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PARTICLE COUNT MONITORING OF REVERSE OSMOSIS WATER TREATMENT FOR REMOVAL OF LOW-LEVEL RADIONUCLIDES

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ABSTRACT

Laser diode particle counting technology and analytical measurements were used to evaluate a pilot-scale reverse osmosis (RO) water treatment system for removal of particulate matter and sub-picocurie low-level radionuclides. Stormwater mixed with Waste Water Treatment Plant (WWTP) effluent from the Rocky Flats Environmental Technology Site (RFETS), formerly a Department of Energy (DOE) nuclear weapons production facility, were treated. No chemical pretreatment of the water was utilized during this study. The treatment system was staged as follows: multimedia filtration, granular activated carbon adsorption, hollow tube ultrafiltration, and reverse osmosis membrane filtration. Various recovery rates and two RO membrane models were tested. Analytical measurements included total suspended solids (TSS), total dissolved solids (TDS), gross alpha (α) and gross beta (β) activity, uranium isotopes $^{233/234}$U and ^{238}U, plutonium $^{239/240}$Pu, and americium ^{241}Am. Particle measurement between 1-150 microns (μ) included differential particle counts (DPC), and total particle counts (TPC) before and after treatment at various sampling points throughout the test. Performance testing showed this treatment system produced a high quality effluent in clarity and purity. Compared to raw water levels, TSS was reduced to below detection of 5 milligrams per liter (mg/L) and TDS reduced by 98%. Gross α was essentially removed 100%, and gross β was reduced an average of 94%. Uranium activity was reduced by 99%. TPC between 1-150μ were reduced by an average 99.8% to less than 1,000 counts per milliliter (mL), similar in purity to a good drinking water treatment plant. Raw water levels of $^{239/240}$Pu and ^{241}Am were below reliable quantitation limits and thus no removal efficiencies could be determined for these species.

NOMENCLATURE

Am	Americium
DPC	Differential Particle Counts
GAC	Granular Activated Carbon
gpm	Gallons per Minute
mg/L	Milligrams per Liter
mL	Milliliters
pCi/L	Picocuries per Liter (1x10^{-12} Ci/L)
Pu	Plutonium
RO	Reverse Osmosis
TDS	Total Dissolved Solids
TPC	Total Particle Counts
TSS	Total Suspended Solids
U	Uranium
α	Gross Alpha Activity (pCi/L)
β	Gross Beta Activity (pCi/L)
μ	Microns

INTRODUCTION

In 1990, stringent stream standards for radionuclides were promulgated by the Colorado Water Quality Control Commission (CWQCC) for the stream segments on and downstream of RFETS. These new radionuclide standards established maximum ambient concentrations for general gross α (7 pCi/L) and gross β (5 pCi/L) water contamination below previously existing federal guidance, and set stringent new standards for Pu (0.05 pCi/L), Am (0.05 pCi/L), and U (5 pCi/L) that did not previously exist. To comply with these new standards, a

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potential need to treat water effluent for radionuclides at sub-picocurie levels was identified.

Because the recent stringent discharge regulations also included many organic and inorganic constituents, physical separation removal technologies such as filtration were emphasized rather than chemical treatment methods. Chemical enhancement of filtration could be subsequently implemented if proven not to adversely impact the water quality.

A technology assessment for radionuclide removal was performed to determine available technologies for removal of radionuclides including gross α, gross β, Pu, Am, and U in the pico- to sub-picocurie activity level in primarily natural waters. Results of the technology assessment revealed that minimal work has been performed for consistent removal at or below 1 pCi/L. In an EPA study (USEPA, 1985), 55 water treatment plants were identified with total U inflow concentrations exceeding 10 pCi/L. Of the 55 plants, only one removed U to approximately 1 pCi/L at best effort (90% percent removal efficiency). This plant utilized enhanced conventional water treatment type systems. Results of a study by Jelinek and Sorg (1988) of an elementary school drinking water system in Colorado showed removal of U from raw groundwater at 50 pCi/L or greater to a treated level of less than 1 pCi/L; and in some cases to 0.10 pCi/L U at best effort. The school water treatment system consisted of prefiltration and ion-exchange columns. Another study (Moritz and Hoffman, 1994) utilized 2μ absolute-rated cartridge filtration without chemicals or physical pretreatment to treat raw pond water with concentrations of Pu and gross β at 0.044 pCi/L and 4.7 pCi/L, respectively. This method reduced the Pu to 0.009 pCi/L in the filtrate, but showed little or no reduction of gross β. Pu and U removal using reverse osmosis was also demonstrated to approximately 1 pCi/L and 4.4 pCi/L, respectively, in Plock et al. (1981), but this method used extensive chemical treatment and pH adjustment. Plock also recommended using ultrafiltration upstream of the RO membranes to reduce scaling and improve membrane life.

Information provided by the technology assessment, as well as general knowledge of radionuclides in natural waters and the environment, suggests that U exists primarily in the dissolved particle size range, ($<0.45\mu$ using standard analytical terms). A significant portion of Pu activity in environmental waters is usually associated with particulate $>0.45\mu$. Am, being a decay product of Pu, is assumed to be associated with the Pu size fraction, but little data exists for this species at sub-picocurie levels in the environment.

The filtration technology evaluated during this study was RO. RO has proven to be an effective method for removing particulate and dissolved solids as well as chemical contaminants to very low levels, and does not require chemical pretreatment. Ultrafiltration was also investigated during this study as a method to reduce RO membrane scaling, thereby eliminating the need to regenerate the membranes by acid rinsing. This also helped achieve the project goal of no chemical treatment.

METHODS/EXPERIMENTAL

An experimental pilot test was conducted at the RFETS stormwater detention Pond A-4 during the summer of 1994. The primary focus of this test was to measure RO system performance for sub-picocurie radionuclide and particulate removal. A RO system was mobilized, set-up, operated, demobilized, and subsequently returned to the subcontractor. Once at the test site, set-up and start-up/operational testing required approximately one day using a crew of two engineers and one field operator. Henceforth, operations were conducted by the operator with engineering technical support provided when necessary. The test was conducted for approximately one month, during which time technical staff evaluated the effects of operational parameters and recovery values on system performance. RO performance was determined by field measurement, chemical and radiochemical analyses, and particle counting methods.

Raw water was collected from the pond with a floating influent line so as not to withdraw sediment, and was pumped continuously through the treatment system at a rate of 3-5 gallons per minute (gpm) during the test sequences. Treated water was discharged back into the pond at a distance so as not to impact the influent water quality.

A schematic of the pilot test is shown in Figure 1. The treatment system was staged as follows: multimedia filtration, granular activated carbon (GAC) adsorption, hollow tube ultrafiltration, and reverse osmosis membrane filtration. A photograph of the interior of the trailer-mounted system is shown in Figure 2.

The water was first treated with multimedia filtration for gross suspended solids and algae removal; then by GAC for the adsorption of any trace organics that might contribute toward fouling and scaling of the RO membranes. A spiral-wound cartridge filter nominally

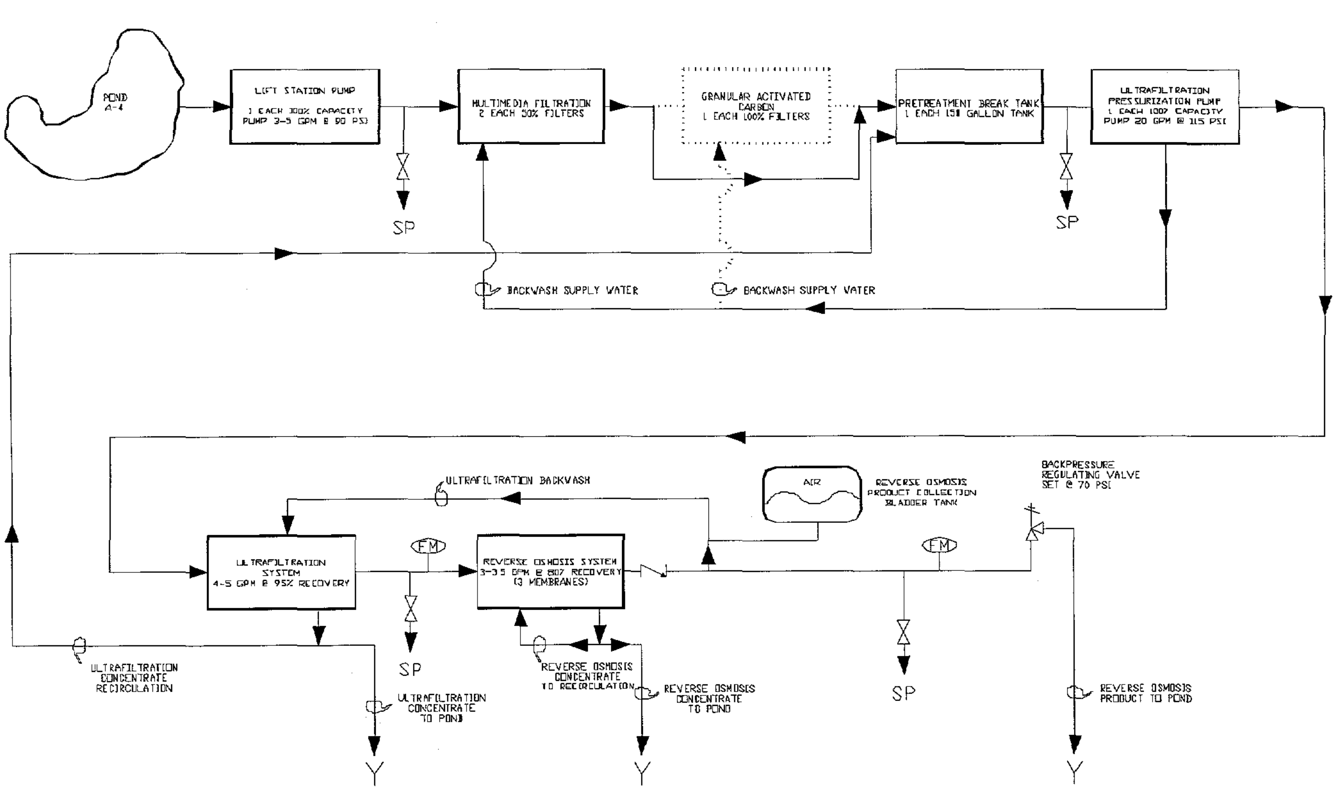

FIGURE 1 - REVERSE OSMOSIS PILOT TEST SCHEMATIC

rated at 0.5μ was positioned downstream of the GAC to prevent any media fines from entering the ultrafilters. Next were two parallel hollow-tube Romicon® model HF53-20-GM80 ultrafilters nominally rated at 0.005μ. The ultrafilters were 3 inches in diameter by 43 inches long, and produced a filtrate product stream and a reject concentrate stream. Flow from the ultrafilters was monitored at the product and concentrate water streams. Another spiral-wound cartridge filter nominally rated at 0.5μ was positioned downstream of the ultrafilters to prevent fines from entering the RO membranes in the event the ultrafilters were bypassed. The final treatment step utilized three parallel RO membranes. The two competitive spiral-wound thin-film composite RO membranes compared during the test were Filmtec® model BW30-4040 and Fluid Systems® model 4021LP. Both membranes were approximately 4 inches in diameter by 40 inches long, and produced a filtrate product stream and a reject concentrate stream. The concentrate streams from the ultrafilters and RO membranes, and the backflushing streams from the multimedia and GAC were discharged back into the pond.

System recovery rates for the RO membranes were varied between 70% and 85% during the test sequences. Recovery rates define the percentage of water produced as product water, with the remaining percentage rejected as concentrate. In this pilot unit the concentrate stream may be fractioned to recirculation back into the treatment system and partially managed as a waste stream.

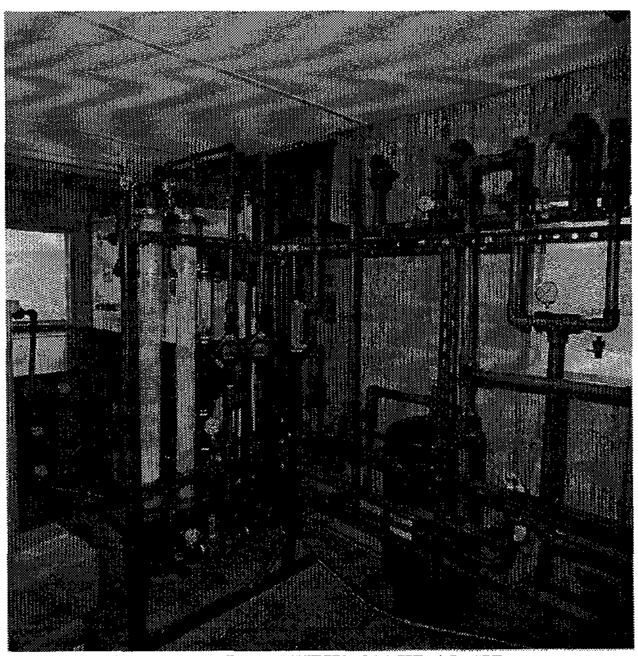

FIGURE 2 - INTERIOR VIEW OF TRAILER

The sample locations are shown in Figure 1 and described as follows. The first location for sample collection was the raw influent water supply line into the trailer. The second sampling point was after multimedia filtration and the third sampling point was after GAC adsorption. The fourth sampling point was after hollow tube ultrafiltration. The fifth sampling point was product water filtrate from the RO membranes. The sixth and final sampling point was the concentrate water stream rejected by the RO membranes.

Field parameter and chemical analysis samples were collected throughout the pilot test and performed according to APHA (1989) and EPA protocol. Radiochemical samples were taken on four days, and included two successive days during each RO element model test. Radionuclide analyses included gross α and β, Pu, Am, and U. Radionuclide analyses were performed according to methodologies reported in EG&G (1993). Particle count samples were collected daily during the pilot test. Particle count analysis was performed with Hiac\Royco® equipment including a model 9064 counter, model HRLD-150 laser diode sensor, and a model 3000 batch sampler. Further details concerning particle counting methods and operations are presented in Moritz and Hoffman (1993).

RESULTS AND DISCUSSION

The TSS, TDS, and radionuclide results are tabulated and presented in Table 1. The particle count data presented in Table 2 and plotted in Figure 3 compares the DPC in raw Pond A-4 water to the DPC after each treatment stage. Figure 4 is a plot comparing the DPC in RO product water to finished water from two local drinking water treatment plants. Filtration efficiency of each stage is also shown in Table 2, and was calculated using the following equation:

Percent Filtration Efficiency = 100 * (1-(Upstream DPC or TPC / Downstream DPC or TPC))

Results from this pilot test indicate the following performance by the RO treatment system:

- TSS were reduced from a maximum level of 38 mg/L to less than 5 mg/L.
- TDS were reduced from a maximum of 310 mg/L to less than 5 mg/L.
- Gross α, gross β, $^{233/234}U$, and ^{238}U were removed by >90% to near the detection limits of the measurement methods.

Analyte (1)	Raw water	Ultrafiltered	Product water
Recovery rate of 70% with Filmtec membranes			
TSS (2)	18	DNA (3)	<5
TDS (2)	310	DNA	<5
Gross alpha	2 ± 1	1 ± 1	0.0 ± 0.6
Gross beta	9 ± 2	4 ± 2	0.8 ± 1.3
U 233/234	0.791 ± 0.082	0.289 ± 0.038	0.002 ± 0.005
U 238	0.744 ± 0.077	0.322 ± 0.041	0.000 ± 0.003
Recovery rate 85% with Filmtec membranes			
TSS	DNA	DNA	<5
TDS	DNA	DNA	<5
Gross alpha	3 ± 1	1 ± 1	0.1 ± 0.6
Gross beta	9 ± 1	3 ± 2	0.3 ± 1.6
U 233/234	0.762 ± 0.087	0.300 ± 0.040	-0.003 ± 0.003
U 238	0.786 ± 0.089	0.308 ± 0.040	-0.003 ± 0.002
Recovery rate 70% with Fluid Systems membranes			
TSS	DNA	DNA	<5
TDS	DNA	DNA	<5
Gross alpha	1 ± 1	0.4 ± 0.8	0.0 ± 0.6
Gross beta	8 ± 2	2 ± 1	0.7 ± 1.5
U 233/234	0.829 ± 0.078	0.249 ± 0.031	0.015 ± 0.006
U 238	0.854 ± 0.080	0.251 ± 0.031	0.013 ± 0.006
Recovery rate 85% with Fluid Systems membranes			
TSS	38	DNA	<5
TDS	280	DNA	10
Gross alpha	2 ± 1	0.5 ± 0.7	-0.1 ± 1.3
Gross beta	10 ± 2	1 ± 2	0.2 ± 1.4
U 233/234	0.931 ± 0.094	0.271 ± 0.033	0.004 ± 0.005
U 238	0.903 ± 0.092	0.269 ± 0.033	0.004 ± 0.004

TABLE 1 - ANALYTICAL RESULTS SUMMARY

(1) All radionuclide data in pCi/L.
(2) TSS and TDS data in mg/L.
(3) DNA, data not available.

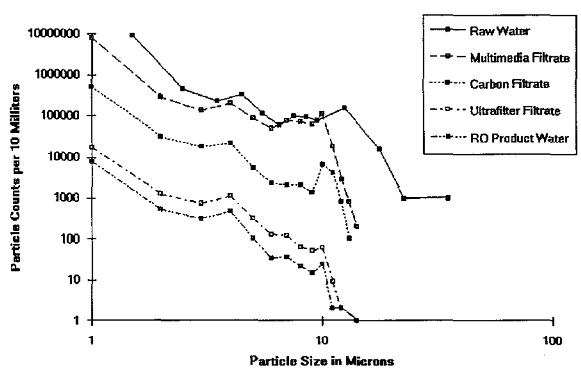

FIGURE 3 - PILOT TEST TREATMENT STAGE PARTICLE DISTRIBUTIONS

- Significant particulate material between 1 and 150μ was removed by each successive treatment stage. The removal efficiency of the multimedia stage was the least effective at 14.1%. The overall treatment system removal efficiency was >99.9%.
- The final RO treatment system product water contained <1,000 TPC per mL compared to raw water levels >1,000,000. This water quality is

Size Microns	Raw Water	Multimedia Filter	Multimedia Efficiency	Carbon Filter	Carbon Efficiency	Ultrafilter	Ultrafilter Efficiency	RO Product	System Efficiency
1	8973000	8004800	10.79%	522900	93.47%	17280	99.78%	7758	99.91%
2	458000	285900	37.58%	32000	88.81%	1284	99.55%	549	99.88%
3	231000	132700	42.55%	17900	86.51%	735	99.45%	320	99.86%
4	333000	205000	38.44%	21800	89.37%	1142	99.44%	482	99.86%
5	114000	85400	25.09%	5400	93.68%	318	99.63%	103	99.91%
6	59000	50000	15.25%	2300	95.40%	132	99.74%	35	99.94%
7	96000	76900	19.90%	2100	97.27%	121	99.84%	36	99.96%
8	91000	71100	21.87%	2100	97.05%	65	99.91%	21	99.98%
9	76000	61400	19.21%	1400	97.72%	52	99.92%	15	99.98%
10	152000	107900	29.01%	6600	93.88%	61	99.94%	24	99.98%
15	16000	17900	-11.88%	4100	77.09%	9	99.95%	2	99.99%
20	1000	2900	-190.00%	800	72.41%	2	99.93%	2	99.80%
25	0	800	n/a	100	87.50%	0	100.00%	0	n/a
30	1000	200	80.00%	0	100%	0	100.00%	1	99.90%
40	0	0	n/a	0	n/a	0	n/a	0	n/a
50	0	0	n/a	0	n/a	0	n/a	0	n/a
75	0	0	n/a	0	n/a	0	n/a	0	n/a
100	0	0	n/a	0	n/a	0	n/a	0	n/a
125	0	0	n/a	0	n/a	0	n/a	0	n/a
150	0	0	n/a	0	n/a	0	n/a	0	n/a
Totals	10601000	9102900	14.13%	619500	93.19%	21201	99.77%	9348	99.91%

TABLE 2 - SYSTEM DIFFERENTIAL PARTICLE COUNTS AND FILTRATION EFFICIENCIES

comparable to finished water from the RFETS and Pine Valley drinking water treatment plants.

- Varying the recovery rate between 70% and 85% had no observable effect on radionuclide removal.
- The radionuclide removal achieved with Film-Tec® RO membranes was indistinguishable from that achieved with membranes from Fluid Systems®.
- $^{239/240}$Pu and ^{241}Am levels in the raw water were below reliable quantitation limits and thus no removal efficiencies are presented.
- The GAC was bypassed early in the test because it caused immediate scaling of the RO membranes at recovery rates >50%. Discussions with the supplier and prior experience revealed this effect was due to the GAC not being acid washed prior to use.
- Chemical analysis results for organics, inorganics, salts, and metals are not presented in this report, but removal of these analytes was also very good.

CONCLUSIONS/RECOMMENDATIONS

Based on the results of this study, the following conclusions and recommendations are made:

- The RO pilot test demonstrated the viability and practicality of deploying mobile water treatment incorporating adsorptive/extractive technologies with no chemical enhancement. Particulate matter and low-level contamination by metals, salts, and radionuclides are effectively managed by this approach.

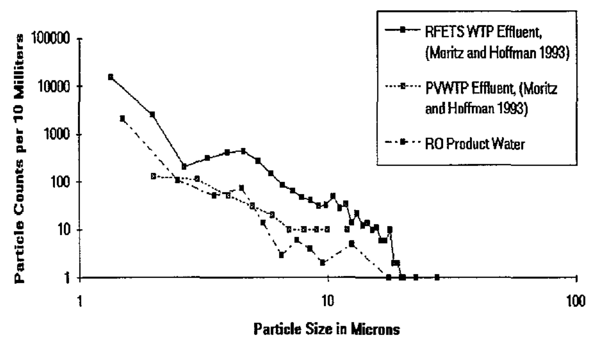

FIGURE 4 - REVERSE OSMOSIS AND WATER TREATMENT PLANT PARTICLE DISTRIBUTIONS

- The overall treatment system removal efficiency of particulate material between 1 and 150μ of >99.9% and final product water levels of <1,000 TPC per mL are comparable to the finished water quality from a good drinking water treatment plant.
- The 14.1% removal efficiency of the multimedia stage was lower than expected, based on data from

other studies of multimedia filters. Alternative configurations and media should be evaluated to improve performance.

- Compared to historical data, radionuclide levels in the raw water were typical for the RFETS ponds and representative of conditions prior to treatment. $^{239/240}$Pu and ^{241}Am levels in the raw water were below reliable quantitation limits and thus no removal efficiencies could be determined. However, Pu removal has been demonstrated in a pilot test performed with absolute rated cartridge filtration. Gross α, gross β, $^{233/234}$U, and ^{238}U were removed by >90% to near detection limits of the measurement methods.
- Since TSS is a gross measurement of suspended solids >0.45μ, it was expected to be removed by this treatment method. TSS were reduced from a maximum level of 38 mg/L to less than 5 mg/L. TDS, a measurement of solids <0.45μ, was also reduced in most cases from a maximum of 310 mg/L to less than 5 mg/L. These results prove this treatment method was highly effective at removing TSS and TDS.
- The GAC should be acid washed prior to use to prevent scaling of the RO membranes at recovery rates >50%.
- During demobilization, spent filter media and reverse osmosis membranes were removed and packaged as waste prior to offsite release of the RO treatment system. In practice, the replacement of these items would be necessary only when the effective service life was reached. As a result, extended periods of operation would normally be possible without removal, regeneration, and/or cleaning of the media or membranes.
- Chemical analysis results of inorganics, salts, and metals are not presented in this report, but removal performance was also very good for these analytes.
- The various recovery rates (70%-85%) and two membrane models very similar in construction produced indistinguishable radionuclide removal results. However, recovery rate drastically affects the system efficiency and amount of concentrate waste water produced. In order to minimize waste water production and maximize efficiency, consistent high recovery rates are required.
- If a large scale RO system (>100 gpm) is required for RFETS pond water treatment, another pilot test of a medium scale system (≅ 50 gpm) performed over an extended period of time at high recovery rates is recommended. This test would result in an accurate determination of low-level radionuclide removal, seasonal effects, membrane service life, waste production, and operating costs.

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