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FILTRATION OF OAK RIDGE NATIONAL LABORATORY SIMULATED LIQUID LOW-LEVEL WASTE

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ABSTRACT

A method for disposal of Oak Ridge National Laboratory's (ORNL's) liquid low-level radioactive waste (LLLW) is being developed in which the material will be solidified in cement and stored in an aboveground engineered storage facility. The acceptability of the final waste form rests in part on the presence or absence of transuranic isotopes.

Filtration methods to remove transuranic isotopes from the bulk liquid stored in the Melton Valley Storage Tanks (MVST) were investigated in this study. Initial batch studies using waste from MVST indicate that >99.9% of the transuranic isotopes can be removed from the bulk liquid by simple filtration. Bench-scale studies with a nonradioactive surrogate waste indicate that >99.5% of the suspended solids can be removed from the bulk liquid via inertial crossflow filtration.

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1. INTRODUCTION

The evolving Underground Injection Control Regulations (Chap. 1200-4-6) of the Rules of the Water Quality Board for the State of Tennessee (first issued May 22, 1985) have led to discontinuation of the hydrofracture process, which was used at ORNL for solidification disposal of LLLW from the mid-1960s until late 1984. Since hydrofracture is no longer authorized, an alternative means for disposal of LLLW concentrate must be implemented. Currently, capacity is limited in the eight 190-m^3 MVST that store the LLLW, so a method for solidifying the waste in cement and disposing of it is being developed to prevent shutdown of the LLLW system at ORNL.

The eight tanks, identified as W-24 through W-31, contain varying quantities of LLLW and precipitated sludge. The supernate contains various beta- and gamma-emitting radionuclides, primarily ^{90}Sr and

^{137}Cs. The transuranic (TRU) content of the liquid is generally less than 100 nCi/mL. The solids contain the same radionuclides, but the TRU content (by gross alpha analyses) ranges up to ~6000 nCi/g.[1] The sodium concentration of the supernate is ~4 M, and other cations are present, such as Ca, Mg, K, Al, Fe, and so forth, in concentrations up to ~0.25 M. Nitrates are in quantities of 3 moles/L. The pH of the supernate ranges from ~12 to ~14. The proposed flow sheet for processing this waste consists of inertial crossflow filtration of the bulk liquid for TRU isotope rejection, treatment of the filtrate via ion exchange for sorption of ^{137}Cs and ^{90}Sr, and solidification of the ion-exchange column effluent in cement. The resultant waste forms will be stored in an aboveground engineered storage facility. The acceptability of the final waste form rests in part on the presence or absence of transuranic isotopes, which data indicate are present as precipitates.[1]

Separation methods are being studied which will remove radionuclides from the bulk liquid. This will ultimately produce a lower-activity, non-TRU waste [i.e., as defined by the Department of Energy (DOE), non-TRU waste is radioactive waste that contains <100 nCi/mL of alpha emitters that have a half-life of >20 years] for final disposal. These separation studies are being performed in two phases: (1) filtration of simulated wastes and (2) filtration of actual MVST wastes. Initial scouting tests were performed with nonradioactive (cold) simulated wastes to select appropriate filters for consideration in tests using MVST waste. This report describes the first phase of these studies.

2. BATCH FILTRATION OF MVST WASTE

Batch filtration of MVST waste was accomplished via Millipore filters with 2.7- and 0.45-μm nominal porosity. Results from one test utilizing a 100-mL aliquot of a sample from W-25 obtained in July 1985[1] and containing ~34 vol % settled solids filtered through the 2.7-μm filter are summarized in Table 1.

Table 1. Filtrate and filter cake assays of Melton Valley Storage Tanks waste

Parameter	Filtrate (nCi/mL)	Filter cake (nCi/g)
Gross alpha	<0.05[a]	2.64 E+3
^{238}Pu	0.019	6.90 E+2
^{239}Pu, ^{240}Pu	N.D.[b]	2.40 E+2
^{233}U	0.014	7.30 E+1
^{241}Am	N.D.	8.00 E+0
^{244}Cm	N.D.	1.62 E+3
Gross gamma[c]	1.81 E+6	2.09 E+7
Gross beta	4.89 E+3	6.51 E+5
^{134}Cs	2.10 E+1	N.D.
^{137}Cs	4.24 E+3	1.20 E+4
^{60}Co	8.00 E+0	6.57 E+3
^{95}Zr	N.D.	2.70 E+2
^{152}Eu	N.D.	9.81 E+3
^{154}Eu	N.D.	6.16 E+3
^{155}Eu	N.D.	2.95 E+3

[a]0.05 nCi/mL is detection limit for gross alpha.

[b]N.D. denotes isotope not detected.

[c]Gross gamma is in counts per min per mL or g.

These data clearly indicate that the TRU isotopes exist primarily as precipitates and can be removed by filtration. Filtration tests with the 0.45-μm filter also produced filtrate containing levels of gross alpha at the detection limit.

3. PREPARATION OF SIMULATED MVST WASTE

A simulated waste was prepared which approximates the contents of the MVST.[1] The base mix was prepared by dissolving various salts in tap water and then brought to a pH of 11.5 using NaOH. This caused the precipitation of most of the metals as hydroxides or carbonates. The resultant slurry, ~3 wt % solids, was then heated to boiling and held at that temperature for 1 h. This step simulates the heating cycle to which the waste is subjected in the LLLW evaporator. This mixture was initially formulated to simulate the worst case expected tank contents for vendor

tests of solidification techniques.[2] The contents of this base mix are summarized in Table 2.

Table 2. Simulated Melton Valley Storage Tanks wastes

Salt	Concentration (moles/L)
Sodium nitrate	3.46 E+00
Calcium nitrate	3.47 E-01
Magnesium chloride	4.09 E-02
Mercury(II) nitrate	2.55 E-04
Cadmium chloride	4.44 E-04
Chromium(III) nitrate	9.57 E-04
Barium nitrate	3.63 E-04
Lead(II) nitrate	1.16 E-04
Silver nitrate	4.63 E-04
Strontium nitrate	5.99 E-04
Cesium chloride	3.77 E-04

Bentonite was added to this base mix to produce a ~10 wt % slurry. This final slurry was used as the feed for all cold tests. A dispersant dye (0.2-μm nominal size) was also added to the feed during the 1000-h test, with the Mott crossflow filter as an indicator of filtrate quality.

4. SCOUTING TESTS WITH SIMULATED WASTE

Cold tests of four filters were conducted using the simulated "worst case" MVST waste described above. Testing began by running a series of short scouting tests to determine the optimum operating conditions for each filter. The crossflow filters were tested in both total recycle and volume-reduction modes. These tests were followed by long-term tests (1000 h) in the total recycle mode to determine the expected service life of each filter. The results of these tests are summarized in the following subsections.

4.1 DEAD-END FILTRATION

A Mott Metallurgical Corp. sintered stainless steel filter disc (70 mm diam) of 0.5-μm nominal porosity was tested to determine the feasibility of this type filter in the MVST solidification system. The filter disc was enclosed within a stainless steel housing. A pressurized and air-agitated feed tank supplied the slurry feed to the filter.

An initial clean-water test was performed with the new filter element for comparison with later data to determine the extent of filter plugging. This clean-water test consisted of measuring the filtration time required for 0.5 L of distilled water (upflow) at 170-KPa feed pressure.

The filtration tests using simulated waste involved upflow filtration at a constant pressure of 584 KPa for ~60 min. During this period, the filtrate flow rate was recorded at discrete time intervals of 5 min and averaged to obtain the flux for each run. The filtration cycle was followed by backwashing, for a period of 30 s, with the approximately 40 mL of filtrate which remained in the upper filter housing. Air at 620 KPa was the driving force for backwashing. A clean-water test followed each filtration test. This technique was used for all runs except the No. 5 series, which was run in a semicontinuous mode (i.e., four cycles consisting of 60 min of filtration, with each filtration cycle being followed by a 30-s backwash). The data from these tests are presented in Table 3.

Table 3. Disc filter test results using a simulated Melton Valley Storage Tanks feed

Run No.	Feed TSS[a] (wt %)	Filtrate flux[b] (L/min•m^2)	Clean water flux[c] (L/min•m^2)
0	0		258.3
1	10.0	4.48	179.3
2	13.5	2.57	99.0
3	12.5	2.77	163.8
4	12.4	2.69	178.8
5A	11.3	2.57	
5B	11.3	2.28	
5C	11.3	2.49	
5D	11.3	1.79	115.7
6	12.2	2.12	121.4

[a]TSS - total suspended solids.

[b]Filtrate flux averaged for 60 min.

[c]Flux measured with clean water at 170-KPa driving force.

Filtrate assays performed for all tests indicated a total suspended solids (TSS) rejection of >99.5%. Evaluation of the data indicates that dead-end filtration of this simulated waste with approximately 10 wt % solids can produce a filtrate essentially free of TSS. However, the decline in flux of >50% (with both simulated waste and clean water) indicates severe plugging of the filter. No effort to clean the filter disc, beyond backwashing with clean water, was attempted. Based on this data and the mechanical problems associated with handling the wet filter cake, dead-end filters were eliminated from further testing.

4.2 INERTIAL CROSSFLOW FILTRATION

Inertial crossflow filtration utilizing sintered metal filter tubes has been tested using filter test units obtained from Mott Metallurgical Corp. and CARRE, Inc. The filter tube supplied by Mott was 0.5-μm nominal porosity sintered stainless steel with a filtration area of 93 cm^2 (0.1 ft^2). The filter tube, 46 cm (18 in.) long by 0.64 cm (0.25 in.) I.D., was enclosed in a 0.95-cm (0.375-in.) I.D. housing. Two units supplied by

CARRE, Inc., were tested. Both tubes were 0.5-μm nominal porosity sintered stainless steel with a filtration area of 121 cm^2 (0.13 ft^2). Each of these tubes was 30.5 cm (12 in.) long by 1.27 cm (0.5 in.) I.D. enclosed in a 1.59-cm (0.625-in.) I.D. housing. One filter tube contained a membrane formed from anhydrous zirconium oxide and sodium silicate (ZOSS). The other contained a membrane formed from proprietary materials.

A laboratory-scale test loop, shown schematically in Fig. 1, was constructed to test the crossflow filters. A low-shear, air-operated, double-diaphragm pump was used as a slurry feed pump to reduce the possibility of shearing the particulates. All three filters were evaluated in the following manner:

1. Initial clean-water tests were run at various flow rates and filtration pressures to obtain baseline data for the as-received filters.

2. Tests were performed with simulated waste in a total recycle mode (i.e., constant TSS content) to determine optimum parameters for filter operation.

3. Operation in a volume-reduction mode was performed to simulate actual operating conditions where the feed will be concentrated.

4. Final clean-water tests were performed to determine the extent of filter element fouling during the filtration runs.

4.2.1 Mott Bare-Tube Filter Element

The as-received Mott inertial crossflow filter was tested with demineralized water to obtain baseline data. An initial test was performed using a water flow rate of 6.36 L/min (3.35 m/s linear velocity through the filter tube) and an average differential filtration pressure of 165 KPa. As shown in Table 4, the filtrate flux declined from an initial flux of 74 $L/min \cdot m^2$ to 5.3 $L/min \cdot m^2$ over a 75-min time span.

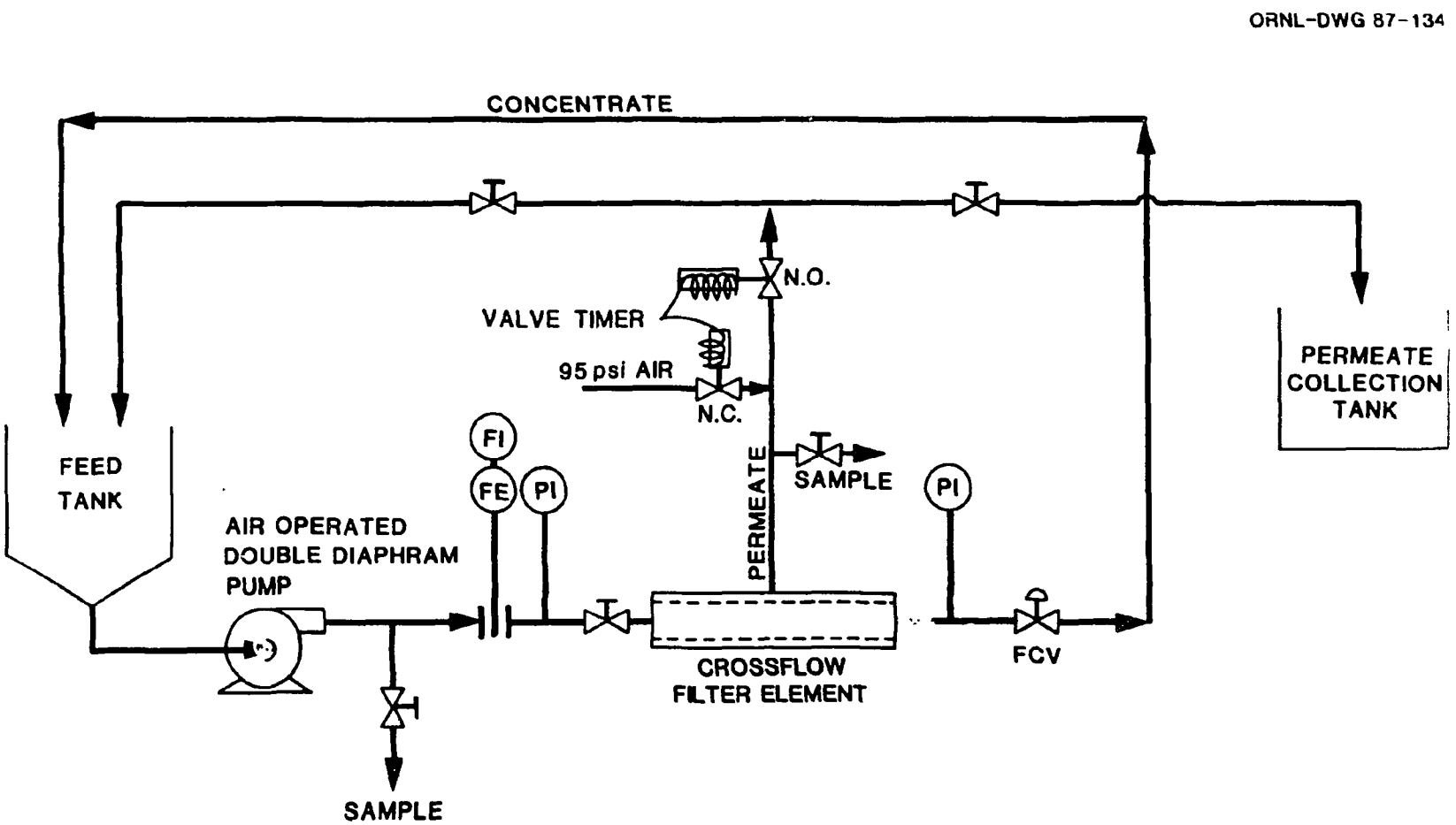

Fig. 1. Crossflow Filtration Cold Test Loop.

Table 4. Mott cross-flow filter, demineralized water test

Time (min)	Filtrate flux ($L/min \cdot m^2$)
0.25	74.1
1.0	53.8
3.0	25.7
5.0	19.6
7.0	17.1
15.0	12.2
30.0	9.0
45.0	6.9
65.0	6.1
75.0	5.3

Based on these data, an additional clean-water test at identical filtration conditions was made, but the filter was backpulsed with 722-KPa water for 0.4 s at intervals of 8 min. During 7.25 h of testing at these conditions, the flux rates were essentially identical (±2%) to those obtained during the first 7 min of operation without backpulsing. This indicates that essentially 100% recovery is obtainable at these backpulse conditions.

The Mott inertial crossflow filter was then evaluated with simulated waste at various feed slurry velocities and filtration pressures in a total recycle mode (i.e., constant TSS content) to determine optimum parameters for filter evaluation in a concentrating mode. The first series of tests was performed to determine the flux decay as a function of time at steady-state conditions with no backpulsing. The resulting filtration rates are presented in Table 5.

Table 5. Mott crossflow filter test using simulated waste without cyclic backpulsing

Slurry[a] velocity (m/s)	Differential filtration pressure (KPa)	Run time (min)	Average filtrate flux ($L/min \cdot m^2$)
1.52	138	57	0.90
3.05	131	211	1.06[b]
5.58	114	67	0.77

[a]Feed slurry solids concentration = 11.2 wt % total suspended solids.

[b]The initial flux, 1.18 $L/min \cdot m^2$, declined to 0.98 $L/min \cdot m^2$ during the first 56 min. No further decline was noted for the remaining 155 min of operation. No measurable flux decay was noted during the tests with slurry velocities of 1.52 and 5.58 m/s.

The data indicate that a drastically reduced flux occurs with filtration of simulated waste when compared to the rates obtained with clean water. This can be attributed to the TSS and the dissolved salt content (increased viscosity) of the simulated waste. A flux increase of 17% was noted when the feed velocity through the tube was increased from 1.5 to 3 m/s. Further increasing the velocity to 5.6 m/s resulted in a 27% decline in flux compared to that obtained at 3 m/s and 14% less than that obtained at 1.5 m/s.

The above tests were repeated, but the filtration pressures were varied to evaluate this variable with respect to filtrate flux (Table 6). Cyclic backpulsing was also employed.

Table 6. Mott crossflow filter tests using simulated waste at varying filtration pressures with cyclic backpulsing

[Feed slurry solids concentration = 11.2 wt % total suspended solids. Backpulse conditions: pressure = 756 KPa, interval = 10 min, duration = 0.4 s (~40 mL displacement liquid)]

Slurry velocity (m/s)	Differential filtration pressure (KPa)	Total run time (min)	Filtrate flux[a] (L/min•m^2)
1.52	138	120	1.55
1.52	276	67	2.49
3.05	138	38	1.14
3.05	276	48	1.87
5.36	138	78	0.86

[a]Filtrate flux averaged over the duration of the run.

These data indicate that the use of cyclic backpulsing results in an average flux increase of 73% at a differential filter pressure of 138 KPa and a linear feed velocity of 1.52 m/s. Increasing the filtration pressure by a factor of 2 increases the flux by 61% at 1.52 m/s. The data also indicate that an increase in feed velocity results in a decreased filtrate rate. Similar results were obtained with a feed velocity of 3.05 m/s. With a feed velocity of 5.36 m/s and a filtration pressure of 138 KPa, the flux obtained was 0.86 L/min•m^2, 57% less than was obtained at 1.52 m/s. Of the conditions tested, a feed linear velocity of 1.52 m/s, with cyclic backpulsing, appears to be optimum.

Following these tests, the system was operated in a volume-reduction mode to simulate actual operating conditions. Tests were made at a filtration pressure of 138 KPa and feed velocities of 1.52 and 3.05 m/s. Cyclic backpulsing was employed at 10-min intervals for a duration of 0.4 s at 756 KPa. The initial feed TSS content was ~12.5 wt %. The runs resulted in a 74 and 67% reduction in volume, respectively. The runs were terminated after ~24 h of operation because of liquid holdup in the system. The filtrate flux obtained during these tests is shown in Fig. 2.

Clean-water tests were performed on the Mott filter following the slurry filtration tests. The data indicate a loss of ~12.0% in filtrate

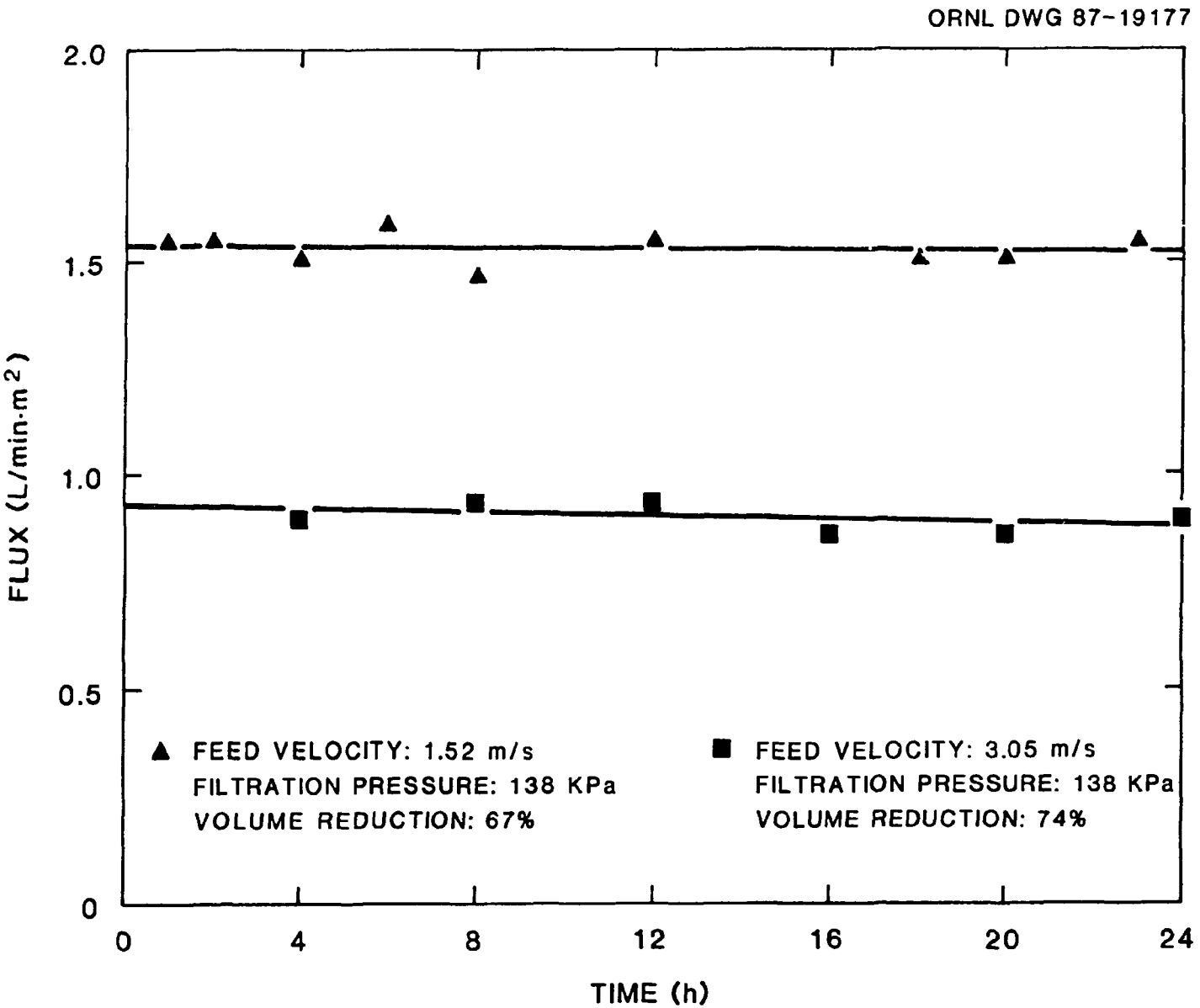

Fig. 2. Mott Crossflow Filter: Filtrate Flux as a Function of Feed Velocity.

flux. Beyond backflushing with distilled water, no attempts were made to clean the filter tube.

The results of these tests indicate that the Mott bare-tube filter element should be considered for further testing with actual MVST waste. Tests should be conducted at a filtration pressure of ~140 KPa to minimize filter blinding at higher pressures. Optimum feed velocity through the tube appears to be ~1.5 m/s.

4.2.2 CARRE, Inc., Membrane Filter Elements

The CARRE, Inc., inertial crossflow filter elements, as-received, were tested with filtered process water without backpulsing. The results are summarized in Table 7.

Table 7. CARRE, Inc., filter tests using process water

Slurry[a] velocity (m/s)	Differential filtration pressure (KPa)	Filtrate flux (L/min•m^2)	
		ZOSS[b] element	Proprietary element
1.52	138	0.41	0.29
1.52	207	0.73	0.49
4.57	138	0.45	0.33
4.57	207	0.49	0.41

[a]Run time for each test was 80 – 100 min.

[b]ZOSS = zirconium oxide and sodium silicate.

The filtrate flux was constant (i.e., no flux decay) at all operating conditions without backpulsing. This is in contrast to the data obtained during evaluation of the Mott filter element, which required cyclic backpulsing to prevent fouling of the element.

Following the clean-water test, three runs were made using simulated MVST waste with each of the CARRE, Inc., units (Table 8). These runs were conducted in a total recycle mode without backpulsing, and the filtrate rates were constant for the duration of each test.

Table 8. CARRE, Inc., filter tests using simulated waste without backpulsing

(Feed slurry - 10.0 wt % total suspended solids)

Slurry velocity (m/s)	Differential filtration pressure (KPa)	Run time (min)	Filtrate flux ($L/min \cdot m^2$)
	Zirconium oxide and sodium silicate membrane		
1.5	140	240	0.24
4.6	140	115	0.24
4.0	200	245	0.33
	Proprietary membrane		
1.5	140	180	0.16
4.6	140	120	0.29
4.0	200	180	0.41

The data in Table 8 indicate that a filtration pressure of 200 KPa and 4.0 m/s linear feed velocity produce filtrate rates ~40% higher than the other conditions tested; therefore, these conditions were selected for volume reduction studies. The filtrate was analyzed for TSS content during these tests (Tables 9 and 10).

Table 9. Volume reduction of Melton Valley Storage Tanks simulated waste using the zirconium oxide and sodium silicate membrane filter

[Slurry feed velocity: 4.0 m/s; filtration pressure: 200 KPa; initial slurry concentration: 9.67 wt % total suspended solids (TSS) (96,700 mg/L); total volume reduction: 64.6%]

Elapsed time (h)	Average filtrate flux ($L/min \cdot m^2$)	Filtrate TSS (mg/L)
3.50	0.24	9.0
7.50	0.29	4.0
11.75	0.24	0.0
15.50	0.29	3.0
17.53	0.24	20.5

Table 10. Volume reduction of Melton Valley Storage Tanks simulated waste using the proprietary membrane filter

[Slurry feed velocity: 4.0 m/s; filtration pressure: 200 KPa; initial slurry concentration: 10.4 wt% total suspended solids (TSS) (104,000 mg/L); total volume reduction: 46.8%]

Elapsed time (h)	Average filtrate flux ($L/min \cdot m^2$)	Filtrate TSS (mg/L)
2.0	0.41	4.0
4.0	0.41	6.0
6.0	0.37	2.0
8.0	0.37	6.5
9.0	0.37	80.5

Tables 9 and 10 show that the CARRE, Inc., proprietary membrane produces a filtrate flux about 46% higher than the CARRE ZOSS membrane and about 50% less than the Mott crossflow filter at identical operating conditions. Filtrate assays for TSS indicated >99.9% solids rejection.

Clean-water tests on both of the CARRE units followed the slurry filtration tests. Initial and final water testing of the ZOSS membrane element produced identical filtrate rates, indicating no loss of filtration efficiency. Final clean-water tests, with the proprietary membrane filter, produced filtrate rates ranging from 75 to 112% greater than the filtrate rates obtained with the as-received filter. This increase in flux and the high TSS content in the final filtrate sample from the volume-reduction run indicate that the proprietary membrane had become depleted.[3] The pH, TSS, or salt content of the feed may have caused the deterioration. Based on the above data, the proprietary membrane is considered unsuitable for use in the MVST system and is thus eliminated from further testing.

5. LONG-TERM CROSSFLOW FILTER TESTS WITH SIMULATED WASTE

Extended runs of 1000-h duration were made with the CARRE, Inc., ZOSS membrane filter and the Mott bare-tube filter to determine the service life of the elements. Operating conditions were selected based on the data presented in Sect. 4.

5.1 MOTT BARE-TUBE FILTER ELEMENT

A new 0.5-μm nominal porosity filter tube was placed in the filter housing and clean-water tested prior to an extended test of ~1000 h. The feed slurry contained 8.4 wt % TSS. When added to the feed, a dispersant dye (50.0 mg/L) with an average particle diameter of ~0.2 μm permitted more accurate measurement of filtrate quality than the previously used TSS assays. Operating parameters were as follows:

Slurry feed rate: 2.8 L/min (1.52 m/s linear velocity);
Average filtration pressure: 138 KPa;
Backpulse conditions
- pressure: 722 KPa;
- interval: 15 min;
- duration: 0.4 s.

Filtrate flux rates and solids rejection data collected during this test are presented in Fig. 3. The system was operated for 429 h at a pH of 11.8. The flux rates declined from an initial value of 1.53 L/min•m^2 to 0.59 L/min•m^2 during the first 380 h of operation and remained essentially constant for the next 50 h. Filtrate analyses by spectrophotometry indicated 100% rejection of the added dye over this 430-h time span. After 430 h of operation, the pH of the feed was increased to >14 (1.1 M hydroxide concentration). Immediately after the pH adjustment, the flux increased to 1.64 L/min•m^2 and the rejection declined to 83%. During the remainder of the test, the flux decreased to 0.37 L/min•m^2, and the filtrate quality increased to 94% rejection of the dye. Before ending the

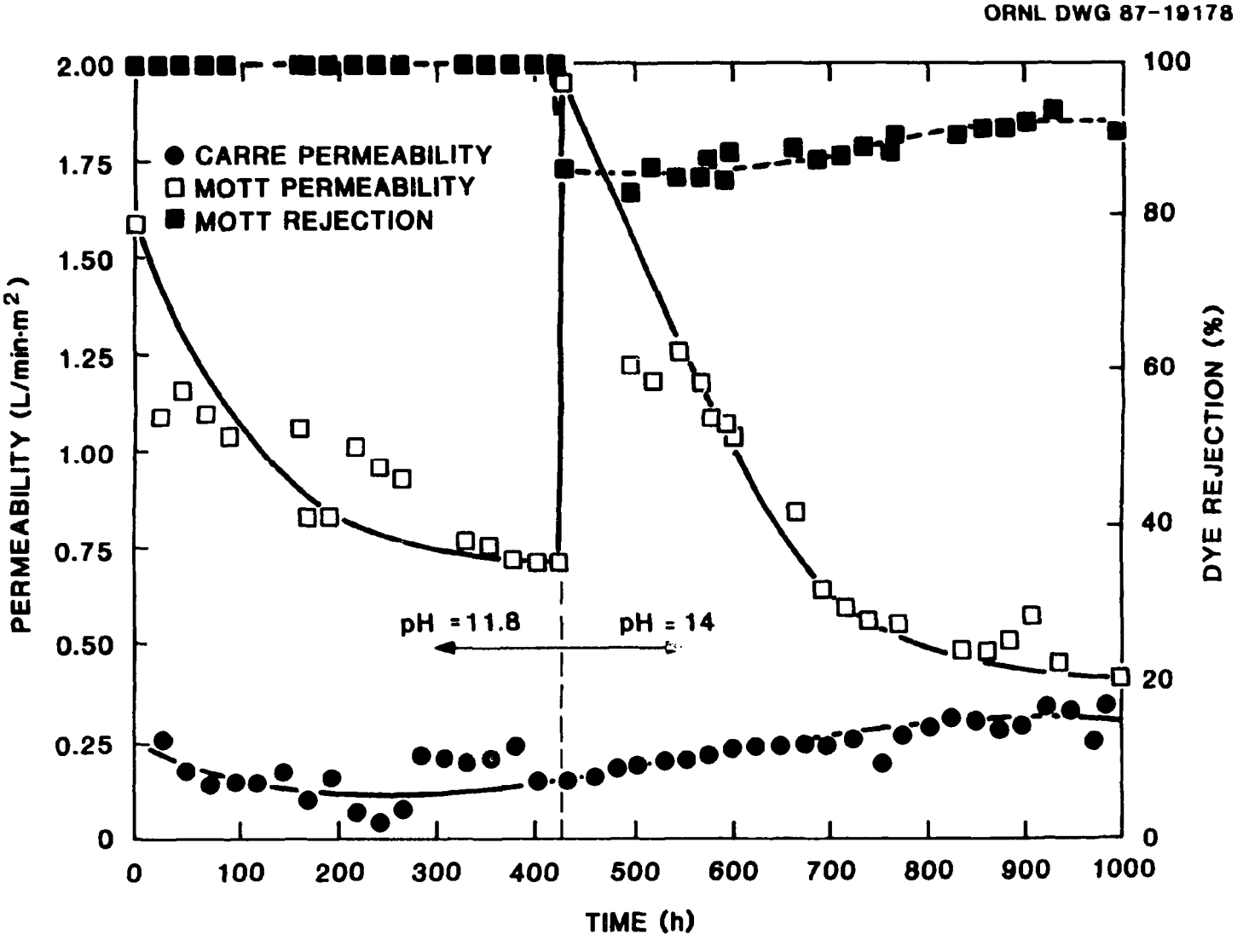

Fig. 3. Extended Cold Test of Carre Zoss and Mott Filters.

test, the backpulse interval was decreased to 5 min, with no increase in filtrate rates noted.

A clean-water test was performed following this test using clean process water at the normal pH of 7.6. The flux had decreased by 99.4% from the as-received filter element (from 144 L/min•m^2 to 0.91 L/min•m^2). Filtrate analyses during the clean-water test indicated 100% dye rejection. Backwashing the filter with water at a pressure differential of 722 KPa for ~1 h did not improve the filtrate rate, which indicates extreme blinding of the filter.

The results of this test indicate that a pH of about 12 or less is necessary for optimum permeability and filtrate quality. At the lower pH, the bentonite apparently forms a very tight membrane, but at a pH of 14 it becomes more porous, permitting ≥10% of the dispersed dye to bleed through. This phenomenon may or may not exist with the actual MVST waste. Analyses of the filtration data obtained with a bare-tube filter element indicate that the need for a membrane filter element is questionable.

5.2 CARRE, INC., ZOSS MEMBRANE FILTER ELEMENT

An extended run of 1000-h duration was made with the ZOSS membrane filter element using a slurry feed containing 9.7 wt % TSS. No backpulsing of the element was employed. Operating parameters were as follows:

Slurry feed rate: 34 L/min (~4.6 m/s linear velocity);
Average filtration pressure: 138 KPa.

The system was operated for ~450 h at a feed pH of 11.8. The pH was then adjusted to >14 (1.45 M hydroxide concentration) and operations continued for an additional 550 h. The filtrate flux data is shown in Fig. 3. The flux decreased from 0.28 L/min•m^2 to 0.03 L/min•m^2 after 250 h. The flux then increased to 0.17 L/min•m^2 at 450 h. The flux rate steadily increased to 0.28 L/min•m^2 (the initial rate for this test) over the final 550 h of operation. The TSS data taken during the first 650 h were not consistent; assays varied from 0 to 568 mg/L. Because of this

inconsistency, filtrate analysis for TSS was not performed after the 650-h sample. The 568 mg/L TSS assay indicated a solids rejection of >99.4% which may be adequate for producing a filtrate of less than 100 nCi/mL of TRU isotopes.

A clean-water test using process water (pH 7.6) was performed after this run using the same feed rates and filtration pressures as in the preceding text. The average filtrate rate for a 60-min period was 0.045 $L/min \cdot m^2$, a factor of 10 less than rates attained with the as-received element.

The filter was then backwashed with process water at 722 KPa for 25 min. The clean-water test was repeated using water spiked with the dispersant dye (50 mg/L, 0.7-μm average particle diameter) in an effort to determine filtrate quality. The filtrate rate for a 2.6-h test period averaged 0.016 $L/min \cdot m^2$. Filtrate analyses by spectrophotometry indicated 100% rejection of the dye.

The pH of the water was adjusted to 13.9 with NaOH and the clean-water test continued. Results of this test are shown in Table 11.

Table 11. Clean-water test with the CARRE, Inc., zirconium oxide and sodium silicate membrane filter element

Time (h)	Filtrate flux ($L/min \cdot m^2$)	Filtrate quality (dye concentration, mg/L)
0.3	0.032	0
0.7	0.035	0
1.8	0.041	4.17
2.3	0.044	6.85
18.5	0.102	8.72
21.0	0.127	9.25
23.8	0.139	9.25
42.6	0.191	9.79
47.2	0.200	9.52

The flux increased by a factor of >6 over the 47-h test period. The flux at the end of this test was 55% less than the as-received unit. Filtrate assays indicated an 80% rejection of the dispersed dye. These tests indicated that at some time during the 1000-h test, the ZOSS membrane was destroyed. This probably occurred when the feed pH was changed from 11.8 to 14. It is hypothesized that a membrane consisting of bentonite

particles was deposited on the tube which essentially replaced the ZOSS membrane. This bentonite layer allowed the filter to operate with a reasonably high TSS rejection and a low flux.

A new ZOSS membrane filter element was then clean-water tested for a 2-h period at the natural pH of the water (7.6) and with the same operating parameters as in the above tests. The filter permeability over this period of time was essentially constant at 0.34 $L/min \cdot m^2$. Filtrate assays indicated 100% rejection of the dye.

The following conclusions concerning the performance of the ZOSS membrane have been drawn:[4]

1. The marked difference in water permeability at neutral pH between the as-received filter used in the 1000-h test, before and after backwashing, indicates that the original membrane had been compromised. Partial dissolution of the membrane had been anticipated during operation at a pH of 14; and, in fact, it is not surprising that the attack was rather extensive over a period of 550 h.

2. The sharp decline in permeability from 0.28 $L/min \cdot m^2$ at the end of the test, with simulated waste at pH 14, to 0.045 $L/min \cdot m^2$ for process water at a neutral pH is not surprising. Presumably, the gaps left in the ZOSS membrane were filled with some material of low permeability in the process water, possibly colloidal alumina. This material would dissolve rapidly at high pH and result in the higher permeability obtained during the final clean-water test at pH 14.

3. The tests with the dispersed dye are also consistent with the above conclusions. High rejection and good permeability were obtained with an intact ZOSS membrane. High rejection and poor permeability were obtained with a tight membrane of possibly colloidal alumina. Some dye permeates the filter when flux is high with an imperfect membrane.

4. Membrane recoating capabilities will probably be necessary for use of the ZOSS membrane when using feed at a pH of 14.

6. SUMMARY AND RECOMMENDATIONS

The CARRE, Inc., proprietary membrane filter became depleted within 20 h of operation probably because of pH or salt content of the feed. The proprietary membrane filter is therefore not recommended for use in the proposed flow sheet, and no further evaluation of this element occurred.

Evaluation of the Mott bare-tube and the CARRE, Inc., ZOSS membrane filter elements at identical filtration pressures (138 KPa) indicates that the permeate quality from each approaches 100% solids rejection with a feed pH of ~12. At a pH of 14 or greater, the filter elements produced filtrate of equal quality, each demonstrating 80 to 90% solids rejection.

After 1000 h of operation with simulated feed, the permeability of the Mott filter had decreased to 0.37 $L/min \cdot m^2$. This is significantly greater than the permeability of the Carre ZOSS filter, which was in the 0.03 to 0.28 $L/min \cdot m^2$ range during the entire 1000-h test.

Using simulated MVST waste and at the operating conditions for these studies, the data indicate that

1. the Mott bare-tube filter and the CARRE, Inc., ZOSS membrane filter produce filtrate of equal quality;

2. feed pH affects filtrate quality;

3. during the 1000-h tests, the average filtrate flux for the Mott bare-tube filter was 3.9 times greater than that produced by the CARRE, Inc., ZOSS membrane filter;

4. the ZOSS membrane in the CARRE, Inc., filter tube became exhausted at some point during the 1000-h test; and

5. optimum feed rates (linear velocities through the filter tubes) appear to be 1.5 m/s for the Mott and 4.6 m/s for the CARRE, Inc., elements.

These cold tests indicate that both filters have the potential for rejection of the TRU isotopes contained in the MVST waste. The data to date indicate that the Mott filter has a higher flux capacity and a better chance of withstanding the harsh environment of the LLLW. Therefore, it is the recommended filter for use in the MVST reference flow sheet. However, tests with actual waste need to be performed to determine if the simulated wastes studies accurately predict the characteristics of the sludge and to determine if TRU material can be removed by crossflow filtration. It is recommended that both filter tubes be tested with actual MVST waste solutions. Parameters of interest should be (1) filtration pressure, (2) feed linear velocity through the filter tube, and (3) pH.

7. REFERENCES

1. F. J. Peretz et al., Characterization of Low-Level Liquid Waste at the Oak Ridge National Laboratory, ORNL/TM-10215 (December 1986).

2. A. J. Mattus, ORNL, personal communication, May 1986.

3. J. S. Johnson, Jr., CARRE, Inc., consultant, personal communication, December 1986.

4. J. S. Johnson, Jr., CARRE, Inc., consultant, personal communication, February 20, 1987.