#### **OPERATION OF A MEMBRANE PILOT PLANT FOR ANSTO EFFLUENT**

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#### ABSTRACT

The Australian Nuclear Science and Technology Organisation (ANSTO) operates Australia's only nuclear reactor. The existing HIFAR reactor, built in the 1950's, is a 10MW research reactor and is due to be shutdown in 2006 with the commissioning of a replacement 20MW pool type research reactor. In conjunction with this, ANSTO plans to upgrade its low level liquid waste (LLLW) treatment plant to further reduce radioactivity discharged to the sewer.

The feasibility of using membrane technology for further removal of radioactivity from ANSTO's industrial effluent was investigated in a membrane pilot plant. The possible reuse of the recovered water for make-up in the reactor secondary cooling circuit was also assessed. This would reduce total effluent discharge by half and reduce fresh water demand for the reactor secondary cooling circuit.

A tubular ultrafiltration (TUF) unit followed by a spiral-wound reverse osmosis (RO) membrane pilot plant was procured for pilot trials. The size of the pilot plant was based on 10% of the estimated full-scale flow of a future plant. The reverse osmosis section consisted of two passes for permeate and two stages for concentrate. The pilot plant also included a cationic and anionic ion-exchange system, for possible use as a polishing step should radioactivity be detected in the Pass 2 RO permeate. The pilot trial was carried out to assess the performance of the system in terms of radioactivity removal, volume recovery, operational stability, and fouling issues for ANSTO's LLLW industrial effluent. Operational and analytical data was also collected for scale-up purposes.

The pilot plant trials demonstrated that membrane technology rejected all radioactivity from ANSTO LLLW and produced very high quality product water for recycle. Sufficient data was obtained for scale up to a full size plant. Control or elimination of surfactant fouling from one of the LLLW streams needs to be resolved before a full-scale membrane plant is built.

#### INTRODUCTION

ANSTO discharges approximately  $100,000 \text{ m}^3$  of liquid effluent annually. Half of this is domestic sewage and the remainder is industrial wastewater. The industrial wastewater includes low-level radioactive wastewater generated from the production of radiopharmaceuticals.

The feasibility of using membrane technology to further increase the removal of radioactivity from ANSTO's industrial effluent was investigated. A membrane pilot plant was procured to assess the performance of a membrane system in terms of radioactivity removal, volume recovery, operational stability and fouling issues. Operational and analytical data was also collected for scale-up purposes. The tests were carried out from August 2001 to September 2003. This paper provides some results from this investigation.

## **Existing Effluent Treatment Plant**

ANSTO has two effluent networks (excluding sewage) which are denoted the 'B' and 'C' lines. Figure 1 depicts the current collection and treatment system.



Fig. 1 ANSTO Wastewater Collection and Treatment System

All treated effluent is currently transferred to the sewer through a 3 km pipeline to a connection point in a southern suburb of Sydney. The 'B' line collects low-level effluent from 'active' areas such as laboratories and radioisotope production facilities. The 'C' line collects effluent from non-active areas such as workshops and cooling towers. Total flow to the sewer is 80-100 ML per year, of which 50% is sewage. Approximately 120 - 140 m<sup>3</sup> of B and C line effluent is collected in total each day. The major component of the 'C' line water is non-radioactive cooling tower blowdown from HIFAR. This is currently 80-90 m<sup>3</sup> per day but may increase with the commissioning of the replacement reactor.

The 'B' line effluent flow is approximately 15 m<sup>3</sup> per day and is collected in a batch tank and treated once per week in batches of 100 m<sup>3</sup>. The existing treatment of 'B' line effluent is chemical dosing with alum (aluminium sulfate), settling of the flocculated sludge, decantation of the supernatant and centrifugation of the sludge. This treatment removes some of the radioactivity, but the removal (30-80%) varies according to the particular species of radionuclide. The treated 'B' liquor is mixed with 'C' line liquor and sewage for discharge to the sewer. The alum sludge is dried in solar evaporation ponds and manually drummed as low level solid waste.

The current treatment system meets ANSTO's Trade Waste Agreement with the local authority, Sydney Water. Table I provides yearly discharge data for 1996-2001. The increase in tritium discharge for the year 2000 was due to the HIFAR major shutdown of February – April 2000.

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Voor	Volume	Total Alpha	Total Beta	Total Tritium	% of
real	$(m^3)$	(MBq)	(MBq)	(GBq)	Authorised
1996	84,112	51.4	1,496	542	22
1997	83,179	135.9	2,422	353	38
1998	96,460	<183	2,224	596	<37
1999	95,364	<63	972	348	<16
2000	80,796	<73	1091	1761	<29
2001	90,470	<88	565	795	<18

Table I Annual Liquid Effluent Discharged for 1996-2001

#### **Membrane Pilot Plant**

The aims of the pilot plant tests were to assess radioactivity removal, obtain design data for a full-scale plant, and maximize recovery of ultra pure water for recycle and minimise the concentrate volume for evaporation/drying. During pilot testing the following parameters were examined: the number of membrane stages required, the need for ion exchange as a final step for permeate quality, potential fouling mechanisms, cleaning cycles, and other operational issues. The configuration of the pilot plant treatment stages for the LLLW is shown in Figure 2. The pilot plant design flow rate was 20 L min<sup>-1</sup> (~29 m<sup>3</sup> per day). This is approximately 10% of the estimated flow required for a full-scale plant.



Fig. 2 Treatment Stages of Membrane Pilot Plant

The pilot plant consists of a tubular ultrafiltration (TUF) unit followed by spiral-wound reverse osmosis (RO) membrane modules. The TUF unit filters the feed to sub micron level, removing large molecular weight materials and colloidal matter. The unit was manufactured by PCI Membrane Systems, and consists of eight 3.6 m long stainless steel housings interconnected with U-bends. Inside each housing is a A19 membrane module, consisting of a bundle of 19 tubes fitted into epoxy end pieces. Wastewater is

processed through the inside of the 12.5 mm tubes and permeate is collected in the shroud (shell).

The reverse osmosis section consists of two passes for the permeate and two stages for the concentrate. The RO units process wastewater that has been pretreated by the TUF. Pass 1 RO (RO1) reduces the dissolved contaminant level of the wastewater. Pass 2 RO (RO2) further reduces the contaminant level of RO1 product water. Stage 2 RO (S2RO) concentrates the reject from RO1 to maximise water recovery and also to minimise the final waste disposal volume. Spiral wound thin-film composite membranes were used in all the reverse osmosis units. The membrane consists of an ultra-thin polyamide barrier layer, a microporous, polysulphone layer and a high strength polyester support web. The membranes for Pass 1(RO1) and Pass 2 (RO2) are FILMTEC extra low energy (XLE) elements while membranes for Stage 2 (S2RO) are FILMTEC fiberglassed brackish water elements. The pilot plant also includes a cationic and anionic ion-exchange (IX) system, for possible use as a polishing step if significant radioactivity is detected in the Pass 2 RO permeate.

Table II provides details of the membranes used in both the TUF and RO units. Table III provides details of the dosing chemicals used for the operation, cleaning and preservation of the RO units.

	TUF	RO1	RO2	S2RO			
Membrane element	PCI FPA10	4" FILMTEC	4" FILMTEC	2.5" FILMTEC			
	100,000 MWCO	XLE-4040	XLE-4040	BW30-2540			
No. of elements	8	6	3	3			
Membrane material	PVDF	Aromatic	Aromatic	Aromatic			
		polyamide thin-	polyamide thin-	polyamide thin-			
		film composite	film composite	film composite			
Total membrane	$20 \text{ m}^2$	$45.6 \text{ m}^2$	$22.8 \text{ m}^2$	$7.8 \text{ m}^2$			

Table III	Dosing	chemical	ls
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CHEMICALS	CONTAINS	USED FOR
Bioclean	Proprietary blend containing inorganic and organic	All RO units
	salts, surfactants and sequestering agents	
Anti-scalant (AS)	AS-1000 = Phosphinocarboxylic acid.	RO1 and S2RO
	AS-1300 = Polycarboxylic acid	only
Biocide	BIO-2000 = 2,2-dibromo-3-nitrilo propionamide	RO1 only
	DIO-2100 – Diena of isounazonni compounds	
Sodium metabisulphite	Na <sub>2</sub> S <sub>2</sub> O <sub>5</sub>	RO1 only
Hydrochloric Acid	HCl (10-36%)	S2RO only

The pilot plant is skid mounted and includes automatic control using a programmable logic controller (PLC). It is sufficiently automated to run 24 hours per day. The PLC is mounted in an electrical control panel with easy operator access via a touch screen display, and records data from the pilot plant every 30 minutes. For maximum flexibility of operation, each section of the pilot plant operates independently of the others. Apart from certain alarm conditions, continued operation of a particular section is only dependent on an adequate level in the respective feed tank to that unit. Initially the pilot plant was tested on 'C' line wastewater (90% of non-sewage effluent flow). Combined 'B' and 'C' wastewater was then tested and a single test was carried out on 'B' line wastewater. 'C' line wastewater spiked with radioactivity was also tested several times. The final tests on wastewater were carried out in September 2003. Figure 3 shows the membrane pilot plant in place on site.



Fig. 3 Membrane Pilot Plant

# **RESULTS AND DISCUSSION**

The pilot plant was run for a total of 40 weeks. Initially the plant was run on a week by week basis with shutdown over weekend periods. This was found to be detrimental as it encouraged the growth of biological activity during shutdown periods and generated more secondary waste in the form of waste cleaning and sterilisation solutions. Biological fouling of the RO modules was persistent until continuous operation was adopted after Week 19.

The pilot plant was commissioned on town water (tapwater) and after 3 weeks C-line wastewater was introduced as feed. C-line was the dominant feed over the duration of the test work period apart from periods where tapwater was re-introduced to keep the plant running while fouling investigations were carried out.

The membrane pilot plant processed radioactive feedwater for a total of 36 days. B-line, mixed B and C line, and C-line spiked with radionuclides (TICAN and AMP waste) were used to investigate the rejection of radioactivity by the membrane plant (TICAN and AMP are radioactive liquid wastes generated from radiopharmaceutical production). The schedule of tests using low level radioactive feedwater is shown in Table IV.

The radionuclides present in the different feed types are given in Table V.

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Week	Feed Type	Days of Operation	Units				
27	B/C (30/70)	7	TUF and RO				
28	B/C (30/70)	4	TUF and RO				
32	В	5	TUF only				
36	C+TICAN	4	TUF and RO				
37	C+TICAN	3	TUF and RO				
38	C+TICAN	4	TUF and RO				
39	C+TICAN	6	TUF and RO				
40	C+AMP	3	TUF and RO (except				

Table IV	Active	Feedwater	Tests

Table V Radionuclides in Feed Type

Feed Type	Radionuclides
B and B/C	<sup>141</sup> Ce, <sup>144</sup> Ce, <sup>60</sup> Co, <sup>51</sup> Cr, <sup>137</sup> Cs, <sup>131</sup> I, <sup>95</sup> Nb, <sup>95</sup> Zr, <sup>103</sup> Ru
C + TICAN	<sup>60</sup> Co, <sup>137</sup> Cs, <sup>99</sup> Mo, <sup>95</sup> Nb, <sup>103</sup> Ru
C + AMP	<sup>60</sup> Co, <sup>134</sup> Cs, <sup>137</sup> Cs, <sup>131</sup> I, <sup>95</sup> Nb, <sup>95</sup> Zr, <sup>103</sup> Ru, <sup>106</sup> Ru

#### Ultrafiltration

The TUF system flux deteriorated with time during the operation of the pilot plant. This was due to slow colloidal fouling of the membrane pores, which could not be cleared by normal cleaning procedures using caustic (NaOH) and hypochlorite (hypo). Figure 4 illustrates the flux decline with time for the first 30 weeks of the pilot plant operation. The most catastrophic flux decline was recorded during Week 32 when B-line only was fed to the plant after new TUF membranes were installed. Decant of the alum dosed B-line (B-alum) was fed to the plant in Week 32 for five days. A starting flux of 66 Lh<sup>-1</sup>m<sup>-2</sup> was recorded. However the flux dropped rapidly, reducing to 25 Lh<sup>-1</sup>m<sup>-2</sup>, about a third of the starting flux. The rapid drop in flux continued on Day 3, where the flux more than halved to 12 Lh<sup>-1</sup>m<sup>-2</sup>. By Day 5, the flux of the new membranes was only 8.2 Lh<sup>-1</sup>m<sup>-2</sup>. Over the five days, the average daily flux decline for B-alum feed was 25%. This rapid flux decline is shown in Table VI.



Fig. 4 TUF Flux vs Time

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	Temp °C	Normalised Flux L h <sup>1</sup> m <sup>-2</sup>							
Day 0	26.4	65.9							
Day 2	37.1	25.3							
Day 3	34.7	11.7							
Day 4	35.0	9.0							
Day 5	35.3	8.2							

Table VI Week 32 B-line Flux

The B-line feed to the TUF was terminated after Day 5. A high temperature Clean-In-Place (CIP) with NaOH/hypo was then carried out. With tapwater as feed, the flux returned to 20  $Lh^{-1}m^{-2}$ . A staged CIP was then carried out, 1.5 hours of nitric acid clean followed by an hour of NaOH/H<sub>2</sub>O<sub>2</sub> (35w/w%) clean. The flux was restored to 40  $Lh^{-1}m^{-2}$ , slightly more than half the stabilised flux of the new membranes.

Investigation into the rapid decline of the TUF flux for this test revealed a high concentration of surfactants in the B-line. These surfactants originate from radiopharmaceutical production facilities. Surfactants are known to cause rapid fouling of polymeric membranes [1]. Laboratory tests were then carried out and these indicated that the surfactants in the 'B' line were destroyed by chemical oxidation with or without UV light. Destruction of surfactants using oxidation technology is not uncommon and is expected to be not technically difficult or costly [2,3].

### **TUF Rejection**

Most of the radionuclides in the wastewater are beta-emitting radionuclides. Alpha radioactivity was only detected in the TUF feed tank in Week 27 and Week 40 when the suspended solids in the TUF feed tank (TK-01) had accumulated significantly during the week.

Gross alpha and beta activity was measured using liquid scintillation. Gamma spectrometry was used to identify and quantify the trace radionuclides present in the samples. Due to the long lead-time in analysis, only selected samples in each test were analysed for radioactivity.

Table VII provides the rejection of gross alpha, beta and various radionuclides by the TUF and RO units. Rejection is calculated from the following equation:

$$Rejection = 1 - \frac{activity of permeate}{activity of feed}$$
(Eq. 1)

When the activity value is less than the detection limit (LDL), the value of the detection limit for that analysis is used in the calculation, resulting in very conservative rejection figures.

The TUF rejects up to 97% of alpha and 83% of beta activity in the feed. As shown in Table VII, <sup>95</sup>Nb, <sup>95</sup>Zr, <sup>106</sup>Ru, <sup>141</sup>Ce, <sup>144</sup>Ce and <sup>60</sup>Co were usually always completely rejected by the TUF. These radionuclides are usually present in the feedwater in colloidal form or they attach easily to particulate material in the feed, and thus can be removed by ultrafiltration.

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Rejection for	alpha	beta	<sup>141</sup> Ce	<sup>144</sup> Ce	<sup>60</sup> Co	<sup>51</sup> Cr	<sup>134</sup> Cs	<sup>137</sup> Cs	<sup>131</sup> I	<sup>99</sup> Mo	<sup>95</sup> Nb	<sup>95</sup> Zr	<sup>103</sup> Ru	<sup>106</sup> Ru
TUF	65.9%	76.9%	70.0%	47.1%	LDL	51.1%		LDL	LDL	LDL	87.5%	76.2%	78.6%	LDL
	96.5%	82.7%	90.0%	70.6%	LDL	48.9%		LDL	LDL	LDL	92.5%	85.7%	96.4%	LDL
	89.5%	46.0%					34.0%	8.2%	6.3%		98.7%	99.1%	80.2%	83.9%
	50.0%	64.3%					47.8%	11.7%			99.8%	99.7%	89.1%	87.4%
		82.4%						8.4%	41.3%		99.5%	99.1%	94.8%	96.6%
Average	75.5%	70.5%	80.0%	58.8%	LDL	50.0%	40.9%	9.4%	23.8%		95.6%	92.0%	87.8%	89.3%
RO1	96.0%	99.8%					76.2%	98.4%	99.6%		80.8%		99.8%	96.8%
	82.5%	99.4%					84.3%	98.5%	99.8%		88.5%	LDL	99.6%	97.9%
		99.2%					82.3%	99.1%	99.6%		90.0%		99.8%	99.0%
Average	89.3%	99.5%					80.9%	98.7%	99.7%		86.4%	LDL	99.7%	97.9%
RO2		66.7%						97.8%	95.2%				85.3%	
		87.9%						97.0%						
		77.8%						97.3%	97.5%					
Average		77.5%						97.4%	96.4%				85.3%	

Table VII Rejection by TUF and RO

The radionuclides  $^{137}$ Cs,  $^{134}$ Cs, and  $^{131}$ I were poorly rejected by the TUF as they usually exist in the feed in ionic form and do not attach easily to suspended solids. On average only 9% of  $^{137}$ Cs, 24% of  $^{131}$ I, and 41% of  $^{134}$ Cs was rejected by the TUF.

The amount of alpha and beta activity associated with suspended solids varied from week to week. No correlation between feed suspended solids and gross beta level of the TUF permeate was found. Table VIII shows the variation in beta activity in the TUF permeate compared to the feed suspended solids for three different operating weeks. There is however a trend of decreasing gross beta level in the TUF permeate with increasing suspended solids concentration in TUF feed tank TK-01 during each week's operation.

	SS in TK-01	Gross beta of TUF permeate
	mg $L^{-1}$	$\operatorname{Bq} L^{-1}$
Week 37	360	154
	545	114
Week 38	18	387
	59	359
	105	315
Week 40	53	810
	108	759
	221	522

Table VIII Feed Suspended Solids and TUF permeate

SS = suspended solids

Conversely for the same batch of feedwater, there was a trend of increasing gross beta with increasing suspended solids in TK-01 as TUF reject was being recycled (Table IX).

	SS in TK-01	Gross beta in TK-01
	mg L <sup>-1</sup>	Bq L <sup>-1</sup>
Week 38	18	1391
	59	2261
	105	2943
Week 39	153	3587
	238	4830
	245	5396
Week 37	360	735
	545	971

Table IX. Feed Suspended Solids and Beta Activity

SS = suspended solids

The data indicates that some beta emitting radionuclides attach to the suspended solids. As the suspended solids in TK-01 increased during the week's operation, more beta activity was associated with the suspended solids that were being rejected by the TUF, and hence less was passed through to the permeate. A reduction in the gross beta values in the TUF permeate was thus observed as the week progressed.

### **Reverse Osmosis**

RO1 was commissioned on tap water and provided a normalised flux of about 35 Lh<sup>-1</sup>m<sup>-2</sup>. The flux dropped to about 25 Lh<sup>-1</sup>m<sup>-2</sup> after operating for 2 weeks. A CIP was carried out but only a slight improvement in flux to 27 Lh<sup>-1</sup>m<sup>-2</sup> was achieved from the cleaning. When C-line wastewater was fed to the plant, RO1 flux dropped immediately to about 10 Lh<sup>-1</sup>m<sup>-2</sup>. Repeated CIP could only partially recover the flux and rapid flux decline occurred when the RO unit was back on line. Biocide dosing of RO1 (300 ppm) for 15 minutes followed by a CIP seemed to restore the flux to the original value of 35 Lh<sup>-1</sup>m<sup>-2</sup> on the first day of the week but it dropped rapidly to 15 Lh<sup>-1</sup>m<sup>-2</sup> by the end of the week. Biological fouling from a contaminated CIP tank caused this rapid drop in flux. The same tank had been used previously to clean the TUF unit and bugs from the TUF unit had been left in the tank and contaminated the RO membranes during the CIP.

A separate 'bug kill program' with the biocide 'kathon' was then carried out which fully recovered the RO1 flux. Other than cross-contamination from the CIP tank, biological contamination was also found in the anti-scalant solution. Long stringy material was found floating in the chemical drum, fouling the membrane as it was being dosed into the RO unit. Improper shutdown of the RO units was also a cause of the flux decline in RO1. A CIP had not been carried out at the end of the run and the membranes were not preserved in SMBS solution when shutdown for more than 48 hours. Weekly biocide shock treatment of RO1 had also not been carried out and all these contributed to irreversible fouling of the RO1 membranes. Flux decline for RO1 was also found to be higher when the feed contained high levels of aluminium (17-20 ppm) and sulphate (300 ppm) ions. At these levels RO1 flux halved (from 22 to 11 Lh<sup>-1</sup>m<sup>-2</sup>) in just four days. After all these operational issues were resolved a flux of about 20 Lh<sup>-1</sup>m<sup>-2</sup> was found to be suitable for stable operation of RO1.

Flux for RO2 started at around 60  $\text{Lh}^{-1}\text{m}^{-2}$  on tapwater. With C-line feed, the flux dropped to around 50  $\text{Lh}^{-1}\text{m}^{-2}$ . Due to biological fouling caused by cross-contamination from the CIP tank and improper shutdown procedures, the flux decreased to 20  $\text{Lh}^{-1}\text{m}^{-2}$ . After these operational issues were resolved the flux recovered slowly to 50  $\text{Lh}^{-1}\text{m}^{-2}$  over time. The flux for RO2 was unaffected by the various feeds to the pilot plant as the quality of permeate from RO1 was consistently high (< 20  $\mu$ S cm<sup>-1</sup> in conductivity). Stable operation of RO2 was achieved by operating at a flux of around 30  $\text{Lh}^{-1}\text{m}^{-2}$ .

Rapid flux decline of the S2RO was observed within the first week of operation with flux dropping from

35 to 5 Lh<sup>-1</sup>m<sup>-2</sup> in just two weeks and reduced to only 2 Lh<sup>-1</sup>m<sup>-2</sup> when C-line wastewater was introduced. This sharp drop in flux continued into the eighth week. It was found that S2RO was operating at less than half the minimum reject flowrate recommended by the membrane manufacturer. A combination of low turbulence, scaling of the membrane surface and possible biological fouling was suspected to be the cause of the sharp flux decline. After the membranes were cleaned and sterilised, flux recovered to about 10 Lh<sup>-1</sup>m<sup>-2</sup> for C-line wastewater. Flux decline was reduced when the membranes were operated at improved operating conditions (e.g. the reject flow rate was adjusted to maintain sufficient cross-flow velocity across the membrane). The flux was maintained between 5-10 Lh<sup>-1</sup>m<sup>-2</sup> for the rest of the test period. The membranes were replaced about halfway into the pilot trials. An initial flux of 34 Lh<sup>-1</sup>m<sup>-2</sup> was obtained and this gradually dropped to 15-20 Lh<sup>-1</sup>m<sup>-2</sup>. Stable operation of S2RO was achieved by operating at a flux of around 15 Lh<sup>-1</sup>m<sup>-2</sup>.

### **RO** Rejection

The membrane pilot plant processed radioactive feedwater for a total of 36 days. B-line, mixed B and C line, and C-line spiked with radionuclides were used to investigate the rejection of radioactivity by the membrane plant. The schedule of tests using radioactivity feedwater is shown in Table X.

Week	Feed Type	Days of Operation	Units					
27	B/C (30/70)	7	TUF and RO					
28	B/C (30/70)	4	TUF and RO					
32	В	5	TUF only					
36	C + TICAN	4	TUF and RO					
37	C + TICAN	3	TUF and RO					
38	C + TICAN	4	TUF and RO					
39	C + TICAN	6	TUF and RO					
40	C + AMP	3	TUF and RO (except S2)					

 Table X
 Active Feedwater Tests

The amount of alpha and beta activity in the feed varied from week to week. Gross alpha values were always below the detection limit in the RO1 permeate and 99.5% of beta emitting species were rejected by RO1 (Table VII). RO1 completely rejected most of the radionuclides except for <sup>106</sup>Ru, <sup>137</sup>Cs, and <sup>131</sup>I. RO1 rejected on average 97.9% of <sup>106</sup>Ru, 98.7% of <sup>137</sup>Cs and 99.7% of <sup>131</sup>I. RO2 completely removed any remaining <sup>131</sup>I and <sup>106</sup>Ru but an extremely small amount of <sup>137</sup>Cs was still found in the RO2 permeate (0.3 BqL<sup>-1</sup>). The rejection of <sup>137</sup>Cs decreased at low feed concentration. The data in Table VII suggests the rejection in RO2 is lower than RO1 although both membranes are identical. The data suggests that the percent radioactive separation increased with feed activity. This same trend was noted by Deshmukh et al. (1987) with cellulose acetate membranes [4].

The radioactivity of the feed to RO was as high as 20 BqL<sup>-1</sup> for alpha activity and 1800 BqL<sup>-1</sup> for beta/gamma activity. Exposure of the RO membranes to the radioactivity at these levels did not seem to affect the rejection properties of the membranes. The rejection of the radionuclides was found to be consistent throughout the runs.

According to Arnal et al. [5], aromatic polyamide composite RO membranes are recognised to be the most resistant to beta and gamma radiation. The authors found the structure and transport parameters of the membranes to be unaffected by both beta and gamma radiation. A high <sup>137</sup>Cs selectivity of more than 90% by these membranes was also reported in their investigation. The data collected from the pilot trials were in agreement with these observations.

The quality of the RO2 permeate was consistently high in all tests (< 5  $\mu$ S cm<sup>-1</sup> in conductivity). The pilot plant IX system was never used to further improve the product water or for further radioactivity

removal.

# Recovery

The membrane pilot plant was run at different recoveries every week. The flow rates of each stream (permeate, reject and recycle) for each membrane unit was adjusted such that the flows were balanced and continuous operation of the plant could be maintained. This allowed minimal overflowing of the feed tanks and start-stopping of any units.

The recoveries from the two stage-two pass reverse osmosis system are given in Table XI. The recoveries were calculated from the average S2RO reject flowrate and the average feed supplied by the TUF for the particular week as given by the following equation:

$$Recovery = 1 - \frac{Average S2RO Reject Flowrate}{Average TUF Permeate Flowrate}$$
(Eq.2)

The assumptions made in the calculation are:

- There is no overflowing of the feed tanks
- Continuous operation of each RO unit (RO1, RO2 and S2RO) was maintained throughout the week.

Table XI Recovery of the RO System							
Week	Feed	% Recovery	Week	Feed	% Recovery		
0	Tapwater	97.2	18	C (hypo)	73.1		
1	Tapwater	97.6	19	C (hypo)	60.5		
2	Tapwater	96.7	20	C (hypo)	67.5		
3	Tapwater	96.8	21	C (hypo)	68.3		
4	C-line	95.9	22	C (alum)/no blowdown	67.2		
5	C-line	94.4	23	C-line	69.0		
6	C-line	no data	24	C-line	72.3		
7	C-line	no data	25	C (hypo)	65.8		
8	C-line	89.9	26	C (hypo)/no blowdown	65.0		
9	Tapwater	97.1	26C	C (5 ppm hypo)	76.5		
10	Tapwater	96.9	27	B/C	69.4		
11	Tapwater	95.7	28	B/C	43.5		
12	Tapwater	96.6	32	B (alum)	69.0		
13	Tapwater	96.5	36	C-active	79.4		
14	Tapwater	95.7	37	C-active	77.9		
15	C-line	90.3	38	C-active	78.3		
16	C (hypo)	89.7	39	C-active	80.1		
17	C (hypo)	72.3	40	C-active	69.3		

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The RO system was run at high recoveries at the start of the pilot trials. Recoveries at more than 95% and 90% were achieved for tapwater and C-line feed respectively from Weeks 0 to 16. These high recoveries were not sustainable for the C-line wastewater and recoveries were lower in the subsequent weeks to control the flux decline in the RO membranes. At recoveries between 65-75%, the flux of the RO units appeared to be more stable. These results are consistent with two stage RO systems. Recoveries of up to 80% could be achieved in the pilot plant without negative impacts on the RO flux.

The performance of RO2 was unaffected by the recovery of RO1 or the type of feed water to the pilot

plant. This was because the feed to RO2 was relatively constant in quality. As long as the quality of the RO1 permeate is maintained the performance of RO2 will be stable and reliable. RO2 will perform consistently provided biological fouling does not occur in RO2 through improper shutdowns (not preserving the membranes in a sterile environment) or cross-contamination from other parts of the plant during cleaning.

### CONCLUSIONS

The following summarizes the results from the operation of the pilot plant:

- For all tests, no significant radioactivity was measured in the product water. The product water was of high purity, typically < 5  $\mu$ S/cm in conductivity. Ion exchange was not required for final product polishing.
- The tubular ultrafilter removed up to 96% of alpha activity and 82% of beta activity.
- Rejection of residual activity in the reverse osmosis unit was close to 100% in Pass 1.
- 'C' line water tended to slowly plug the pores of the ultrafilter with colloidal/organic species (over the course of a year). This is not considered difficult to alleviate, as changing membrane type, pore size and or using a backflush system would control this.
- 'B' line water caused dramatic and irreversible fouling of the ultrafilter membrane. This was determined to be due to the very high surfactant loading in the 'B' line water, originating from radiopharmaceutical production activities. Removal of these species by pre-treatment would be required to eliminate this negative effect, and requires further investigation.
- Design data has been obtained for the full-scale plant.

The operation of the pilot plant has been successful, although further work on the pre-treatment of 'B' line wastewater is required before full-scale implementation.

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