Cesium Removal from R-Reactor Building Disassembly Basin Using 3M Empore® Web-Membrane Filter Technology

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**WESTINGHOUSE SAVANNAH RIVER COMPANY
SAVANNAH RIVER TECHNOLOGY CENTER**

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USING 3M EMPORE® WEB-MEMBRANE FILTER TECHNOLOGY (U).**

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SAVANNAH RIVER SITE

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Keywords:

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SUMMARY

A seven-day demonstration of the use of 3M Empore® membrane filter loaded with ion exchange material (potassium cobalt hexacyanoferrate (CoHex)) for cesium uptake was completed at the R-Disassembly Basin. The main goal of the demonstration was to evaluate the ability of the Process Absorber Development unit (PADU), a water pre-filtration /CoHex configuration on a skid, to remove cesium from R-Disassembly Basin at a linear processing flow rate of 22.71 liters (6 gallons) per minute.

The average radioactive cesium concentration in the R-Disassembly Basin water is 8.340 E-02 $\mu\text{Ci/L}$. Over fifty-five thousand gallons of R-Disassembly Basin water was processed through the unit without a cesium breakthrough, that is, the effluent after treatment with CoHex, contained less than detectable amounts of radioactive cesium. The following conclusions can be drawn from the seven-day demonstration:

- After processing over 210,000 liters (>55,000 gallons) of the R-disassembly basin water there was no cesium-137 breakthrough.
- The flow rate, approximately 6 gallons of R-Disassembly Basin water per minute, is sufficiently low enough to ensure that there is sufficient contact

time for ion exchange reactions in the spiral membrane/CoHex arrangement. This flow rate translates to approximately 29 gallons per minute per square foot, which would meet most enduser flow rate requirements for polishing applications to remove cesium from aqueous streams.

- The minimum cesium-137 decontamination factor (D_f) obtained with this CoHex in a cartridge treatment is 366. At lower cesium detection limit extrapolation ($6.523\text{E-}08 \mu\text{Ci/mL}$) the calculated D_f is greater than 1600.
- Cesium concentration in the Vertical tube storage area of the R-disassembly basin, where this demonstration was carried out, is $8.34 \pm 0.245\text{E-}02 \mu\text{Ci/L}$. This value is in agreement with the documented average R-Building Disassembly Basin cesium-137 concentration of $8.25\text{E-}02 \mu\text{Ci/L}$.
- The calculated total amount of cesium-137 chemisorbed on the CoHex in a cartridge unit (six cartridges per unit) is 17,528 μCi , which is approximately 2,921 μCi per cartridges.
- The pressure change across the PADU coarse and fine pre-filter units was 9.4 psi per 24 hours time period. Pressure reading changes were highest on the coarse pre-filter at 10.4 psi per 24 hours
- The coarse pre-filter will need to be changed approximately every 6.7 days and the fine pre-filter every 70 days for continuous operation of the PADU.
- At the end of the 7 day demonstration the gamma radiation dose rate on the external areas around the 11 gauge stainless steel housing (CoHex cartridge housing) was 70 mR/hr at 30 cm.

1.0 INTRODUCTION

The R-Reactor Disassembly Basin at the Savannah River Site (SRS), up until 1964 when the R- reactor was shut down, was used mainly for temporary storage of irradiated fuel to remove radioactive decay heat from the fuel assemblies. The basin contains no fuel, target assemblies or other nuclear reactor components. The R-Disassembly Basin pool has an estimated volume of (18,925,000 liters (5 million gallons) of tritiated water and 5.1 –7.6 cm of radioactive sludge on the basin floor. The sludge is primarily made up of corrosion products from aluminum-clad fuel assemblies and debris from underwater machining operations (1). The principal radionuclides of environmental release concerns in the Disassembly Basin are cesium-137, strontium-90 and tritium. The concentration of cesium-137 and strontium-90 in the R-Disassembly Basin [Table 2a] exceed

the Department of Energy (DOE) release limits of 3000 pCi/L for Cs-137 and 1000 pCi/L for Sr-90 (DOE order 5400.5).

One option being considered for the R-Reactor Building Disassembly Basin stabilization (removal of traces of radionuclides) calls for the use of ion exchange-based deactivation technologies to remove cesium-137 and strontium-90 (1). This ion exchange treatment of the basin water will ensure that the deactivated basin water will meet DOE ingestion limits and thus become acceptable for release to site surface water. However, although the treated water can be released to site surface water, such direct release decision has not been made yet, because tritium will still be a problem after cesium and strontium have been removed.

The R-Reactor disassembly basin is subdivided by thick concrete walls into seven other smaller sections: Vertical Tube Storage (VTS); Machine Basin (MB); Horizontal Bundle and Bucket Storage (HBBS); Emergency Disassembly Basin (EDB); Monitor Pin Basin Canal; Dry Cave; and Transfer Bay (TB). These concrete division walls have narrow vertical openings at strategic points to enable transfer of materials between sections without removing material from the water. The openings were closed by the installation of portable stop-logs when it became necessary to isolate and drain a section for maintenance. At present, most of the stop-logs in the R-Reactor disassembly basin are in the open position. The remaining stop-logs are inoperable.

Depth-wise, the basin can be divided into two sections: the west end, with a general depth of 9.15 meters (30 ft) and floor slab thickness of 2.3 meters (7.5 ft) and the east end, with 5.19 meters (17 ft) depth and 1.53 meters (5-ft) thick floor slab. Each section contains many relatively small areas called the Vertical Disassembly Machine areas that exceed 15.25 meters (50 ft) in depth.

This demonstration project, which was carried out in the VTS area of the R-Reactor Building Disassembly Basin, was aimed at evaluating technology which allows ion specific separations to concentrate low levels of cesium with the generation of minimum secondary waste. The 3M empore[®] technology that was evaluated for this cesium removal project was developed with support from the Department of Energy Efficient Separations and Processing (ESP) Crosscutting Program. Previous low-volume flow rate demonstrations with this technology had been carried out at the Pacific Northwest National Laboratory (PNNL) (2) and 3M at Idaho National Environmental Engineering Laboratory (INEEL) (3).

2.0 Scope of Project

The objectives of this ion exchange treatment project were:

- (1) To demonstrate the use of 3M empore[®] membrane/ion exchange filter integrated system as a unit that could be used to remove cesium from R-Disassembly Basin at a flow rate up to 22.7 liters (6 gallons) per minute.
- (2) To process a larger volume of cesium contaminated wastewater than had been previously demonstrated with the Process Absorber Development unit (PADU).
- (3) To obtain comparative cesium removal field data on the effectiveness of the new configuration for the (spiral wound) web-like 3M membrane in a cartridge.
- (4) To acquire field data on the regularity with which the coarse and fine pre-filters of the PADU will need to be changed under the R-Disassembly Basin conditions.
- (5) To provide field data for the scale up of the technology to a 190 liters (50 gallons) per minute using the R-Disassembly Basin pool.

This report documents the cesium removal data collected in the demonstration and the operation of the Process Absorber Development unit at a flow rate of 22.71 liters (6 gallons) per minute.

3.0 Equipment Description

3.1 Empore[®]CoHex Technology

The Empore[®]web-like membrane developed by 3M is an inert matrix of polytetrafluoroethylene fibrils, which can be loaded with element selective ion exchange material. In this case of cesium removal, potassium cobalt hexacyanoferrate (CoHex) is the active agent. The resulting membrane is configured into a cartridge and inserted into commercial filter housing. It is in this cartridge form that the CoHex is used in the R- Disassembly Basin cesium removal demonstration. The 3M absorber cartridge physical properties are summarized in Table 1. An effective pre-filtration component of the Process Absorber Development unit (0.45 micron coarse filter and 0.1 micron fine filter) was used upstream of the empore cartridges to remove R-Disassembly Basin particulate and prevent the clogging of the empore[®] cartridges during lengthy operation periods without supervision.

3.2 PADU Skid Design/ Modification

The Process Absorber Development unit (PADU) consists of: a portable skid unit containing pumps, piping, pre-filter system, and the 3M empore® membrane/CoHex cartridge. Figures 1a, 1b and 1c are, respectively, the PADU schematic diagram, the engineering design flow diagram and photo. As mentioned above, this PADU was previously used to treat the waste-water at 105-N Basin at Hanford for cesium-137 and strontium-90 removal. In order to meet the needs of the R-Disassembly Basin demonstration, the PADU was decontaminated and modified at the Savannah River Technology Center. In detail, the modified PADU consisted of:

- Two pre-filters assemblies. Each pre-filter is housed in a high-pressure stainless steel vessel. Both the coarse filter (first pre-filter unit) and the fine filter (second pre-filter unit) contained six filter cartridges with pore size of 0.45 and 0.1 microns, respectively. The six filter cartridges were arranged in parallel inside each vessel and the two vessels were connected in series. The coarse filter was connected directly to the R-Disassembly Basin water intake line via a 3.1 meter long stainless steel pipe (ID of 2.54 cm). Each pre-filter unit was equipped with a pressure gauge, a sampling port and a pressure release/water drain valve.
- Two positive displacement pumps arranged in parallel. The pumps could deliver water at approximately 3.78 and 22.71 liters per minute, respectively. Thus, the PADU could be operated at either 3.78 or 22.71 liters per minute. In this demonstration the PADU was operated in the 22.71 liters per minute mode.
- A high-pressure stainless steel vessel containing the 3M empore®/CoHex membrane. This unit trapped the radioactive cesium and was lowered about three feet into the basin water for shielding purposes. This vessel was also equipped with pressure gauge/pressure release valve and was connected to the rest of the PADU unit (about 30 meters away via a one-inch copper tubing) via a totalizing flow meter. The 3M empore®/CoHex membrane vessel was lowered into the basin water with a winch on a swing-A-frame crane unit. A 10-meter long copper line (effluent line) from the 3M empore®/CoHex membrane vessel contained a valve for obtaining samples before the treated R-Disassembly Basin water was returned into the disassembly basin.
- A 24-hour clock wired to indicate a self shut-off time, which by design was expected to take place when the pressure readings on the pre-filters is either above 100psi or below 15 psi.
- A high and low-pressure guard circuits. The high-pressure switch, located on the intake side of the PADU pre-filter system, was designed to initiate a shut-off of the PADU if the pressure reading on any of the pre-filters goes above 100 psi due to clogging. The low-pressure switch located after the fine guard filter, on the other hand, initiates the PADU shut-off when the PADU pressure is lower than 15 psi due to a leak in the PADU.

- The R-Disassembly Basin water intake orifice, about 1.53 meters down the basin, was guarded with an inlet strainer made of stainless steel. The strainer prevented large particles from going into the pre-filter units.

Adsorber	CoHex
Adsorber weight per cartridge	130 gm
Cartridge height	25.4 cm
Cartridge diameter	6.35 cm
Membrane Length	101.60 cm
Membrane width	24.13 cm
Membrane thickness	0.1524 cm
Adsorber bed volume [volume of active membrane]	0.374 Liters

Table 1. Adsorber cartridge properties⁺⁺.

4.0 Basin Water Quality and Field Test Operations

4.1 R-Reactor Disassembly Basin Water Quality

During the winter months when the major part of this PADU demonstration was carried out, a thin off-white layer of glassy and light reflecting film was observed on the surface of the R-Disassembly Basin pool. The film layer, which is probably composed of calcium precipitate and dust particles, made it difficult to see any item introduced into the basin water. During the warmer spring/summer months most of the film material was gone, and it was now quite easy to operate remote handling tools in the basin without any transparency problems associated with the water clarity. The appearance and disappearance of the off-white film seems to be associated with basin water temperature, which is quite cold in winter (R-Building Disassembly Basin areas are not heated in winter). The surface film material may be transition phases resulting from solubility changes of calcium compounds in cold winter months versus warmer months. The Disassembly Basin contains more than 10 ppm calcium.

The pH of the R-Disassembly Basin ranges from 7.06 to 7.66 with an average of 7.35 (ref. 1). The R-basin water quality data are summarized in Tables 2a and 2b below. The average values are based on data from individual sections of the R disassembly basin. The machine (MB) and the emergency basins (EB) contain the highest amounts of cesium-137 at 0.0001 $\mu\text{Ci/mL}$ in the supernate.

⁺⁺ Information from 3M

4.2 System Check, Sampling and Data Acquisition

Before running the R-Disassembly Basin water through the PADU, a final system check involved the pumping of more than 6,800 liters of distilled water through the PADU. During these trial runs the PADU was checked for leaks, sampling procedures, auto shut down responses resulting from system pressure greater than 100 psi and lower than 15 psi. The last two conditions were simulated by valve restrictions and intentionally inducing leaks in the PADU. After these check tests, the PADU required no monitoring other than periodic check for leaks during sampling periods. No leaks were observed during the demonstration.

The stainless steel housing containing the six CoHex cartridges was lowered into the basin (1.53 meters) and remained submerged in the basin through out to provide radioactive shielding. The processed water was returned to the basin about 30 meters from the intake point near the PADU to prevent agitation of the disassembly basin water.

Samples were collected from three different sampling locations or ports on the PADU (Figure 1a and 1b): pre-filtration samples (samples collected before entering filter housings #3 and 1), post pre-filtration samples (after leaving filter housing #3 and 1) and post cesium decontamination through 3M Empore® filter samples (filter housing #2). Each sampling port was fitted with a toggle switch, which could be flushed before sample collection. During the first four days of continuous operation of the PADU, samples were collected from each of the three ports at intervals of two hours beginning at 0900 hours (0900, 1100, 1300 and 1500 hours). A total of 12 samples were collected on a typical 24 hour time period (Table 3). Samples were collected in labeled 250-mL high-density polyethylene bottles, which were pre-rinsed with liquids from each specific sampling port before being filled to the 200-mL mark. External radiation measurements were made on the outside of the stainless steel pre-filter housings during each sampling period. The maximum radiation reading on the external pre-filter housing, 200 cpm (β/γ), was not that different from the initial background reading of the decontaminated PADU. After approximately 74 hours of continuous running of the PADU the coarse pre-filter was changed due to clogging of the filter with particulate from the basin. The maximum activity on the clogged coarse pre-filters was 2000 cpm (β/γ).

A sodium iodide (NaI (TI)) scintillation detector, housed in a lead shielded cavity to eliminate background influence from the disassembly basin, and interfaced to a computer was used in the field to obtain approximate activity for cesium-137 in all influent and effluent samples collected (gamma ray spectrum for Cs-137 at $\approx$ 662 KeV). A one-hour delay time in the estimation of cesium activity in the samples using the field detector was employed to minimize the effect of barium-137 (daughter products of cesium-137 with a half-life of 30 minutes). The Analytical Development Section (ADS) carried out the final analytical

quantification for cesium in all collected samples at the Savannah River Technology Center.

Radionuclide	R-Basin Liquid
Total beta/gamma	243 d/m/ml
Tritium	0.041 ($\mu\text{Ci/mL}$)
Cesium-137	8.25E-05 ($\mu\text{Ci/mL}$)
Strontium-90	23.5 pCi/mL
Co-60	Not determined
Am-241	Not determined

Table 2a. Summary of average radionuclide concentration in R-Reactor Disassembly Basin (Ref. 1). Average values are based on concentrations from VTS, MB TB, and EB sections of the R-Disassembly Basin.

Cation/Anion	R-Basin Liquid (Average concentrations) (mg/L)
Ca	12.107
B	< 0.011
Al	0.116
Ba	< 0.008
Ag	6E-05
Cd	< 0.004
Co	0.001
Cr	0.009
Cu	0.014
Fe	< 0.009
K	27.570
Mg	0.444
Mn	< 0.003
Na	15.220
Ni	< 0.015
Pb	< 0.007
Se	0.093
Si	1.020
Sn	7.1E-4
Sr	0.152
Zn	< 0.004
NO ₃	1.10 [#]
NO ₂	0.23
SO ₄	3.3
PO ₄ (Total)	0.02
F	0.31
Cl	12.2
Br	26.8

Table 2b. Summary of principal cation and anion concentration in R-Reactor Disassembly Basin.

[#] All anion data are from reference 1.

Process** Time, (hour)	Coarse filter pressure (psi)	Fine filter pressure (psi)	Clock	R-Basin Feed. [Cs-137] ⁺ ($\mu\text{Ci/mL}$)	CoHex Effluent. [Cs-137] $\mu\text{Ci/mL}$	Volume processed (Liters)	Bed volume (BV) BV= 2.244L (0.374 L *6)	Sampling Date
0.5	29	29	1440	8.79E-05	0.0 ⁺	5,095.06	2,70.53	2/10/98
1	29	29	1540	8.19E-05	0.0	6,345.60	2,827.80	2/10/98
16	36	30	0900	8.03E-05	0.0	29,385.11	13,095.00	2/11/98
18	38	31	1100	8.63E-05	0.0	32,059.40	14,286.72	2/11/98
20	39	31	1300	- ⁺⁺	0.0	34,741.46	15,481.93	2/11/98
22	40	31	1500	8.73E-05	0.0	37,398.75	16,666.11	2/11/98
40	47	32	0900	-	0.0	61,373.02	27349.83	2/12/98
42	48	32	1100	8.09E-05	0.0	64,044.85	28,540.49	2/12/98
44	49	32	1300	-	0.0	66,690.94	29,719.67	2/12/98
46	50	32	1500	8.48E-05	0.0	69,328.33	30,894.98	2/12/98
64	58	32	0900	-	0.0	93,141.28	41,506.81	2/13/98
66	58	32	1100	8.18E-05	0.0	95,624.24	42,613.30	2/13/98
68	60	32	1300	8.41E-05	5.14E-8	98,943.67	44,092.55	2/13/98
70	60	33	1500	7.82E-05	0.0	100,591.67	44,826.95	2/13/98
72	60	33	1300	8.54E-05	0.0	106,599.98	47,504.45	2/17/98
74	61	33	1500	8.76E-05	0.0	108,732.07	48,454.58	2/17/98
coarse	filter	changed			coarse	filter	changed.	3/09/98
0	38	36	1300	7.76E-05	0.0	113,036.38	50,372.72	3/09/98
2	38	36	1500	8.45E-05	0.0	115,488.68	51,465.54	3/09/98
20	45	38	0900	-	0.0	139,825.47	62,310.82	3/10/98
22	55	39	0900	8.10E-05	0.0	171,501.00	76,426.47	3/11/98
25.5	56	39	1430	8.33E-05	0.0	178,493.03	79,542.35	3/11/98
44	65	40	0900	8.73E-05	0.0	203,140.95	90,526.27	3/12/98
49.45	67	40	1445	8.07E-05	0.0	210,167.80	93,657.67	3/12/98
Average	NA= Not applicable	NA	NA	8.340E-05 $\pm$ 0.245E-05	$\approx$ 0.0	NA	NA	NA

Table 3. Summary of PADU data. Coarse pre-filter was changed after the first 74 hours of continuous operation.

5.0 RESULTS AND DISCUSSIONS

External radiation activity measurements for the stainless steel pre-filter housings were never greater than 400 dpm/mL β/γ at 30 cm, indicating that no appreciable amount of radioactive cobalt particles were present or trapped on the pre-filter units as was expected. The slightly higher than background reading on the housings were due to incomplete decontamination of the PADU.

** Process time in hours is interval between sampling periods.

⁺ Weight mean activity; ADS #s 3-104464 to 3104468 and 3107658 to 3107706.

^{*} Less than minimum detection limit; averaged 2.277E-07 and 6.523E-08 $\mu\text{Ci/mL}$ for 1000 seconds and 10,000 seconds real time counting, respectively.

⁺⁺ Feed was not analyzed.

Field estimates of cesium activity in all samples collected, using the portable sodium iodide scintillation detector, showed that the CoHex treated samples had no detectable cesium activity. Both the pre-filter and post filter basin water samples responded positively to cesium activity. The analytically determined average cesium concentration in the R-Disassembly Basin (Vertical tube storage area) is $8.34\text{E} \pm 0.245 \text{E-}05 \text{ } \mu\text{Ci/mL}$ (Table 3). This value is consistent with a value of $8.25 \text{E-}05 \text{ } \mu\text{Ci/mL}$ (Table 2a) documented in previous reports on the R-disassembly basin water chemistry (Ref. 1). The cesium concentration in the basin water samples coming through the CoHex filter, after processing more than 210,000 liters (55,000 gallons) of basin water, was less than the cesium-137 minimum detection limit of $2.277\text{E-}07 \text{ } \mu\text{Ci/mL}$ (gamma scan counting time of 1000 seconds with average mean activity uncertainty of 2.44%). As a result, a value of zero was assigned to the entire CoHex treated samples; in other words, there was no cesium breakthrough after processing over 210,000 liters of the disassembly water.

Since the cesium decontamination factor, D_f (initial cesium concentration in the basin/final cesium concentration), at this detection limit is undefined because the final cesium concentration in the CoHex treated samples was assigned a value of zero, the minimum D_f was estimated by using the magnitude of the minimum detection limit as being equivalent to the final cesium concentration. Based on this assumption, the estimated D_f value is 366 ($8.34\text{E} \text{E-}05 \text{ } \mu\text{Ci/mL} / 2.277\text{E-}07 \text{ } \mu\text{Ci/mL}$). To estimate an upper limit for the D_f value, one of the CoHex treated samples was re-analyzed for cesium by counting for 10,000 seconds instead of 1000 seconds. A new minimum detection limit of $6.523\text{E-}08 \text{ } \mu\text{Ci/mL}$ with a higher activity uncertainty of 28.29% was established. The value of cesium concentration in this particular sample, at this new and lower detection limit was $5.14\text{E-}08 \text{ } \mu\text{Ci/mL}$ (Table 3). Thus, the upper calculated D_f limit is 1,623 ($8.34\text{E} \text{E-}05 \text{ } \mu\text{Ci/mL} / 5.14\text{E-}08 \text{ } \mu\text{Ci/mL}$). This large D_f value shows that the cesium uptake efficiency by CoHex, in the presence of competing ions like potassium and sodium (cesium selectivity), is significantly large.

The DOE release limit for cesium-137 is 3000 pCi/L (3pCi/mL). To attain this compliance limit for the R-disassembly basin cesium concentration of $8.34\text{E} \text{E-}05 \text{ } \mu\text{Ci/mL}$ using CoHex ion exchange technology, the expected D_f is 27.8 ($8.34\text{E} \text{E-}05 \text{ } \mu\text{Ci/mL} / 3\text{pCi/mL}$). Hence, using PADU/CoHex set up for treating the R-disassembly basin, the minimum estimated cesium decontamination factor, D_f , is 13 times better than what the DOE order calls for (366/27.8), and 58 times better when the upper D_f value estimate is used for the comparison.

The calculated total amount of adsorbed cesium in the CoHex cartridges is 17,528 μCi ($55,526.5 \text{ gallons} \times 3.785 \text{ liters/gallon} \times 1000 \text{ mL/Liter} \times 8.34\text{E-}05 \text{ } \mu\text{Ci/mL}$), which is approximately 2,921 μCi per cartridge. At the end of the demonstration,

the external radiation dose rate on the stainless steel housing holding the CoHex cartridges was 70 mR/hour at 30 cm (gamma).

Since treated basin water was released back into the disassembly basin only about 30 meters away from the PADU intake line, one would expect a dilution effect, that is, lower cesium concentration at intake line with time. The data collected shows that this is not the case (Table 3). There are, in fact, no significant or measurable changes in cesium concentration from the basin water acquired at the beginning and at the end of the demonstration. The average cesium concentrations for the first 20 hours of PADU operation and the last 20 hours are, respectively, $8.41 \pm 0.43 \text{ E-05 } \mu\text{Ci/mL}$ and $8.31 \pm 0.37 \text{ E-05 } \mu\text{Ci/mL}$. The above result was expected because of the large volume of water in the R-Building disassembly basin (5,000,000 gallons) and the possibility for the leaching of equilibrium cesium in the basin sludge back into the liquid phase.

Data used in the plots in Figures 2-5 are from the first 74 hours of continuous operation of the PADU, that is, before the change out of the coarse pre-filters. Pressure reading on the coarse pre-filters housing showed a significant change every 24 hours. The increase in pressure per 24 hours time period averaged about 10.4 psi, Figure 2. The fine pre-filter pressure changes were significantly lower at about 1.0 psi per 24 hours, Figure 3. These changes in pressure due to the clogging of the pre-filters can be attributed to the presence of particulate matter in the R-disassembly basin. However, recent discussions and a better understanding of the basin water chemistry have shown that the basin water is super saturated with calcium ions (Table 2b). The super saturated calcium ion media, when disturbed such as when the water comes into contact with the pre-filter membranes, may precipitate as carbonates and thus may be the main pre-filter clogging culprit. A plot of the PADU continuous operation time (hours) versus pressure difference across the coarse and fine pre-filter cartridges, Figure 4, shows that the pressure changes across the pre-filter units was 9.34 psi per 24 hours.

After 74 hours of continuous processing of R-disassembly basin water through the PADU, the coarse pre-filter pressure reading went up to 61 psi and since that was on a weekend, the PADU was shut down. The six coarse pre-filters cartridges were changed after that weekend and the unit brought back into service. At 10.4 psi per 24 hours pressure increase the PADU would have automatically shut itself down by the end of that weekend, as it was designed to operate if the pressure of the system went above 100 psi.

The R-Disassembly Basin water flow rate across the PADU, based on the totalizing flow meter, averaged 22.14 liters (5 gallons) per minute. The flow rate determined from a plot of total volume of water processed with time, Figure 5, showed that the actual flow rate was 22.71 liters (6.0 gallons) per minute (slope of the linear plot).

The calculated CoHex cartridge intake surface area is 0.019 m^2 (all six cartridges in parallel), and based on a linear flow rate of 22.71 liters per minute the flow rate per unit area was $1,195.8 \text{ liters/minute/m}^2$ ($29.35 \text{ gallons/minute/ft}^2$). This number will seem to suggest a very short contact time between the CoHex and the basin water, probably in seconds. This flow rate per unit area meets industrial requirements. Most ion exchange based polishing applications in the industry are operated at flow rates of 611.31 to $2,037.1 \text{ liters/minute/m}^2$ ($15\text{-}50 \text{ gal/min/ft}^2$) (4).

The decontamination plan for the R-disassembly basin would eventually involve simultaneous removal of cesium-137 and strontium-90. However, due to the high concentration of calcium ions (up to 12 ppm in the supernate, Table 2b), which is an effective competing agent in Sr-90 removal using ion exchange approach, calcium would have to be removed before any effective Sr-90 decontamination can be achieved. Among some of the proposed approaches to limiting the effect of calcium is the adjustment of the basin water pH to about a pH of 10 with either sodium hydroxide or preferably sodium carbonate. This would result in the precipitation of the calcium as the carbonate followed by the application of centrifuge/filter techniques to remove the resulting calcium carbonate precipitates. The Cs-137 and Sr-90 bearing effluent would then be passed over cesium and strontium removing ion exchange columns or cartridges in series. The concentration of magnesium in the basin water, at 1.19 mg/L in the supernate, is also quite large in comparison to the concentration of strontium-90. Its effect as a competing ion in Sr-90 removal using ion exchange techniques may need further investigation.

6.0 CONCLUSIONS

- The linear flow rate, approximately 22.71 liters of R-Disassembly Basin water per minute, is sufficiently low enough to ensure that there is sufficient contact time for ion exchange reactions in the spiral membrane /CoHex arrangement.
- This linear flow rate translates to approximately $1,195.8 \text{ liters/minute/m}^2$ ($29.35 \text{ gallons per minute per square feet}$), which would meet most enduser flow rate requirements for polishing applications to remove cesium from aqueous stream (4).
- Cesium concentration in the Vertical tube storage area of the R-disassembly basin, where this demonstration was carried out, is $8.34 \pm 0.245\text{E-}05 \text{ }\mu\text{Ci/mL}$. This value is in agreement with the documented average R-disassembly basin cesium-137 concentration of $8.25\text{E-}05 \text{ }\mu\text{Ci/mL}$.

- The estimated minimum cesium-137 decontamination factor (D_f) obtained with this CoHex in a cartridge treatment, based a minimum cesium detection limit of $2.277 \mu\text{Ci/mL}$, is 366. This D_f value, at the R-Disassembly basin cesium-137 concentration of $8.34\text{E-}05 \mu\text{Ci/mL}$, is at least 13 times higher than the DOE requirement for the R-Disassembly basin. At lower cesium-137 detection limit extrapolation ($6.523\text{E-}08 \mu\text{Ci/mL}$) it can be shown that the actual D_f is greater than 1600.
- After processing over 210,000 liters (>55,000 gallons) of the R-disassembly basin water there was no cesium-137 breakthrough.
- The calculated total amount of cesium-137 adsorbed on the CoHex in a cartridge unit is 17,528 μCi , which is approximately 2,921 $\mu\text{Ci/cartridge}$.
- The pressure change across the PADU coarse and fine pre-filter units was 9.4 psi per 24 hours time period. Pressure reading changes were highest on the coarse pre-filter at 10.4 psi per 24 hours. Pressure reading changes on the fine pre-filter was approximately 1.0 psi per 24 hours.
- At this filter clogging rates in the R-Disassembly Basin, the coarse pre-filter will need to be changed approximately every 6.7 days and the fine pre-filter every 70 days for continuous operation of the PADU. Note that the PADU auto shut-off mechanism comes into play, when the pressure across the pre-filters or any part of the system is greater than 100 psi.
- At the end of the approximately 7 days demonstration the external gamma radiation dose rate on the 11-gauge stainless steel housing (CoHex cartridge housing) was greater than 70 mR/hr at 30 cm.

7.0 RECOMMENDATIONS

It would be of scientific interest to operate the PADU continuously until the CoHex is saturated with chemisorbed cesium-137 (breakthrough point). Such breakthrough information is useful in determining and comparing the theoretical and the actual volume of cesium bearing waste water that can be processed, as well as the validity of the batch distribution coefficient (K_d) for the ion exchange material (potassium cobalt hexacyanoferrate). However, such lengthy PADU operations will lead to radiation safety issues for the operators. Each CoHex cartridge will be overloaded with chemisorbed cesium up to the point where it becomes too "hot" to work with safely during dislodging/CoHex change and transport operations. In other words, a high radiation environment (>100 mR/hr) would be created. Therefore, only a maximum of seven days of continuous PADU operation before change of CoHex cartridges is recommended. These CoHex

cartridge change operations (opening of the 11-gauge stainless steel housing) must be carried out under water (inside the disassembly basin) using appropriate tools. The cesium-137 bearing cartridges must be shielded during transportation above the disassembly basin.

An appropriate strontium-90 ion exchange absorber, preferably an inorganic material such as sodium titanate, should be incorporated in series with the CoHex to evaluate the efficiency of Sr-90 removal in the presence of ppm levels of competing calcium ions in the R-Building Disassembly Basin.

Future plans call for the designing of another PADU, which will be capable of processing the R-Disassembly Basin water at 190 liters (50 gallons) per minute.

8.0 QUALITY ASSURANCE

All the demonstration activities and other related work were conducted in accordance with SRS site rules and regulations including quality assurance procedure manual 1Q43, SRTC Conduct of Research and Development (WSRC-IM-97-00024) and R-Reactor Area Work Clearance permit. The experimental procedures and data collected during this demonstration program are contained in laboratory notebook WSRC-NB-94-194.

The following Radiological Work Control Permits (RWPs) and PADU Operation Procedures were used in this demonstration:

- 98R-108
- 98FDD-104
- WPTS101

9.0**ACKNOWLEDGEMENTS**

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Figure 1a. R-Disassembly Basin PADU test equipment schematic

Figure 1b. R-Disassembly Basin PADU Engineering Design Schematic

Figure 1c. R-Disassembly Basin PADU Photo

Pressure changes with hours of operation for coarse filter

Figure 2. Pressure changes as a function of time for the 0.45 micron (coarse) Filter. Pressure changes per 24 hours on the coarse filter was 10.4 psi (see slope).

Pressure changes with hours of operation for fine filter

Figure 3. Pressure changes as a function of time for the 0.1-micron (fine) filter. From the slope data, pressure changes on the fine filter was 1.0 psi per 24 hours.

Pressure difference across the coarse and fine filters.

Figure 4. Pressure difference across the coarse and fine pre-filters as a function of time. The slope of the plot gives a value of 9.34 psi per 24 hours. Hence, filters had to be changed approximately every 7 days during continuous operation [initial system pressure ≈ 30 psi and auto upper level shut-off pressure of 100 psi].

Figure 5. Volume of R-Disassembly Basin water processed before change of coarse filter. R-Basin water flow rate through the COHEX cartridges, based on slope of the plot, was 6.0 gallons per minute. The calculated flow rate was 5.85 gallons per minute.

**CESIUM REMOVAL FROM R-REACTOR BUILDING DISASSEMBLY BASIN
USING 3M EMPORE® WEB-MEMBRANE FILTER TECHNOLOGY (U).**

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