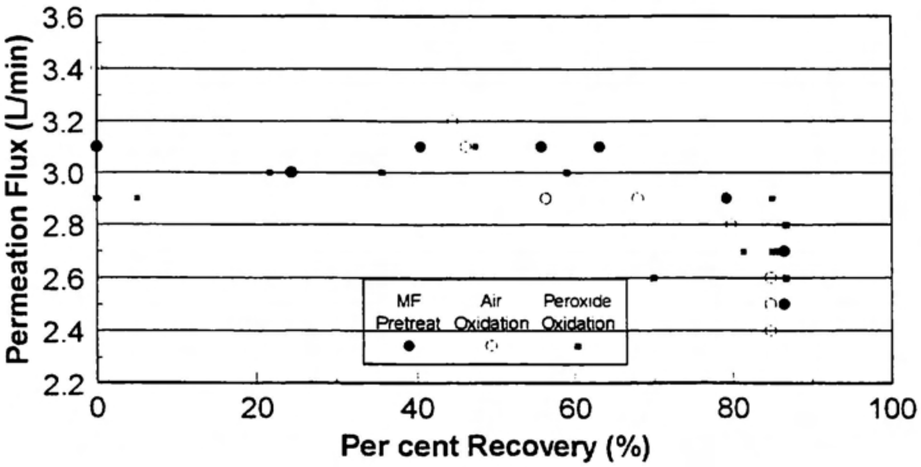
Following the initial 75 hours of operation, the fresh feed to the pilot plant was modified to a 50:50 blend of the CD/DC waste streams {stream 1}, and the performance was evaluated. The pilot-plant was run in parallel with the plant-scale system, and both of the systems were targeted to achieve 85% volumetric recoveries overall. In comparison with the plant-scale system, which was only able to maintain 0.5 L/min/element (at the start of a processing campaign), the pilot-scale system was able to sustain a permeation rate of 2.7 L/min. During the course of a run, the flux typically decreased from 2.7 L/min to about 1.7 L/min, depending upon the final volumetric recovery that was sought. However, the loss of flux during a run was always reversible (after flushing the system with permeate), indicating it was due to concentration polarization. If the system was placed in total recirculation at any time during the run the permeate flows would not decrease further. It seems evident that the permeate flux loss in the pilot-scale work is linked to concentration polarization at the membrane surface and not to the formation of a permanent fouling layer.

Effect of Oxidation as Pretreatment for Stream {1}

The flux loss that has been encountered on the plant-scale system since the initiation of full-scale operation in 1991 (2) could be attributed to several factors including: fouling due to coagulation of colloids, metals fouling, and organic fouling. One of the key milestones was to determine if oxidation of the MF filtrate with a UV/ozone system or with enhanced catalytic oxidation would decrease the flux loss that had been observed in the past (6). The objective was to determine if a significant fraction of the flux loss could be attributed to blockage of the membrane pores with organic contaminants.

Figure 4 shows the results of a series of experiments for which stream {1} was pretreated in three different ways. In the first, stream {1} was pretreated by microfiltration alone for the removal of suspended solids. The 0.2 µm backwashable filtration system was used upstream of the pilot-scale unit to lower the silt density index, and thereby prevent the deposition of particulate matter on the RO membranes.

Fig. 4
Effect of MF and Oxidation Pretreatment on Permeate Flux
Processing of Stream {1} with Pilot System



Following filtration with the MF system to remove suspended solids, the filtrate from processing of stream {1} was directed to the feed tanks of the pilot-scale RO system. Approximately 800 L of filtrate was collected for processing. For some experiments the filtrate was oxidized using a titanium dioxide catalytic oxidation rig with dissolved air and hydrogen peroxide (at a concentration of 500 mg/L). Results are shown in Figure 4, comparing the effects of MF pretreatment with and without catalytic-aided oxidation technologies. Using permeation flux as a performance indicator, there is no significant advantage in employing oxidation upstream of the pilot-scale unit for stream {1}. The permeation flux decreased from about 3.1 L/min initially, to approximately 2.2 L/min (at a volumetric recovery of 85%) for all three cases. The permeation flux initially started declining at a volumetric recovery of 65%. This suggests that saturation of salts in the boundary layer leading to concentration polarization becomes significant at a recovery in excess of 60%. Chemical analysis of the oxidized filtrate showed that there was no significant decrease of organic carbon content. Usually, the distribution of organic species changed after oxidation, but the overall concentration did not change.

Effect of Oxidation as Pretreatment for Stream {2}

Reactor Drains (RD) waste {stream 2} was also processed through the combined oxidation/RO system configuration. The RD waste is a dilute radioactive stream originating from the operating nuclear reactors at the AECL site. The stream contains virtually no suspended solids and very little chemical loading. The majority of the radioactivity of this stream is associated with tritium which is not removed by membrane technologies.

Figure 5 shows the effect of catalytic oxidation of the MF filtrate on the process ability of stream {2} by the pilot-scale unit. Catalytic oxidation was carried out in the presence of a titanium dioxide catalyst employing hydrogen peroxide as an oxidant, at a concentration of 50 mg/L (6). The permeation flux from the pilot-scale system was plotted versus the system volumetric recovery for three cases including: i) no pretreatment; ii) MF pretreatment; and iii) MF pretreatment with catalytic oxidation.

For a volumetric recovery of less than 90% there was no impact of any of the pretreatment options on the permeation flux curve for stream {2}. For recoveries exceeding 90% the permeation flux was sensitive to slight perturbations of recovery, and it rapidly decreased from approximately 2.8 L/min to 2.2 L/min. There is no evidence to indicate that the use of MF and/or preoxidation decreases the decline of permeation flux in the pilot-scale system. The results corroborate earlier speculation that the primary cause of premature membrane fouling is related to insufficient turbulence at the membrane boundary layer leading to a thicker hydrodynamic boundary layer. The thicker boundary layer leads to precipitation and growth of a fouling layer on the membrane surface. Chemical analyses of the oxidized filtrate confirmed that the distribution of the organics species had changed, but the overall organics concentration had not.

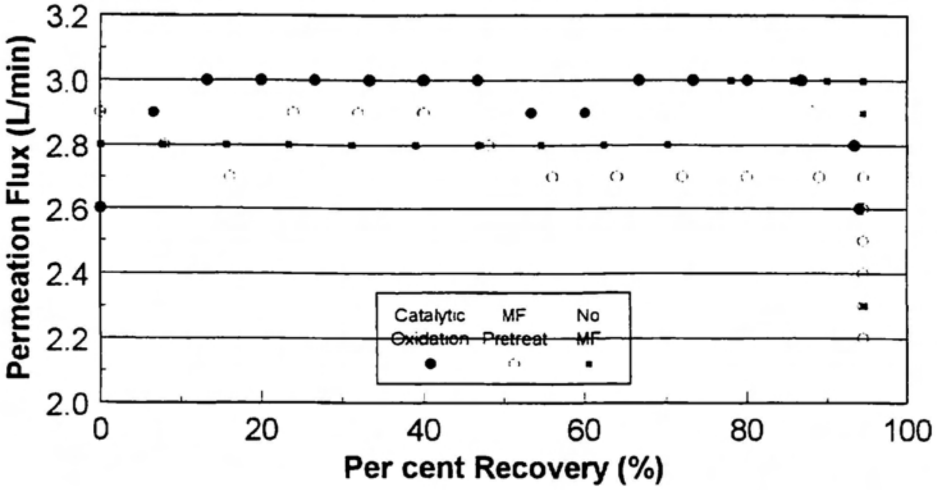
Processing of Blended IX Regenerant Waste {stream 3}

Origin of IX Regenerant Waste. Water from the reactor rod bays at Chalk River Labs (where spent fuel rods are stored in large pools) are continuously recirculated through independent pre-coat filter systems for particulate removal. A portion of the filtered water from each rod bay is directed to separate level-controlled concrete surge tanks. It is then processed through a mixed-bed ion exchange column to remove ionic contaminants in a 1m diameter vessel, containing about 0.3 m^3 of cation resin and 0.6 m^3 of anion resin. The effluent from the column is directed to deionized water reservoirs that are continuously circulated.

Fig. 5

Effect of MF and Oxidation Pretreatment on Permeate Flux

Processing of Reactor Waste {Stream 2} with Pilot System



Typically the mixed-bed column requires chemical regeneration four times a year. Regeneration is accomplished by first separating the cation and anion resins in the column by backwashing with deionized water, then simultaneously pumping dilute sodium hydroxide and nitric acid solutions through the anion and cation resins respectively. Approximately 6 m^3 of intermediate level radioactive liquid is created with each regeneration which is known as the IX waste.

Following the regeneration sequence, the IX waste is processed through a small falling film evaporator. The distillate from the evaporator is pumped to the chemical drain system and becomes a portion of the stream {1} that is currently treated at AECL with membrane technology. The concentrate is currently being stored in a large tank awaiting processing. The composition of the CD/DC waste stream {1} is compared with the blended CD/DC/IX stream {3} in Table 1.

Table 1
Comparison of the Existing WTC MF Feed for Stream {1} and Stream {3}

Parameter	MF Feed		Increase (%)(*)
	DC/CD {1}	DC/CD/IX {3}	
Volume (m^3/yr)	3 300	3 324	0.7
Conductivity (mS/cm)	950	1 200	26
Total Solids (mg/L)	1 400	1 700	21
Cl (mg/L)	120	119	0
PO_4 (mg/L)	240	240	0
SO_4 (mg/L)	120	120	0
NO_3 (mg/L)	295	440	49
Na (mg/L)	265	335	26
Gross α (Bq/ml)	3.9	4.6	18
Gross $\beta\gamma$ (Bq/ml)	2 700	3 400	26
Tritium (Bq/ml)	1 200	1 300	8
Co-60 (Bq/ml)	127	170	34
Cs-134 (Bq/ml)	81	103	27
Cs-137 (Bq/ml)	580	1 077	86

* Increase of parameter for stream {3} in comparison with stream {1}.

The goal of the pilot-scale treatability test performed was to investigate the impact of treating IX waste, blended with CD/DC waste {stream 3}, on the permeation flux of the Filmtec RO membranes. The test results could be compared to results obtained from similar tests for stream {1}.

Experimental Procedures for Processing Stream {3}. The distribution of radionuclides present in the IX regenerant waste is dominated by highly soluble Cs-137 and Cs-134. These species are not significantly rejected by the MF system and, therefore, do not concentrate in the MF backwash liquid. The IX waste contains primarily soluble species, since the feed stream, which is reactor bay water, is filtered by a Bowser pre-coat filter system for removal of particulate matter prior to the deionization step.

Five separate one litre samples of IX waste were collected over the one hour chemical addition step of the regeneration procedure. All the samples contained a significant mass of a white gelatinous precipitate, thought to be aluminum nitrate, and consisting of up to 25% of the sample volume. The radiation fields on contact with the bottles were usually between 200 to 400 mR/hr. The samples were returned to the analytical lab where they were agitated, and a 125 ml composite sample containing the precipitate was prepared for analysis. The remainder of the five litre sample (about 4.8 L) was mixed with 850 L of stream {1} to simulate the blend which would occur if the ongoing IX regenerant waste were to be treated at the WTC as {stream 3}.

Stream {3} waste was passed through the MF system for removal of suspended solids. The suspended solids free stream, or filtrate, from the MF was directed to the feed tanks of the pilot-scale reverse osmosis system. Approximately 800 L of filtrate was collected for each experiment.

Treatment of IX Regenerant Waste Stream {3}: The pilot-scale test parameters and performance summary for stream {3} and the previous stream {1} experiments are shown in Table 2. A 95% confidence band (representing two standard deviations around the mean value) is given. The test parameters for the two data sets are about the same, although the final volumetric recovery was slightly higher for the stream {3} batch (88% versus a mean value of 84%).

Table 2
Pilot-Scale System Performance for Waste Streams {1} and {3}

Parameter	DC/CD {1}	DC/CD/IX {3}
Final Volumetric Recovery (%)**	84.1 ± 1.4	88
Feed Pressure (kPa)	2 440 ± 40	2400
Feed Temperature (°C)	20.9 ± 1.7	20
Feed pH	6.4 ± 1.7	7.1
Initial Normalized Permeate Flow at Zero Volumetric Recovery (L/min/element)	2.8 ± 0.4	2.8
Normalized Permeate Flow Loss (%)	14.3 ± 15	18
Crossflow Per Vessel (L/min)	53.1 ± 0.9	55
Crossflow Pressure Drop (kPa)	155 ± 29	138
Crossflow Pressure Drop Increase (%)	7.7 ± 18	5

** Volumetric recovery (ratio of permeate to feed volume) achieved by batch volume reduction with complete rejection of permeate and recirculation of retentate.

The most critical parameter recorded was the permeate flow decline associated with volumetric reduction of the feed stock. The initial permeate flowrate of 2.8 L/min/element obtained from the stream {3} experiment was equivalent to that observed for the processing of stream {1}. The normalized permeate flow loss was 18% for stream {3} at 88% volumetric recovery. This compares well with the mean value of 14% (at 84% volumetric recovery) obtained for the processing of stream {1}. The permeate flow loss during the run was reversible with a simple permeate flush after the completion of the batch of waste. It was strictly a function of concentration polarization for stream {3}, and not the result of the formation of a permanent fouling layer on the membrane surface.

In addition to permeate flow loss, fouling of RO membrane elements can result in increased pressure drop due to coagulated colloidal particles or precipitates depositing in the element feed channels. At a crossflow of about 55 L/min the initial pressure drop across the membrane vessel was 138 kPa for the stream {3} test, and it increased by only 5% during the test. These values are consistent with those obtained from the stream {1} experiments, indicating that there was no increased tendency for fouling or flux loss due to any other source.

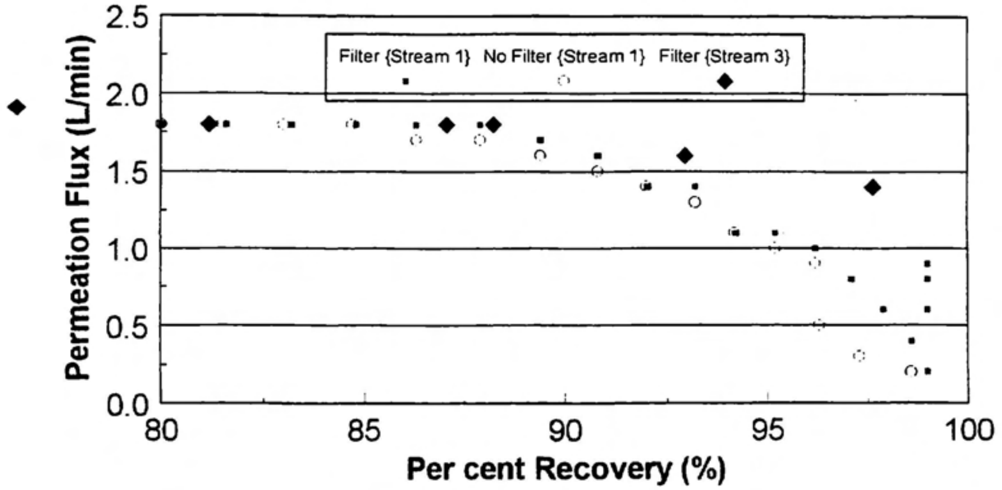
Table 3 shows the contaminant rejection efficiencies determined for both the MF and pilot-scale RO systems. The rejection efficiencies for the monitored contaminants are consistent with those observed for stream {1}.

Table 3
MF and Pilot-Scale RO Contaminant Rejections for Stream {3}

Parameter	MF Contaminant Rejection (%)	Pilot-Scale RO Contaminant Rejection (%)
Conductivity	Nil	99.93
Total Solids	47.5	99.00
Na	19.6	99.61
NO_3	12.6	99.30
Cl	Nil	99.86
SO_4	13.8	99.87
PO_4	21.8	99.97
Gross α	90.1	100
Gross $\beta\gamma$	19.9	99.46
Co-60	17.2	99.77
Cs-134	16.7	99.52
Cs-137	12.8	99.5
Total γ	17.8	99.65

Effect of Volumetric Recovery on Permeation Rate for Stream {3}. An experiment was carried out to determine if the presence of IX regenerant waste (in waste stream {1}) would significantly affect the decline of permeation flux observed at volumetric recoveries above 80%. Figure 6 shows an experiment for which a batch of CD/DC waste {stream 1} was passed through the pilot-scale Filmtec membranes for three cases including: i) with 5 µm filtration and CD/DC waste; ii) without 5 µm filtration and CD/DC waste; and iii) with filtration for CD/DC/IX waste.

Fig. 6
Effect of Filtration on Permeation
Processing of IX Regenerant Waste {Stream 3} with Pilot System



The permeation flux for the combined CD/DC/IX {stream 3} was initially at about 1.8 L/min/element at a recovery of 80%. For volumetric recoveries exceeding 85%, the decrease of permeation flux for CD/DC waste {stream 1} was greater than that for stream {3}. At a volumetric recovery of 97%, the permeation flux for stream {3} was 1.4 L/min, compared to about 0.7 L/min for stream {1}. It is concluded that the presence of the mixed IX waste should have no adverse impact on the routine processing of AECL wastes {stream 1} by reverse osmosis technology.

Treatment of Boron-Containing Waste {Stream 4} with Reverse Osmosis

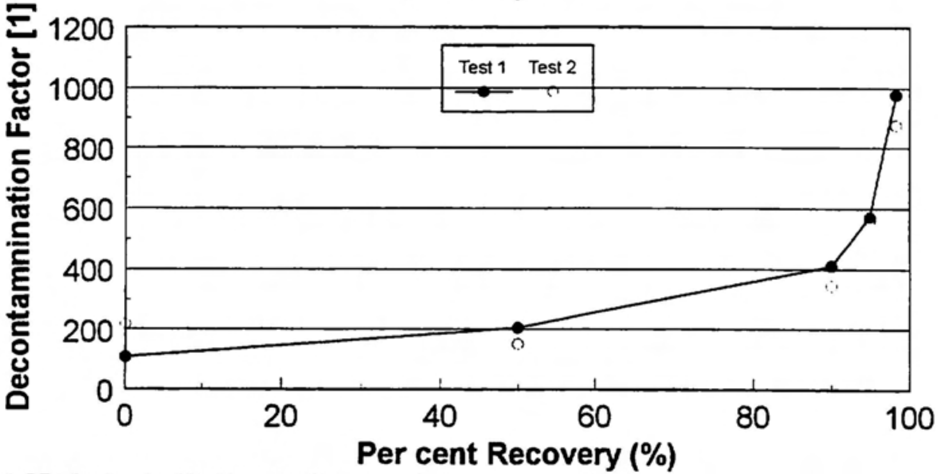
Origin of Boric Acid Waste. Boric acid is used in pressurized water reactors (PWRs) as a soluble neutron poison to control reactivity, and also to assure a safety margin in the spent fuel pool during refuelling operations. A typical US PWR starts its fuel cycle with about 5000 kg of boric acid dissolved in the coolant. As the fuel is exhausted the boric acid is removed from the reactor coolant, until substantially none remains prior to the refuelling outage. Most of this boric acid is removed by bleed and feed dilution of the reactor coolant system by the chemical and volume control system, and is collected for concentration. This stream may be called reactor letdown. In some cases, when the boric acid concentration is low, smaller quantities of boric acid are be removed by ion exchange.

Most Western European and US PWR stations have waste streams with boric acid concentrations of up to about 0.8% (7). These streams usually have to be processed before they can be released to the environment. The liquid radwaste containing boric acid generated from a PWR is generally evaporated and the concentrate then solidified. The liquid radwastes are concentrated as far as possible to yield the highest volume reduction for the selected immobilization scheme, since the volume of consolidated waste is critical to the determination of the ultimate disposal cost. Another more economical method for volume reducing the boric acid radwaste may be to employ reverse osmosis technology at an elevated pH. The operating pH of the equipment is critical, and it should be in the alkaline region, since boric acid permeates the membrane in the acidic regime.

Recovery of Boric Acid by RO. Figure 7 shows a plot of DF versus volumetric recovery of the RO system for a 0.6% boric acid synthetic (fresh feed) solution, at an operating pH of 9.

Fig. 7

Removal of Boron {Stream 4} with RO
Pilot-Scale System



1. DF refers to ratio of feed to permeate concentrations.

The *decontamination factor* (DF), was used rather than a rejection efficiency for clarity on the figure. The decontamination factor (DF) is defined by equation (3).

$$DF = \frac{Feed\ Concentration}{Permeate\ Concentration} \equiv \frac{1}{1 - Fraction\ Rejected} \qquad (3)$$

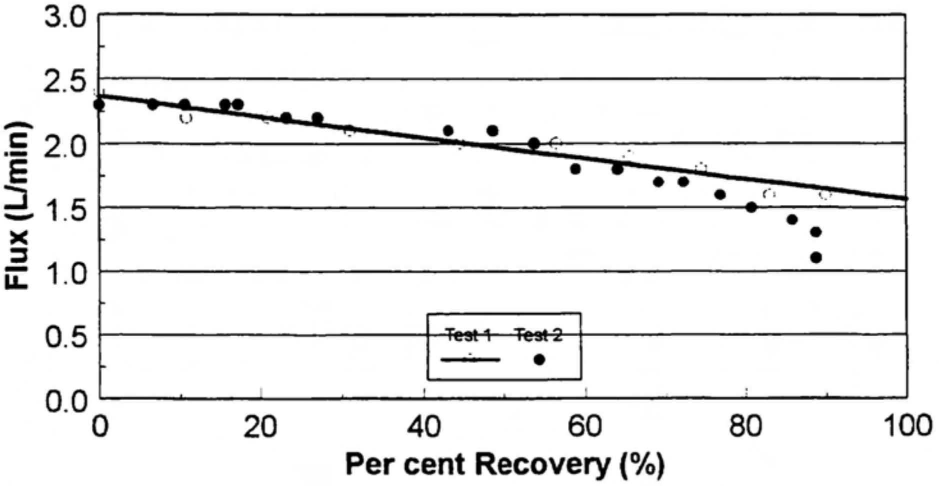
Regardless of the incoming feed concentration of boric acid, the effluent concentration in the permeate decreases to a constant low value. This is contrary to results obtained for other chemical contaminants (1), where it has been shown that the rejection efficiency for a given membrane was fixed. The DF for boric acid (0.6% starting solution concentration) increases with the volumetric recovery of the pilot-scale system. Further tests will determine if there is an upper limit above which the DF levels off with volumetric recovery.

Figure 8 shows that there is a slight decrease in permeation flux for the processing of a 0.6% boric acid solution to 90% recovery. The flux decreases gradually from about 2.4 L/min to approximately 1.5 L/min at a volumetric recovery of about 90%. Considering that the total solids concentration at a volumetric recovery of 90% is about 6% (in boric acid alone), the decline of flux is not substantial. The slight decrease of flux with recovery may be attributed to the supersaturation of boric acid, which has a solubility of about 5.5% in the absence of other salts. Since the bulk concentration of the boric acid was about 6%, the concentration of boric acid in the boundary layer next to the membrane surface would be several times the supersaturation level. In spite of this, there is only a modest reduction of permeation flux.

Fig. 8

Boron Recovery from Stream {4} with RO

Concentration of 0.6% Boric Acid in Pilot System



CONCLUSIONS

Spiral wound reverse osmosis technology has been demonstrated as an effective treatment for a variety of different aqueous radioactive waste streams. Although there is usually a reduction of permeation flux during the volumetric reduction of waste due to concentration polarization effects, there is no permanent loss of flux if the crossflow velocity remains at 55 L/min/vessel.

If the feed is filtered, there does not appear to be any significant advantage of additional pretreatment technologies (such as oxidizing the feed stream with enhanced oxidation for organic destruction) for the four waste streams investigated.

Contaminated reactor drains water at AECL was treated with the pilot-scale reverse osmosis system operating at a volumetric recovery of 98 to 99%. No flux decline occurred for this waste stream prior to a 95% volumetric recovery. Minimal pretreatment was required to achieve this volumetric recovery, and chemical scaling concerns were not significant. Pretreatment of the reactor waste stream with enhanced oxidation technologies including UV-ozone and catalytic oxidation did not improve the permeation flux performance.

The mixed radioactive waste that results from the regeneration of ion exchange columns (when blended with other waste streams) was effectively treated with reverse osmosis. The rejection efficiencies of all contaminants investigated exceeded 99.3%, after partial removal of contaminants upstream by microfiltration. Gross $\beta\gamma$ removal was 99.5% and gross α removal was virtually 100%. The performance of the RO membranes- either for permeation flux or rejection efficiency- was not affected by the higher activity IX regenerant waste stream. The permeation flux in the system decreased by 18% for the IX regenerant blended waste, compared with a reduction of 14% for the unblended waste. The pressure drop increased by between 5-10% after processing of either waste stream.

Boric acid, used as a moderator in pressurized water reactors (PWRs), can be concentrated from 0.6% to 6% by reverse osmosis. The decontamination factor for the boric acid increased from 200 to 1000, as the volumetric recovery increased from 50% to 99.5%. The final concentration of boric acid in the permeate was independent of the incoming feed concentration of boric acid. The variation of rejection efficiency with initial feed concentration was opposite to that previously observed for decontamination of other chemical contaminants. The results would suggest that the Filmtec membrane is slightly permeable to boric acid to give some nominal value in the permeate, regardless of the concentration in the fresh feed.

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