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Reverse Osmosis Applications to Low-Level Radioactive Waste

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REVERSE OSMOSIS APPLICATIONS TO LOW-LEVEL RADIOACTIVE WASTE

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The Westinghouse Hanford Company has been testing reverse osmosis for decontaminating liquid effluents and groundwater. Data demonstrating the capability of reverse osmosis to remove uranium, technetium, tritium, strontium, and cesium are reported. All of these contaminants, except tritium, were removed to less than regulated limits. Total alpha- and beta-emitter rejection data are included. The minimum secondary waste volume also was determined in the tests. Program data on the effectiveness of reverse osmosis on nonradioactive constituents were also collected but are not included in this paper.

1.0 INTRODUCTION

The Hanford Site at Richland, Washington, is operated for the U.S. Department of Energy (DOE) by Westinghouse Hanford Company. Since the Hanford Site was established in the 1940's, the operation of the various facilities has resulted in the contamination of liquid effluents and some groundwater with radioactive constituents. Westinghouse Hanford Company has been testing various technologies to determine their effectiveness in decontaminating these two types of liquids. Reverse osmosis (RO) has been applied to two process effluents and two groundwaters. Rejection data have been collected for uranium, technetium, tritium, strontium, cesium, and total alpha and beta.

2.0 CRITERIA FOR EVALUATION

The capability of RO to remove the radioactive contaminants was evaluated by comparing the final contaminant concentrations in the permeate with the administrative control values (ACV) or the Drinking Water Standards (DWS) (EPA 1976). The tests with the process effluents were compared with ACVs; the tests with groundwater were compared to the DWSs. The ACVs regulate the radionuclides received by the liquid waste disposal facilities and are regulated as a direct discharge to groundwater. The ACVs have been established so that groundwater concentrations meet 4% of the Derived Concentration Guides (established by DOE Order 5400.5) (DOE 1990) at the Hanford Site boundary on an annual average. The DWSs used are the most restrictive drinking water regulations that have either been adopted or are proposed.

2.1 TERMS

In this paper, the term permeate means "pure" water that permeates the membrane. The term concentrate means a substance that does not permeate the membrane. Also, the following terms are used in this paper.

$$\% \text{ Recovery} = \frac{\text{Permeate}}{\text{Permeate} + \text{Concentrate}} \times 100$$

$$\% \text{ Rejection} = \frac{\text{Feed concentration} - \text{Permeate concentration}}{\text{Feed concentration}} \times 100$$

A reference run is defined as the processing of a 0.2% sodium chloride solution through the membrane. The results are compared with previous reference runs to verify membrane integrity.

3.0 EXPERIMENTAL

3.1 SYSTEM DESCRIPTION

All of the tests were performed with an RO pilot system located in the 1706-KE Laboratory on the Hanford Site. A schematic of the system is shown in Figure 1. The RO system consists of two feed tanks, a pump, membrane, assorted pressure gauges, flowmeters, conductivity sensors, temperature

sensor, and valves. The membrane used was a Filmtec* FT-30, which is a thin film composite polyamide. It is spiral wound, and the dimensions are 2.5 in. by 40 in. with 23 ft^2 of membrane area.

The pilot system used for the first three studies consisted of one cartridge. Before the fourth test, the system was upgraded. The single membrane was replaced with a vessel containing two Filmtec FT-30 membranes, and the instrumentation was improved.

Because a full-scale operating RO system consists of several (usually six) membrane cartridges assembled in series in one vessel, the operation of a full-scale system was simulated in the pilot system by saving the concentrate from each pass and using it as feed for the next pass.

3.2 APPLICATIONS

3.2.1 B Plant Process Condensate

3.2.1.1 Description of Feed. The B Plant Process Condensate (BCP) is essentially distilled water with low levels of dissolved salts. The aqueous species present are either from entrainment of the salts or from condensation of volatile species with boiling points at or below the nominal boiling point of water. An approximate recipe is listed in Table 1. Cesium and strontium concentrations, which are of special interest, are 50 ppt and 20 ppt, respectively.

A synthetic BCP solution was prepared. The isotopes in the actual BCP are ^{137}Cs and ^{90}Sr. Dilute concentrations of the radionuclides ^{134}Cs and ^{85}Sr were used in the synthetic solution. These particular isotopes were selected because of their short half-life periods (^{134}Cs is 2.06 yr, and ^{85}Sr is 65.2 d) and because they can be measured by Gamma Energy Analysis. The percent rejection is expected to be the same for both isotopes of the individual species.

3.2.1.2 Description of Test. The synthetic feed solution was processed through a single membrane; the concentrate was collected and used as feed for a subsequent run. This procedure was repeated for a total of three passes. Feed pressure ranged from 182 to 187 lb/in^2. Temperature was about 28 °C. Feed flow was 2 gal/min, and concentrate flow was 1.6 gal/min. Samples were taken of the feed, concentrate, and permeate streams from each pass and analyzed for cesium and strontium.

3.2.1.3 Results. The results from the analysis are shown in Table 2. Cesium was rejected at 99.1% with less than 45 pCi/L in the permeate. Strontium was rejected at 99.9% with 149 pCi/L in the permeate. At these concentrations of radionuclides, the percent rejections attained produced a permeate well within the regulated limits.

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*Filmtec is a trademark of the Filmtec Corporation, Minneapolis, Minnesota.

At test concentrations, the strontium and cesium were reduced to well below the regulated limits. However, the percent rejection will decrease as the feed concentration increases. In addition, because of different contaminant limits, a rejection that will be adequate for one isotope may not be adequate for another. At a concentration of 50 ppt of ^{137}Cs in the stream, a rejection of 99.3% will be required to reach the ACV of 30,000 pCi/L. This percent rejection was not reached at the low concentration of 0.06 ppt in the synthetic solution and is not likely at higher concentrations. At a concentration of 20 ppt ^{90}Sr in the stream, a rejection of 99.29% will be required to reach the ACV of 20,000 pCi/L. This rejection may possibly be attained, because the percent rejection is 99.9% at the low concentration of 1.5 ppt in the synthetic solution. Further testing at higher concentrations is needed to provide confirmation of these assumptions.

An overall recovery rate of 48% was achieved. This is an acceptable recovery because the test consisted of only three membrane passes, which is the equivalent of half a full-scale assembly. About 50% recovery would be expected from one pass through a full-scale assembly.

After the BCP test was completed, the unit was rinsed thoroughly and a reference run performed. The pressures and rejection rates were the same as the initial reference run performed.

3.2.2 UO_3 Process Condensate

3.2.2.1 Description of Feed. The feed for this test was actual UO_3 Process Condensate. The UO_3 Process Condensate is a waste generated by the concentration and calcination of uranium nitrate hexahydrate to produce UO_3 powder. Water vapor and nitric acid fumes from the process are condensed, buffered with phosphoric acid, and neutralized with potassium hydroxide. Small amounts of other constituents that are either volatile species or entrained salts are also found in the stream. The expected constituents for the stream are shown in Table 3. The radionuclides of interest in this stream are uranium and ^{99}Tc.

3.2.2.2 Description of Test. This test consisted of three steps. In the first step, 55 gal of the unneutralized condensate were processed through the RO unit. Because the pH of the unneutralized condensate was below the tolerance range of the membrane (pH = 0.8), the manufacturer was consulted. A short run of 30 min or less was recommended. A 20 min, single-pass test was performed. Feed pressure was about 200 lb/in^2, temperature was 22 °C, feed flow was 1.8 gal/min, and concentrate flow was 1.5 gal/min. Samples of the feed, permeate, and concentrate were taken.

In the second step, the product streams were collected and combined, neutralized with potassium hydroxide to produce a pH of 2.5, and processed through the RO unit. The concentrate was collected and used as feed for subsequent passes. A total of six passes were completed in the second step. Samples of the product streams were taken after each pass.

In the third step, the pH of the product streams from the second step was raised to 5. Upon this addition, a precipitate formed. The precipitate was allowed to settle, and the test was completed by pumping from the top of the feed tank, taking care not to pump any of the precipitate. Approximately 1 L of feed was lost in this procedure. The remaining feed was processed for six passes. Samples of the product streams were taken after each pass.

The percent recovery was kept at about 14% for each pass throughout the second and third steps. Some general trends in other parameters were observed as follows.

- Feed flow rates ranged from 1.8 to 1.02 gal/min, decreasing gradually with each pass (Hodgson and Garrett 1989).
- System pressures ranged from 210 to 245 lb/in^2 and increased slightly with each pass (Hodgson and Garrett 1989).

3.2.2.3 Strontium Uptake. A sample of the precipitate that formed in step three was analyzed by Gamma Energy Analysis and trace amounts of ^{85}Sr were detected. The BCP synthetic solution (the previous test) was the only possible source for the ^{85}Sr.

The test plan for the BCP solution did not include any evaluation of radionuclide uptake; therefore, no activities were designed to determine this parameter. The ^{85}Sr was detected in the concentrate stream of a later, unrelated test. Because it was unexpected, the ^{85}Sr levels were not quantified. However, a mass balance was calculated with the analytical values and found to be within measurement uncertainties. Given the limited evidence, it seems that very little of the total ^{85}Sr was retained in the system.

3.2.2.4 Results. The rejection rate for uranium was greater than 99% and the rejection rate for ^{99}Tc ranged from 74% to 81%. The concentrations of uranium and technetium that remained in the permeate were well below the required ACVs and showed no evidence that pH adjustment affected the rejection rates. As expected, tritium was not rejected. (See Tables 4, 4a, and 4b.)

Overall recovery for six passes (one full-scale assembly equivalent) was about 60%. About 50% recovery would be expected from one pass through a full-scale assembly.

3.2.3 Groundwater from Well 200-W19-03

3.2.3.1 Description of Feed. The chemical constituents found in this groundwater are listed in Table 5. The total dissolved solids (TDS) are about 360 ppm.

3.2.3.2 Description of Test. The feed for the RO test was 770 gal of groundwater. As in the previous test, the concentrate stream was saved and used as feed for the next pass. Samples were taken of the permeate after each pass and the concentrate. The permeate was collected and sampled after each pass. After nine passes, a precipitate formed that was determined to

be calcium carbonate. Hydrochloric acid was added to dissolve the precipitate and processing was continued for five more passes. During the fourteenth pass, the flux decreased to 60% of the starting flux. At this time, the test was terminated.

Feed pressures ranged from 182 to 210 lb/in^2. Feed flow ranged from 1.6 to 1.8 gal/min, and concentrate flow ranged from 1.3 to 1.4 gal/min.

3.2.3.3 Results. The acid addition changed the test parameters considerably, so results will be discussed as before acid addition and after acid addition. The results before acid addition (Table 6) show that uranium and technetium were removed to well below the DWS. After hydrochloric acid addition, the rejection rates decreased for the measured constituents. This was expected because the salt passage through the membrane is a function of the concentration differential across the membrane. Usually, this rate decreases gradually and makes very little difference in the overall percent rejection. However, the addition of hydrochloric acid changed the concentration of ions in the feed by a considerable amount, and rejection rates of all the constituents were affected. The difference in rejection rates was most noticeable in the technetium results (compare Table 6 with Table 6a). Concentrations in the permeate were still below the DWS in all cases.

After processing through 14 passes (equivalent to 2-1/3 full-scale assemblies), the concentrate stream had been reduced to about 31 gal for an overall recovery of 96%.

3.2.4 Groundwater from Well 1-H3-2B

3.2.4.1 Description of Feed. The feed for this test was purge water from Well 1-H3-2B. Purge water is the water pumped from a monitoring well to purge the system before taking a sample for analysis. The constituents detected in the feed are shown in Table 7. The TDSs are about 90 ppm.

3.2.4.2 Description of Test. Approximately 58 gal of purge water were processed through the RO unit. The unit used for this test contained two spiral-wound membranes. The feed was pumped through the unit at pressures of about 150 lb/in^2. Pressures and temperatures varied slightly during the test. Conductivities of the feed and permeate increased with each pass as expected. Seven passes were performed. Samples were taken of the feed before processing, the permeate after each of the first six passes, and the final concentrate.

After the sixth pass, the volume of the feed had been reduced to about 12 gal. This volume allowed for only 5 min of operation in the seventh pass. This was not long enough for conductivity equilibrium to be established, so no permeate sample was taken after this pass.

3.2.4.3 Results. All of the measured constituents in the 183-H purge water were removed by RO to concentrations less than the DWS. Table 8 shows the values for the radioactive constituents compared to the DWS.

Masses for uranium did not balance; so each data point was investigated and a determination made as to its validity. The concentration in the feed was close to the value measured in monitoring activities for this well. The concentrations of the six permeate samples were all consistent with each other and at the values expected from previous RO testing experience. In addition, it was found that uranium analysis accuracy is highly dependent on the matrix, and as the salt concentration increases in the sample, the uranium analysis accuracy decreases. Therefore, the final concentrate value was disregarded and the feed and permeate concentration values for uranium were used in this evaluation.

The mass of the total beta-emitters that was in the concentrate was greater than that in the feed. Even if no beta-emitters were assumed to be in each of the permeate samples, the mass would not balance. Because the beta concentration in the feed was close to the value measured in monitoring activities for this well and the concentrations in all the permeate samples were below the detection limits, the final concentrate value was determined to be inaccurate. The percent rejection was determined by using the less than detection values measured in the permeate samples.

The final concentrate volume after seven passes through the two-membrane unit (equivalent to 2-1/3 full-scale assemblies) was 8.4 gal, and the overall recovery was 86%.

4.0 SUMMARY

The RO was effective at removing the cesium, strontium, technetium, and uranium to regulated limits at the concentrations tested in the four applications reported in this paper. Rejection rates were acceptable at low concentrations of contaminants in a variety of matrices. However, rejections of ^{137}Cs and ^{90}Sr at higher concentrations may not be adequate. Maximum volume reduction was not achieved in all cases because of the small feed volume or other test limitations, but recovery rates were better than expected.

5.0 REFERENCES

DOE, 1990, *Radiation Protection of the Public and the Environment*, DOE Order 5400.5, U.S. Department of Energy, Washington, D.C.

EPA, 1976, *National Interim Primary Drinking Water Regulations for Radiological Constituents*, EPA-570/9-76-003, U.S. Environmental Protection Agency, Washington, D.C.

Hodgson, K. M. and L. Garrett, 1989, *Demonstration of Technologies to Remove Contamination from Groundwater*, WHC-SA-0367-FP, Westinghouse Hanford Company, Richland, Washington.

WHC, 1989, *Environmental Compliance*, WHC-CM-7-5, REV 0, Westinghouse Hanford Company, Richland, Washington.

Table 1. B Plant Process Condensate Constituents.[a] (Units are in parts per million unless otherwise specified.)

Constituent	Concentration
$NaNO_3$	210
NaOH	101
$NaNO_2$	37
$Al(NO_3)_3 \cdot 9H_2O$	25
NaF	5
$Cr(NO_3)_3 \cdot 9H_2O$	1
Cesium	50 ppt
Strontium	20 ppt

[a]Based on an estimated analysis of B Plant feed and expected decontamination factors.

Table 2. Reverse Osmosis Testing of B Plant Effluent.

Nuclide	Feed (pCi/L)	Permeate (pCi/L)	Rejection (%)	ACV[a] (pCi/L)
Cesium	5,030	<45	>99.1	30,000
Strontium	214,000	<149	>99.9	20,000

[a]WHC, 1989, *Environmental Compliance*, WHC-CM-7-5, REV 0, Westinghouse Hanford Company, Richland, Washington.
Recovery = 48%
ACV = administrative control value

Table 3. UO_3 Process Condensate Constituents. (All units are parts per billion unless otherwise specified.)

Constituent	Concentration
Ammonium	78
Cadmium	2
Calcium	325
Chromium	66
Iron	290
Magnesium	26
Manganese	6
Mercury	1
Nickel	32
Sodium	380
Zinc	43
Nitrate	1 E+07
Phosphate	3 E+06
Total Organic Carbon	2 E+04
Total Organic Halide	2 E+02
Gross Alpha	1.8 E-06 μCi/mL
Gross Beta	2.4 E-06 μCi/mL
Technetium-99	7.2 E-04 μCi/mL
Gross Uranium	2.6 E-06 μCi/mL

NOTE: The condensate, as produced, has a pH of less then 2. It is neutralized to a pH between 6.5 and 8.5 before disposal.

Table 4. Reverse Osmosis Testing of UO_3 Process Condensate. (pH = 0.86)

Constituent	Feed (pCi/L)	Permeate (pCi/L)	Rejection (%)	ACV[a] (pCi/L)
Alpha	3,050	<23	>99.2	
Beta	4,630	276	94.0	
Technetium-99	1,350	<252	>81.3	4,000
Tritium	5.6 E+07	5.5 E+07	-	-
Uranium	4,627	6	99.9	200

[a]WHC, 1989, *Environmental Compliance*, WHC-CM-7-5, REV 0, Westinghouse Hanford Company, Richland, Washington.
Recovery = 12% (one pass)
ACV = administrative control value

Table 4a. Reverse Osmosis Testing of UO_3 Process Condensate. (Neutralized with KOH to pH = 2.5)

Constituent	Feed (pCi/L)	Permeate (pCi/L)	Rejection (%)	ACV[a] (pCi/L)
Alpha	190	<35	>81.6	
Beta	11,600	3,610	68.9	
Technetium-99	1,480	<381	>74.3	4,000
Tritium	4.9 E+07	5.0 E+07	-	-
Uranium	5,310	8	99.9	200

[a]WHC, 1989, *Environmental Compliance*, WHC-CM-7-5, REV 0, Westinghouse Hanford Company, Richland, Washington.
Recovery = 59% (6 membrane passes)
ACV = administrative control value

Table 4b. Reverse Osmosis Testing of UO_3 Process Condensate. (Neutralized with KOH to pH = 5)

Constituent	Feed (pCi/L)	Permeate (pCi/L)	Rejection (%)	ACV[a] (pCi/L)
Alpha	379	<36	>90.3	
Beta	12,600	3,680	70.8	
Technetium-99	946	175	81.5	4,000
Tritium	4.9 E+07	4.9 E+07	-	-
Uranium	3,180	11	99.6	200

[a]WHC, 1989, *Environmental Compliance*, WHC-CM-7-5, REV 0, Westinghouse Hanford Company, Richland, Washington.
Recovery = 60% (6 membrane passes)
ACV = administrative control value

Table 5. Analysis of Water from Well 200-W19-03. (All units are in parts per billion unless indicated otherwise.)

Constituent	Concentration
Barium	49
Calcium	42,900
Chloride	19,100
Copper	12
Fluoride	965
Iron	245
Magnesium	14,100
Manganese	9
Nitrate	93,000
Potassium	4,630
Sodium	77,100
Strontium	207
Sulfate	55,400
Zinc	15
Uranium	8,590
Total Organic Carbon	643
Total Organic Halide	69
Total Alpha	3,930 pCi/L
Total Beta	4,050 pCi/L
^{137}Cs	5 pCi/L
^{60}Co	3 pCi/L
^{99}Tc	1,800 pCi/L

Table 6. Reverse Osmosis Treatment of Hanford Groundwater Well 200-W19-03.

Constituent	Feed (pCi/L)	Permeate (pCi/L)	Rejection (%)	DWS[a] (pCi/L)
Uranium	4,700	4.7	99.9	600
Technetium-99	1,219	57.3	95.3	900

[a] EPA, 1976, *National Interim Primary Drinking Water Regulations for Radiological Constituents*, EPA-570/9-76-003, U.S. Environmental Protection Agency, Washington, D.C.
Recovery = 89% (9 membrane passes)
DWS = Drinking Water Standard

Table 6a. Reverse Osmosis Treatment of Hanford Groundwater (HCl added to dissolve $CaCO_3$).

Constituent	Permeate (pCi/L)	Rejection (%)	DWS[a] (pCi/L)
Uranium	7.8 ppb	99.8	600
Technetium-99	192.7	83.9	900

[a] EPA, 1976, *National Interim Primary Drinking Water Regulations for Radiological Constituents*, EPA-570/9-76-003, U.S. Environmental Protection Agency, Washington, D.C.
Recovery = 96% (14 membrane passes)
DWS = Drinking Water Standard

Table 7. Constituents Above Detection Limits in Purge Water from Well 1-H3-2B. (All units are parts per billion except as indicated.)

Constituent	Concentration
Boron	20
Calcium	26,500
Chloride	2,800
Chromium	198
Magnesium	9,710
Manganese	7
Nitrate	5,700
Potassium	5,120
Silicon	8,250
Sodium	4,430
Strontium	196
Sulfate	23,500
Zinc	678
Alpha	1.94 pCi/L
Beta	5.91 pCi/L
Uranium	2.58 pCi/L

Table 8. Analytical Summary of Reverse Osmosis Testing of Purge Water from Well 1-H3-2B.

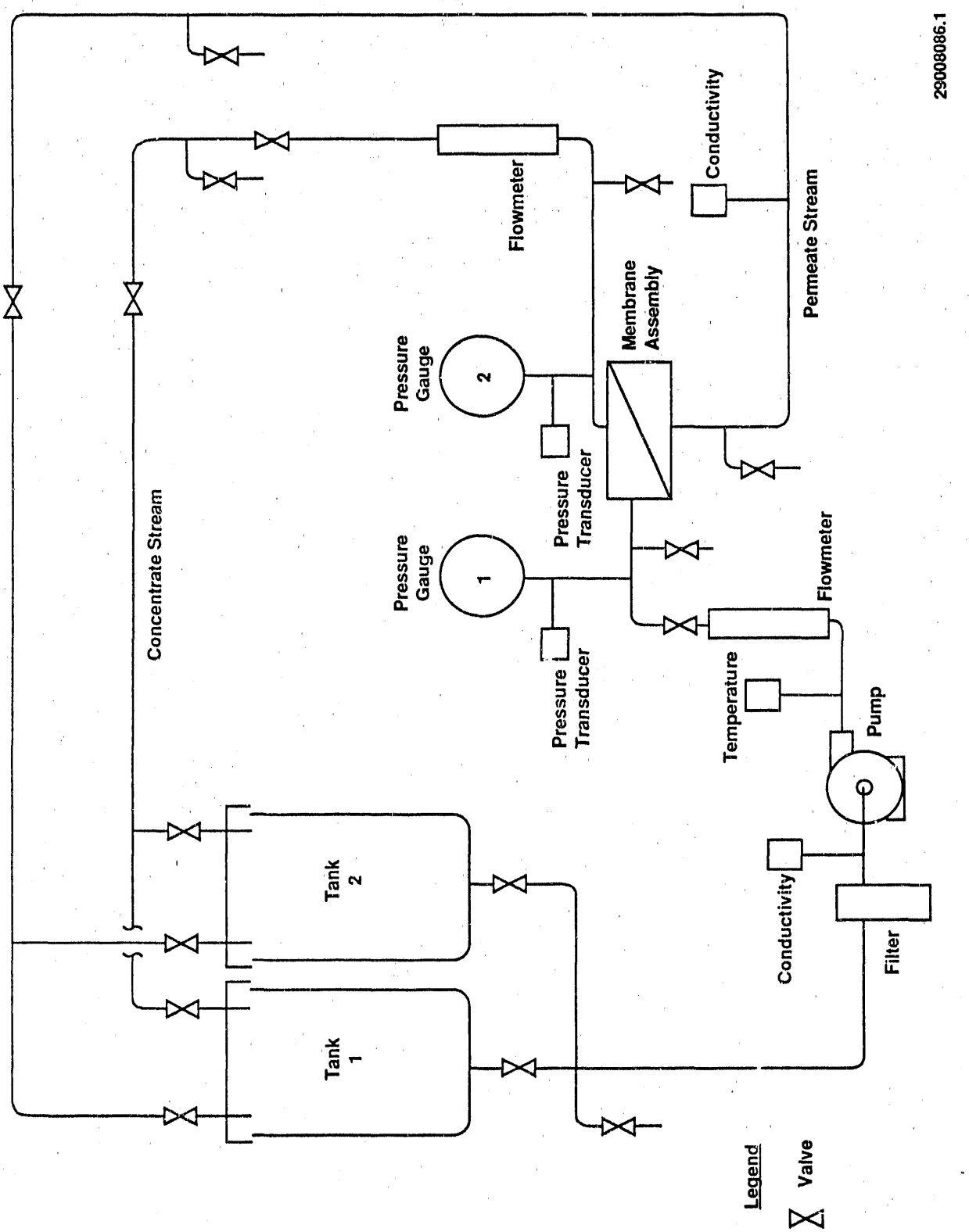
Nuclide	Feed (pCi/L)	Permeate (pCi/L)	Concentrate (pCi/L)	Rejection (%)	DWS[b] (pCi/L)
Alpha	1.94	0.25	457	87.6	15
Beta	5.91	0.70[a]	92	88.2	50
Uranium	2.58	0.45	812	82.5	600

[a] Denotes a result less than the detection limit.

[b] EPA, 1976, *National Interim Primary Drinking Water Regulations for Radiological Constituents*, EPA-570/9-76-003, U.S. Environmental Protection Agency, Washington, D.C.

DWS = Drinking Water Standard

Figure 1. Reverse Osmosis Unit--Flow Schematic.



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