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Cross Flow Filtration of Oak Ridge National Laboratory Liquid Low-Level Waste

V. L. Fowler
J. D. Hewitt

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Chemical Technology Division
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Liquid Low-Level Waste Solidification Program
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CROSS FLOW FILTRATION OF OAK RIDGE NATIONAL LABORATORY LIQUID LOW-LEVEL WASTE

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Engineering Development Section

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CROSS FLOW FILTRATION OF OAK RIDGE NATIONAL LABORATORY LIQUID LOW-LEVEL WASTE

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ABSTRACT

A new method for disposal of Oak Ridge National Laboratory liquid low-level radioactive waste is being developed as an alternative to hydrofracture. The acceptability of the final waste form rests in part on the presence or absence of transuranic (TRU) isotopes.

Inertial cross flow filtration was used in this study to determine the potential of this method for separation of the TRU isotopes from the bulk liquid stored in the Melton Valley Storage Tanks. These studies indicate that the filtrate produced by cross flow filtration, using filter elements of 0.5- and 2.0-μm nominal porosity, contains less than 1 nCi/mL of TRU isotopes.

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 the discontinuation of the hydrofracture process, which was used at Oak Ridge National Laboratory (ORNL) to dispose of liquid low-level radioactive waste (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, the eight 190-m^3 Melton Valley Storage Tanks (MVST) that store the LLLW have limited capacity. A method for solidifying and disposing of LLLW 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. Data exist indicating that the TRU contaminants are present as precipitates.[1,2] The sodium concentration of the supernate is ~4 M. Other cations are present, such as Ca, Mg, K, Al, and Fe, in concentrations up to ~0.25 M. Nitrates are present in quantities of about 3 mol/L. The pH of the supernate ranges from ~12 to ~14.

One proposed flow sheet for processing this waste includes inertial cross flow filtration of the bulk liquid for TRU isotope rejection. Since the TRU contaminants are probably present as particulates, this separation method to remove the radionuclides from the bulk liquid was studied. This should ultimately produce a lower-activity, non-TRU waste for final disposal. (As defined by the Department of Energy, non-TRU waste is radioactive waste that contains <100 nCi/mL of alpha emitters that have a half-life of >20 years.)

These separation studies were performed in two phases: (a) filtration of simulated wastes and (b) 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.[2] Based on these initial studies, inertial cross flow filtration was the filter of choice for studies with MVST waste. This report describes the second phase of these studies.

2. BACKGROUND: OPERATING PRINCIPLES OF INERTIAL CROSS FLOW FILTRATION

Inertial cross flow filtration is a nonclogging cross flow type of filtration designed for particulate or sludge concentration and liquid recovery. This contrasts with dead-end filtration, in which the process stream passes perpendicular to the plane of the filter medium and the particles accumulate on the filter medium surface. Two types of cross flow filters were used in this study: bare-tube and membrane. Both types were formed from sintered stainless steel. The membrane filter was pretreated by the manufacturer to form a subsurface layer of fine particulates, and the bare-tube filters were not pretreated. During operation with a bare-tube filter element, fine particulates penetrate a short distance into the porous media, creating a subsurface membrane. As a result, this membrane becomes a finer filter than the porous media itself and rejects or prevents fines from passing through the porous wall.

At the beginning of operation with a clean bare-tube filter element, the filtrate rate will be relatively high. It will begin to decline with time as the subsurface membrane develops within the porous tube structure, and this decline will continue until a steady state flow is achieved. This steady state filtrate flow will be a small percentage of the total flow capacity of the porous tube. Therefore, a periodic back flush is used to dislodge the membrane that develops and increase the filtrate flow accordingly.

Operation with a filter element that contains a preformed membrane does not exhibit this decline in filtrate rate. Thus, steady state flow is achieved at startup and back flushing is unnecessary.

The feed stream to be concentrated is pumped through the inside diameter of the filter tube at linear velocities in the range of 1.5 to 6.0 m/s. A high-velocity inertial effect causes particulate depletion at the tube wall because of a "pinch effect." The kinetic energy of the fluid is sufficient to cause the particles to pass straight through the tube because of their inability to turn into the porous wall. This prevents gross amounts of the particulate matter from penetrating the porous media. A valve or restriction placed on the downstream side of the filter tube creates a differential pressure across the porous filter tube. Because of this pressure differential, a portion of the liquid phase of the feed stream flows through the porous wall as clear filtrate, free of particulates, and exits from the filtrate outlet. The filtrate rate is directly proportional to the filtration pressure and inversely proportional to the viscosity of the filtrate.

Filters are evaluated in terms of flux or permeability. Flux is defined as the amount of filtrate collected in a given time divided by the surface area of the filter tube. Permeability is defined as the flux through the filter tube wall divided by the pressure drop across the filter tube wall. All data and discussions presented in this study will be in terms of flux and will have the units $L/min \cdot m^2$.

3. EQUIPMENT DESCRIPTION

A hot cell test loop (HCTL), diagrammed in Fig. 1, was constructed to test filtration and ion exchange decontamination methods. The system consisted of feed and collection tanks, an air-operated, double diaphragm feed pump, connections for two cross flow filter elements, a sand bed

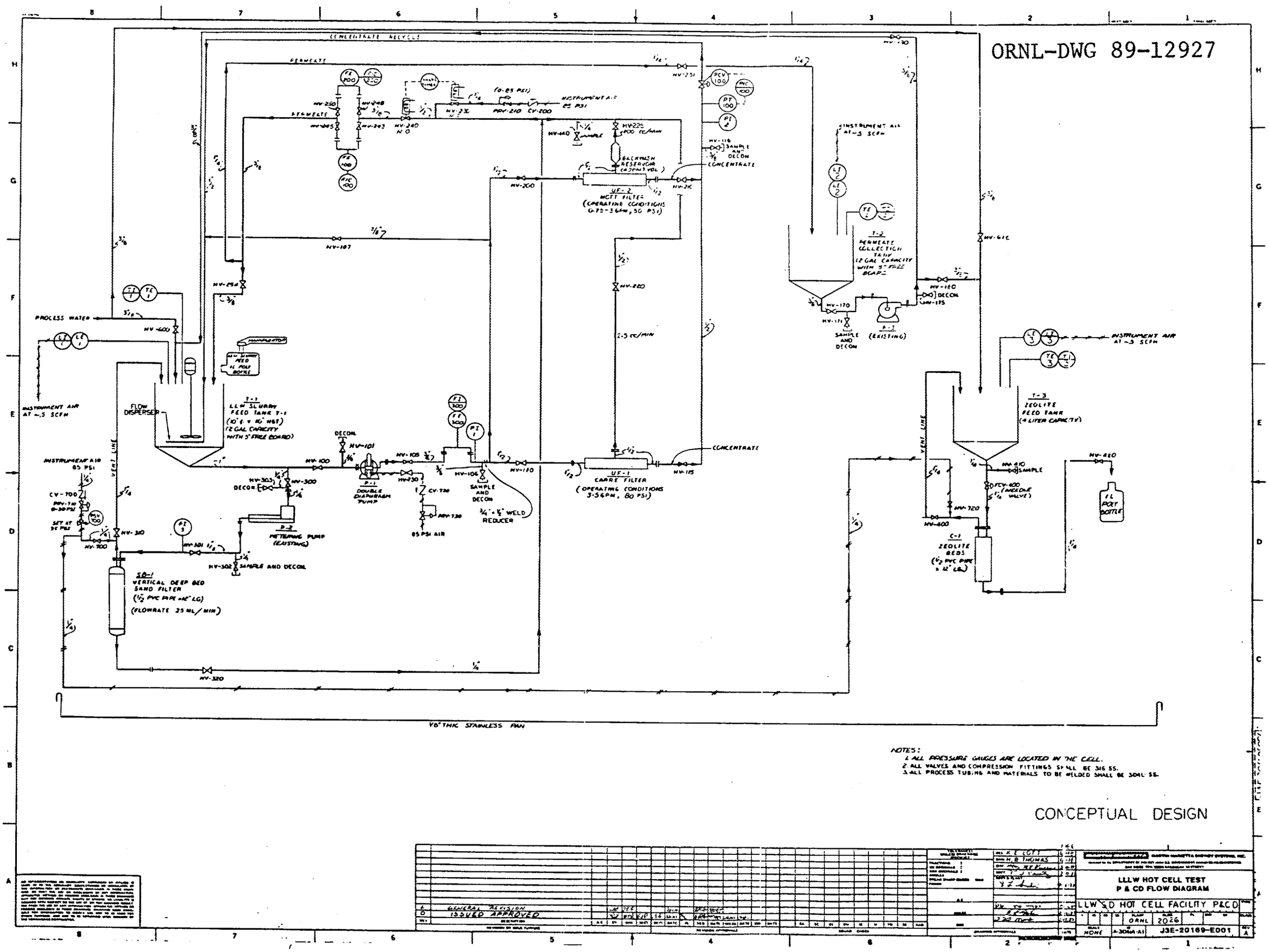

Fig. 1. Hot cell test loop process and instrumentation diagram.

filter, and an ion exchange column. Stainless steel pipe or tubing was used in the construction of the test loop unless otherwise noted.

The filter units tested in this study were manufactured by Mott Metallurgical Corp. and CARRE, Inc. The two filter tubes manufactured by Mott (bare-tube type) were 0.5- and 2.0-μm nominal porosity sintered stainless steel (type 316L), each having a filtration area of 93 cm^2 (0.1 ft^2). The filter tubes, 46 cm (18 in.) long by 0.64 cm (0.25 in.) ID, were enclosed in a 0.95-cm- (0.375-in.-) ID housing.

The one filter tube manufactured by CARRE, Inc., was 0.5-μm nominal porosity sintered stainless steel (type 316) with a filtration area of 121 cm^2 (0.13 ft^2). The filter element was 30.5 cm (12 in.) long by 1.27 cm (0.5 in.) ID, enclosed in a 1.59-cm- (0.625-in.-) ID housing. This filter tube contained a membrane formed from anhydrous zirconium oxide and sodium silicate (ZOSS).

In addition to the cross flow filters, a vertical deep-bed sand filter (SB-1) was incorporated into the system. This filter was fabricated from a 30.5-cm length of clear 0.5-in. polyvinyl chloride (PVC) pipe, schedule 40. The sand bed filter was installed so that the system would have an alternative method of TRU filtration if the cross flow filters failed to adequately reject these isotopes. The HCTL also contained an ion exchange column (C-1) identical in dimensions to SB-1. The ion exchange column was installed for future studies directed at decontamination of the filtrate (i.e., removal of ^{137}Cs and ^{90}Sr). Neither of these two units was utilized in these studies, so they will not be further discussed.

A low-shear, air-operated, double diaphragm pump (P-1), equipped with teflon diaphragms, was used as a slurry feed pump to reduce the possibility of shearing the particulates. Feed from an agitated feed tank (T-1) was pumped to either of the two cross flow filters, and the concentrate was returned to T-1. The feed line was equipped with a flowmeter to monitor feed flow rates. The concentrate line was equipped with an automatic pressure control valve (PCV-100) that controlled the filtration pressure. During these studies, filtrate from the units was returned to T-1 when operating in a total recycle mode to maintain a constant total suspended solids (TSS) content. The filtrate was collected in T-2 for volume reduction tests. The filtrate piping was equipped with timer-controlled solenoid valves (HV-230 and HV-240) and a backwash reservoir to enable periodic backwashing of the filters. Two flowmeters were installed in the filtrate line to continuously monitor the filtrate flow rate. These flowmeters became unreliable shortly after startup because of trapped air under the floats, so they were not used in this study. Air, as the driving force for liquid backwashing, was introduced into the line during the periodic backwashes. To accurately measure the filtrate flow, it became necessary to collect the filtrate in the filtrate collection tank (T-2) periodically and to determine the filtrate rate by tank level depletion and increase (T-1 and T-2, respectively). After determining the flow rate, the filtrate in T-2 was returned to T-1 via pump P-3. For future decontamination studies, this collected filtrate could be pumped to T-3 as feed for C-1.

Each of the feed and collection tanks was equipped with air bubblers and magnehelic gauges for liquid level determination. Each tank also

contained a thermocouple for liquid temperature measurement. Pressure gauges were placed at various locations throughout the system. Sample valves placed at strategic locations enabled sampling of the various streams, and these valves could also be used as "decontamination valves" (i.e., water entry and exit points through individual system components). With the exception of PCV-100, HV-230, and HV-240, all valves in the system were manually operated ball valves.

After construction and initial checkout, the HCTL was installed in the equipment assembly room (mock-up cell) in Bldg. 2026 for verification that the system was adequate for remote operation with manipulators. After placement in the mock-up cell, the completed test loop, along with the control panel for remote operations, is shown in Fig. 2. After verifying the integrity of the test loop, the system was placed in a Bldg. 2026 hot cell, and services were connected.

4. EXPERIMENTAL RESULTS

Filtration studies were begun by running a series of short scouting tests to determine the potential of cross flow filtration for gross alpha rejection. These tests were performed in a total recycle mode (i.e., constant TSS content). Operating parameters were based on the results obtained in earlier simulated waste studies.[2] These studies were followed by a long-term test (1000 h) to determine the expected service life of the filter that exhibited the greatest potential for accomplishing the objectives. Baseline data using process water was obtained on each new filter element. Additional clean water tests were periodically performed on each filter to determine the extent of filter element fouling.

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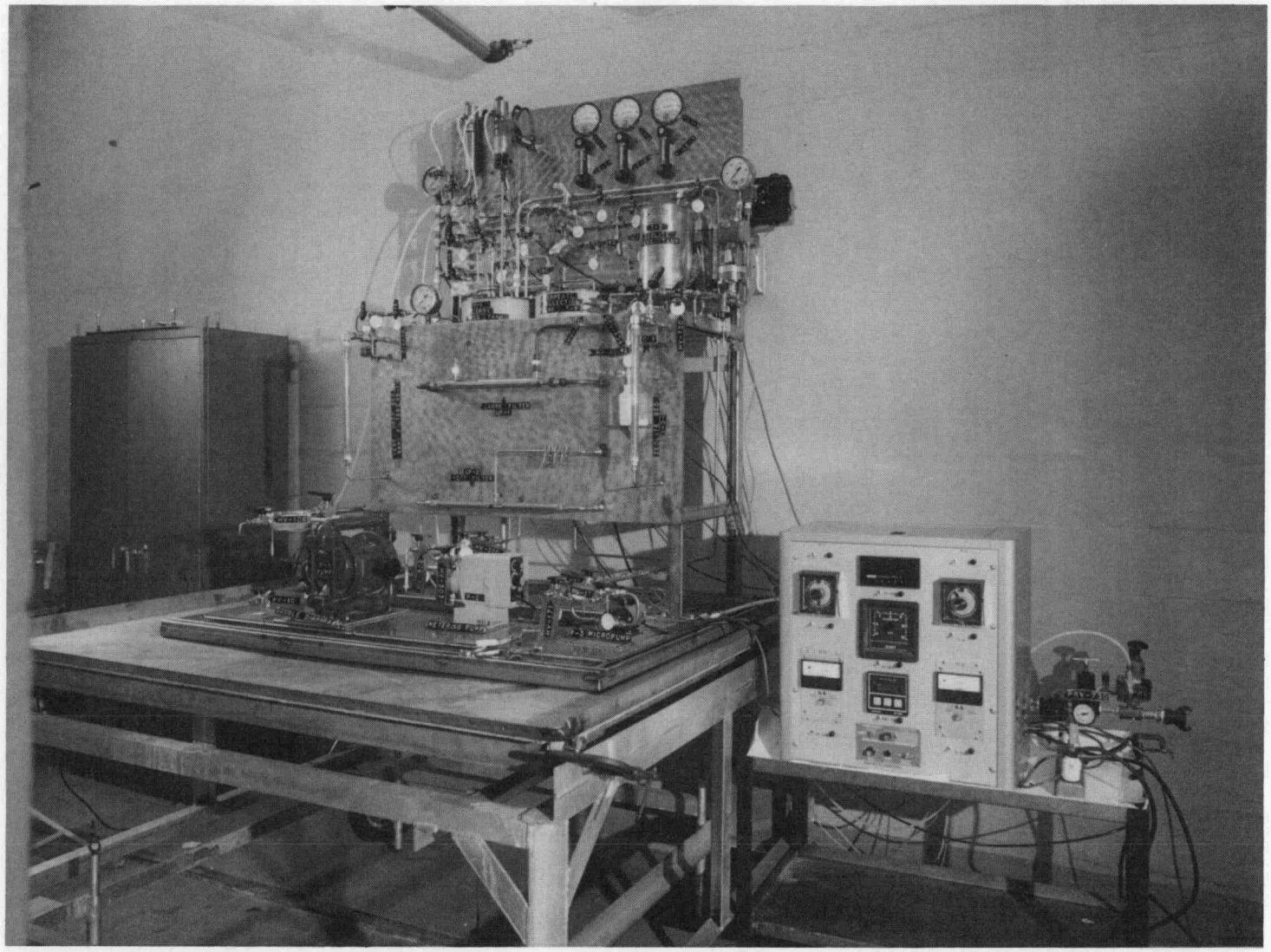

Fig. 2. Hot cell test loop.

All of the filtration tests were performed using a sample of MVST waste obtained from tank W-29 (on August 13, 1987) at the liquid and solids interface. The composition is presented in Table 1.

Table 1. Characterization of Melton Valley Storage Tanks W-29 waste used as feed material for cross flow filtration studies

Parameter	Analyses	Units
Gross alpha	1.82E+04[a]	Bq/mL
Gross beta	3.46E+05	Bq/mL
Gross gamma	2.20E+06	c/m/mL
^{233}U	3.73E+03	Bq/mL
^{238}Pu	5.81E+03	Bq/mL
^{239}Pu, ^{240}Pu	1.75E+03	Bq/mL
^{244}Cm	6.92E+03	Bq/mL
^{60}Co	2.06E+04	Bq/mL
^{134}Cs	7.07E+03	Bq/mL
^{137}Cs	2.25E+05	Bq/mL
^{152}Eu	1.01E+04	Bq/mL
^{154}Eu	6.00E+03	Bq/mL
^{155}Eu	1.91E+03	Bq/mL
^{90}Sr	8.06E+04	Bq/mL
Na^+	8.75E+01	mg/mL
NO_3^-	2.50E+02	mg/mL
pH	13.3	
Specific gravity	1.2466	
Total solids	384	mg/mL
Total dissolved solids	379	mg/mL

[a]Equivalent to 492 nCi/mL.

No cations other than sodium are reported in the table. Because of the high dilution factors necessary for cation analysis by the Inductively Coupled Plasma procedure (ICP) in a "cold" laboratory, all other cation assays resulted in concentrations at or below the detection limits.

Radiation readings using a "cutie-pie" were obtained at various points on the feed tank after charging with 11 L of the MVST waste. The radiation readings and a schematic drawing of the tank are shown in Fig. 3.

Temperature changes of the feed, in the range of 20 to 30°C, were encountered because of heat input from the feed pump and varying air temperatures within the cell. Because of the inverse relationship between viscosity and flux, all flux data reported in this study were normalized to a liquid viscosity consistent with 25°C through use of the following equation:[3,4]

$$\mu = Ae^{B(1/T)}, \tag{1}$$

where

μ = absolute viscosity, g/cm·s;

A = a function of the density of the fluid specific for momentum transfer;

B = an energy function related to the latent heat of vaporization, assumed to be the same for viscosity;

T = filtrate temperature, K.

Since flow is inversely proportional to the viscosity,

$$Q_1\mu_2 = Q_2\mu_1 \ . \tag{2}$$

Substituting Eq. (1) into Eq. (2) then becomes

$$Q_2 = Q_1 e^{B[(1/T_2)-(1/T_1)]} \ , \tag{3}$$

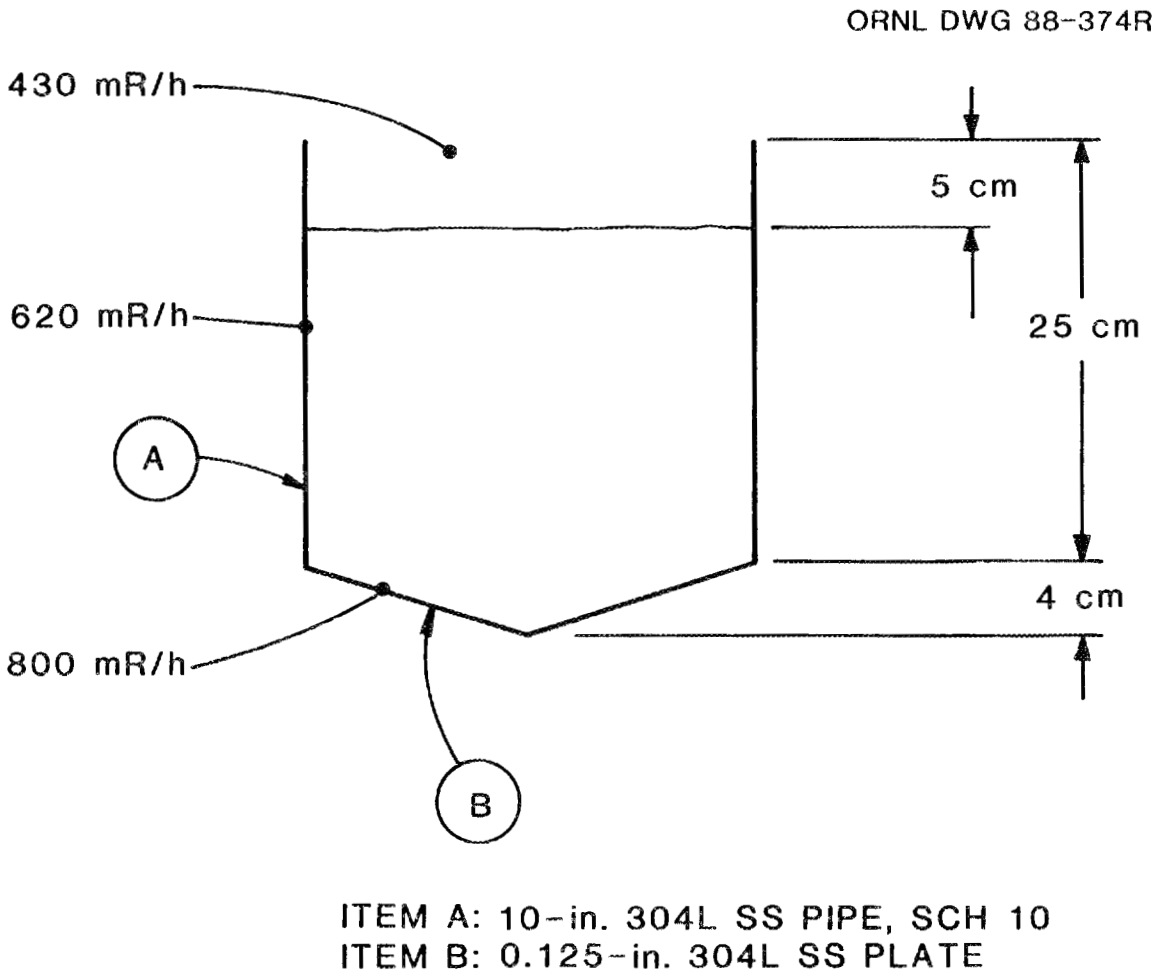

Fig. 3. Hot cell test loop feed tank schematic with radiation levels resulting from 11.0 L of Melton Valley Storage Tanks W-29 waste.

where

Q_1 = flux at T_2, L/min·m^2;

Q_2 = flux corrected to 298 K, L/min·m^2;

B = 2500;

T_1 = 298 K;

T_2 = actual liquid temperature, K.

The value of B in Eq. (3) was determined from absolute viscosity measurements of the filtrate at 24 and 30°C, which were 0.0135 and 0.0113 g/cm·s, respectively.

4.1 SCOUTING TESTS

Several scouting tests, each of ~6-h duration, were performed with the Mott and CARRE, Inc., filters to determine optimum operating conditions with respect to filtrate rate and TRU isotope (or gross alpha) rejection. Parameters evaluated were the feed flow rate (linear velocity through the filter tube) and filtration pressure. These tests were performed with the system operating in total recycle mode.

Five scouting tests were conducted with Mott 0.5-μm filters, one test with a Mott 2.0-μm filter, and three tests with the CARRE, Inc., ZOSS membrane filter. Cyclic backwashing of the Mott filter elements during operation was employed. Backwashing occurred at 15-min intervals for a duration of 0.4 s using air at 90 psig. The air pressure provided the driving force for forcing a portion of the filtrate from the backwash reservoir back through the filter tube wall. No backwashing of the CARRE filter occurred. The operating conditions and the resulting filtrate rates for all three filters are shown in Table 2, as is the gross alpha

Table 2. Filtrate rates as a function of feed rates and filtration pressures obtained with the cross flow filters

Feed[a] velocity (m/s)	Filtration pressure (KPa)	Filtrate flux[b] ($L/min \cdot m^2$)	Filtrate alpha conc. (nCi/mL)
	Mott filter, 0.5 μm bare tube		
1.5	140	4.1	0.72
3.0	140	3.2	0.32
1.5	280	7.0	0.07
3.0	280	5.0	0.02
1.5	140	3.7	0.14
	Mott filter, 2.0 μm bare-tube		
1.5	280	6.0	0.17
	CARRE filter, 0.5 μm membrane		
1.5	140	0.14	0.01
3.0	240	0.27	0.39
1.5	280	0.32	0.12

[a]Feed gross alpha content = 492 nCi/mL.
[b]The reported values for flux and gross alpha content are averages for the duration of each test.

content of the filtrate. The reported alpha concentrations are at essentially background levels. The low gross alpha concentration prevented analyses of the filtrate for TRU isotopes, but it is obvious from the reported assays that >99% of these isotopes were rejected by the three filter elements.

The filtrate samples were also analyzed for beta-gamma-emitting isotopes. The radionuclide concentrations of the feed and filtrate of samples obtained during the first Mott filter test are shown in Table 3.

Beta-gamma assays of samples obtained during tests with all three filters were ±5% of these values.

Table 3. Typical beta-gamma assays of feed and filtrate samples

Constituent	Feed (Bq/ml)	Filtrate (Bq/ml)
Gross beta	3.45E+05	2.02E+05
Gross gamma	2.20E+06[a]	1.76E+06[a]
^{90}Sr	8.05E+04	1.53E+03
^{134}Cs	7.07E+04	6.69E+03
^{137}Cs	2.25E+05	2.21E+05
^{60}Co	2.06E+04	1.11E+03
^{152}Eu	1.01E+04	N.D.[b]
^{154}Eu	6.00E+03	N.D.
^{155}Eu	1.91E+03	N.D.

[a]Reported as Ci/min/mL.
[b]N.D. denotes isotope not detected.

The above data indicate that ≥95% of the ^{60}Co and ^{90}Sr are removed by filtration. Essentially all of the europium isotopes are also removed. These data indicate that of the isotopes detected in MVST W-29, only ^{137}Cs exists almost entirely in the soluble state.

Based on the data presented in Table 2, the optimum feed velocity with the Mott 0.5-μm bare-tube filter element appears to be ~1.5 m/s with respect to filtrate flux, which is consistent with the results obtained during filtration of the simulated waste.[2] Increasing the filtration pressure from 140 to 280 KPa resulted in an average increase in flux of ~80% at a feed velocity of 1.5 m/s. At the conditions tested, the filtrate rates produced by the Mott 0.5-μm bare-tube filter were in the range of ~10 to ~20 times greater than that produced by the CARRE ZOSS

filter. The average filtrate rate obtained with the 2.0-μm Mott filter was 14% less than the flux obtained in earlier tests with the 0.5-μm Mott filter at the same conditions. These data are compared in Fig. 4. Filtrate assays were essentially identical to those of all earlier tests with the Mott 0.5-μm filter. The flux data indicate that the MVST solids penetrate deeper into the 2.0-μm pores than into the 0.5-μm pores and are more difficult to remove at the backwash conditions used in these tests. The end result is that the average filtrate rate is lower for the larger pore diameter filter. Based on this data, the 2.0-μm Mott filter was not tested further.

All of the data (filtrate rates) collected during the scouting tests with the 0.5-μm Mott and CARRE filters is shown in Figs. 5 and 6, respectively. As can be seen in Fig. 5, the filtrate rates obtained with the 0.5-μm Mott filter element appear to be inversely proportional to the feed velocity at a given filtration pressure. However, because of the limited data, no absolute correlations can be made.

Filtrate samples obtained during filtration tests with the CARRE membrane filter indicated that the ZOSS membrane may have been compromised. During the MVST filtration tests with the Mott filter (~30 h total time), all of the 30 filtrate samples were free of any discoloration. The first filtrate sample obtained 30 min after startup with the CARRE filter was also clear. The second filtrate sample, obtained 2.5 h later, was yellowish in color as were all subsequent samples from the remainder of the CARRE studies. This sudden appearance of color was a possible indication of some dissolution of the ZOSS membrane; therefore, only three filtration tests with the CARRE filter

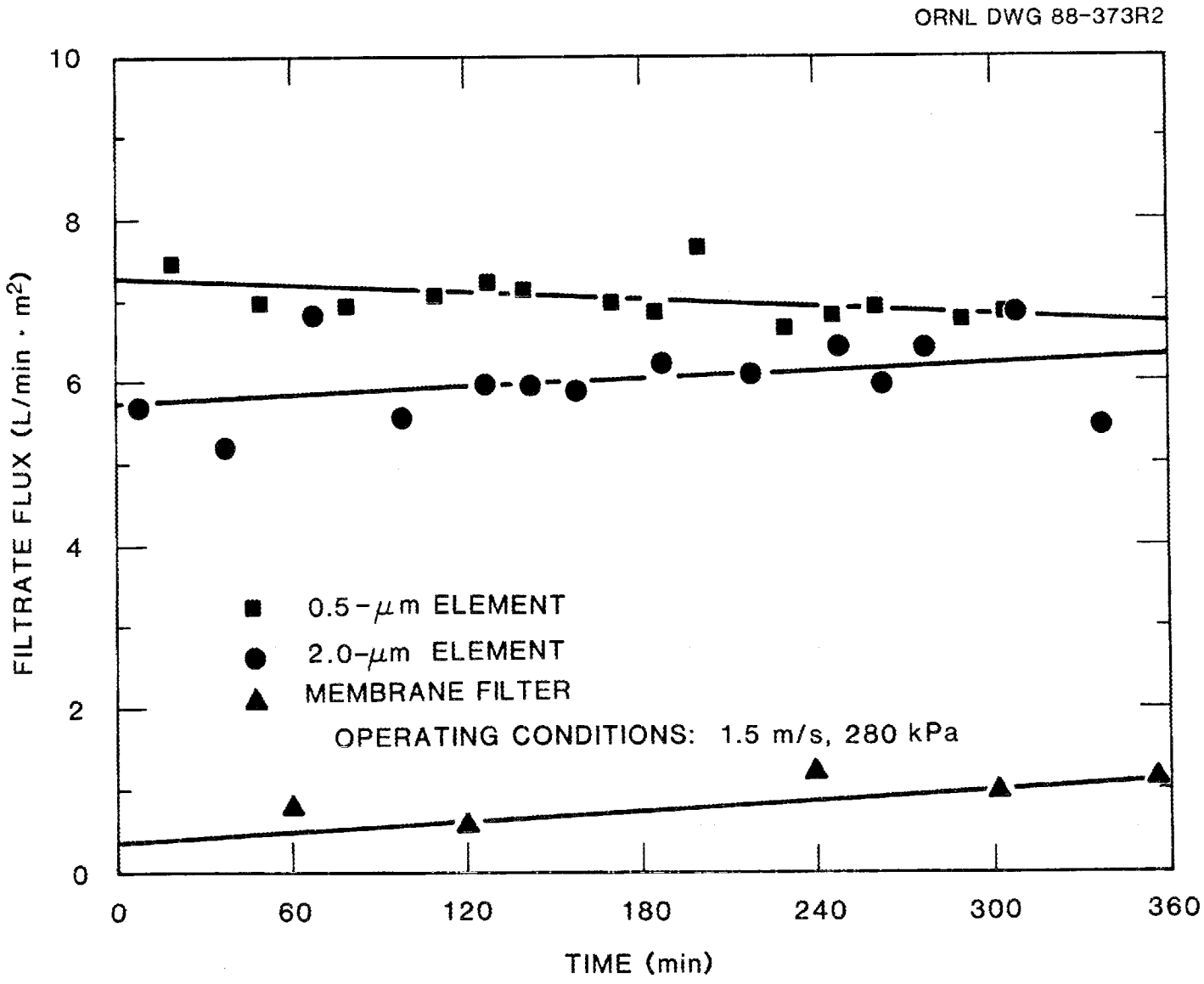

Fig. 4. Scouting studies: filtration of Melton Valley Storage Tanks waste with Mott filter elements; flux as a function of pore size vs time.

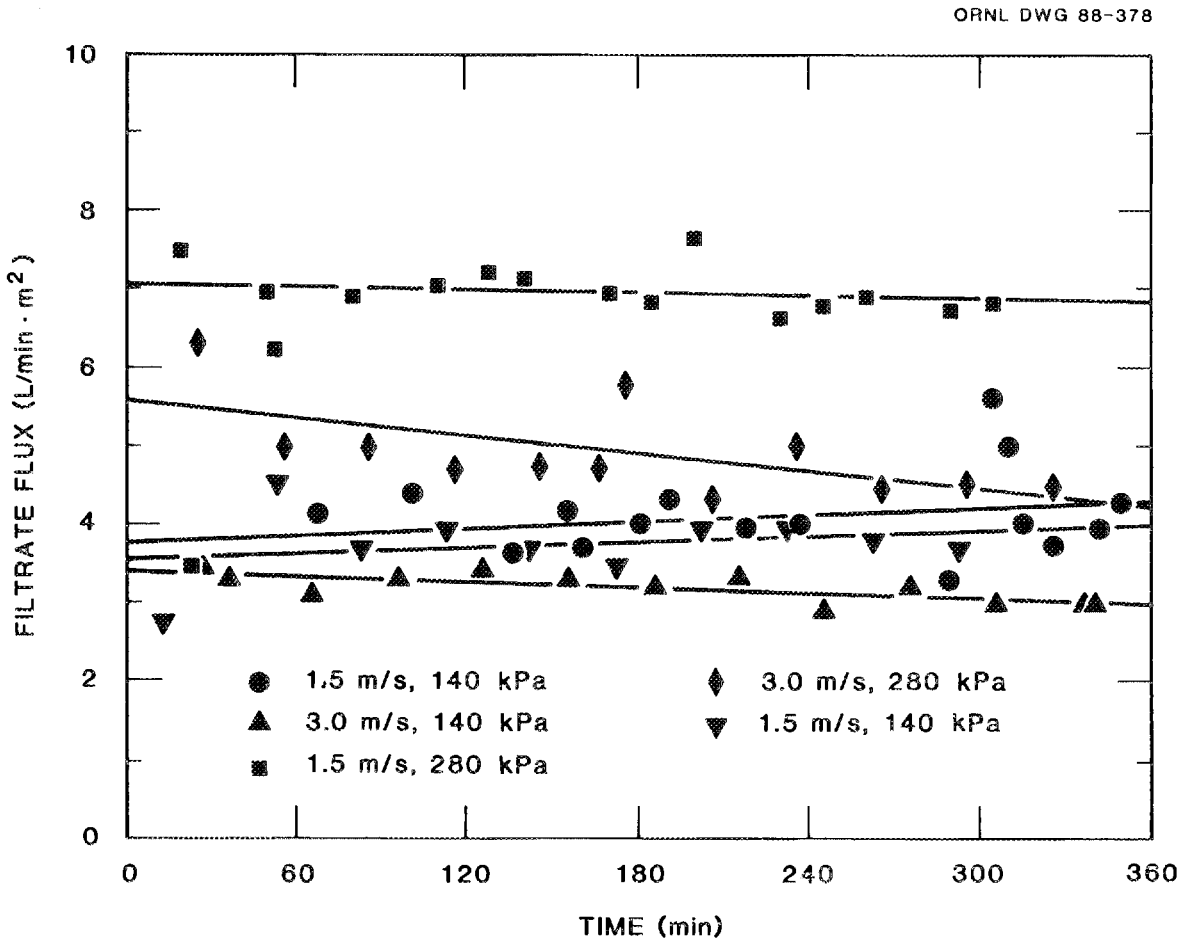

Fig. 5. Scouting studies: filtration of Melton Valley Storage Tanks waste with a Mott 0.5-mm filter element; flux as a function of flow rate and pressure with respect to time.

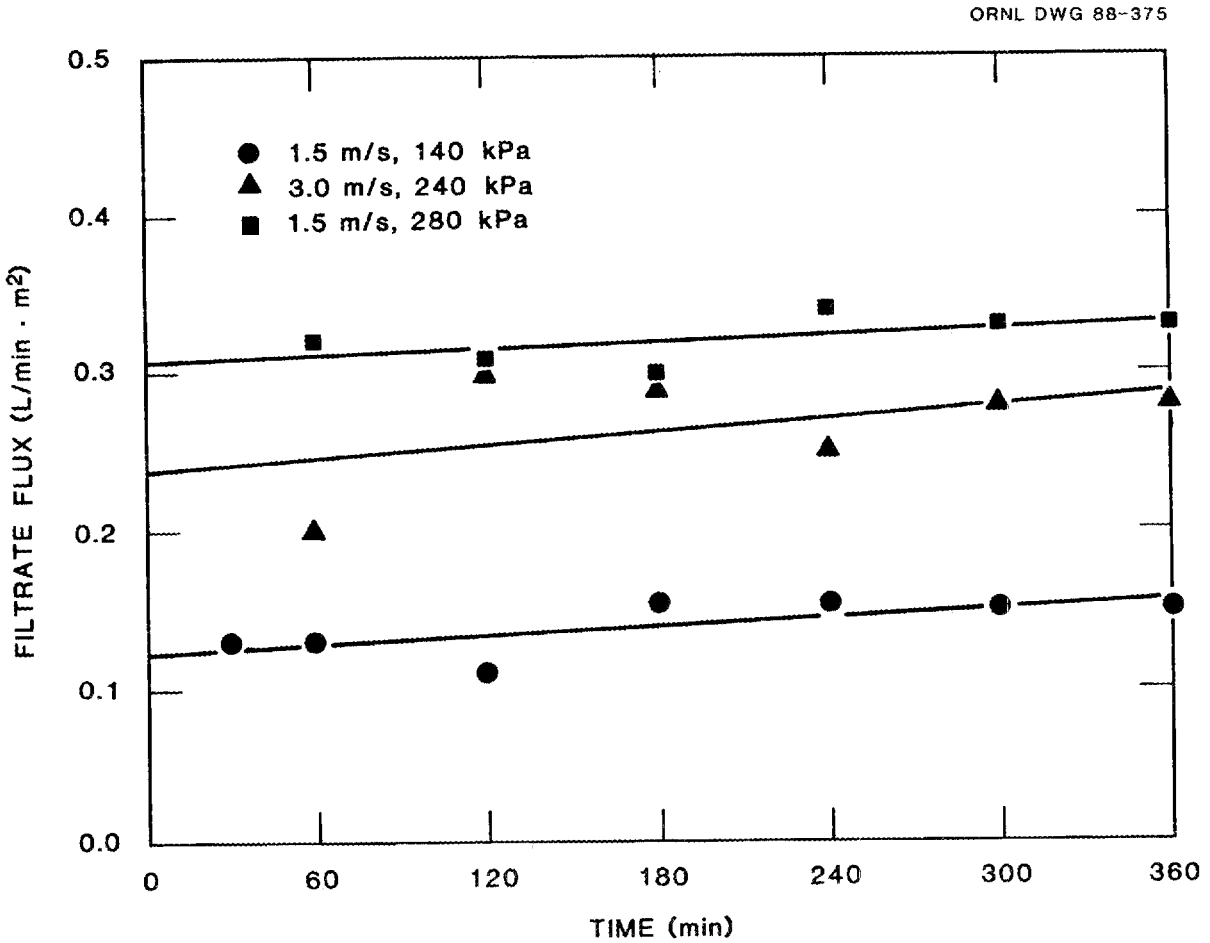

Fig. 6. Scouting studies: filtration of Melton Valley Storage Tanks waste with a CARRE, Inc., ZOSS membrane filter element; flux as a function of flow rate and pressure with respect to time.

were made. Dissolution of the membrane was expected at the pH of the MVST feed (~13), which did occur during the simulated waste studies,[2] but it is surprising that the attack occurred within 3 h of operation. Earlier projections of the expected life of the ZOSS membrane in the MVST environment was on the order of six months.[5] The limited data obtained does indicate that by increasing the filtration pressure from 140 to 280 KPa (at 1.5 m/s feed velocity), an increase in flux of 129% was realized. No correlations between feed velocity and filtrate rates can be drawn from these data.

Following the CARRE studies, one additional test was made with the Mott 0.5-μm filter. The filtrate samples obtained during this run also contained the yellow color, but the filtrate rate and gross alpha rejection were essentially the same as those obtained in all of the earlier Mott filter tests.

Since the average filtrate rate for the 0.5-μm Mott filter was higher than that obtained with the Mott 2.0-μm and the CARRE, Inc., filters, and because the stability of the CARRE membrane in the MVST environment is questionable, the 0.5-μm Mott filter was selected for additional testing.

4.2 LONG-TERM TEST

A long-term filtration of MVST waste was undertaken to determine the expected service life of a new 0.5-μm Mott bare-tube filter element. Baseline clean water data indicated that the new filter element was essentially identical (with respect to flux) to the 0.5-μm filter used in the scouting tests. Initial operating conditions were as follows:

Filtration pressure:	140 KPa
Linear feed velocity:	1.5 m/s
Backwash conditions:	15-min intervals
	0.4-s duration
	90-psig air pressure

The system was operated continuously for 575 h at these conditions and in a total recycle mode (i.e., constant feed TSS content). At this time, the filtration pressure was increased to 280 KPa, and operation continued for an additional 425 h. The filtrate flux data is shown in Table 4.

The data shown in Table 4 indicate that the flux at 140 KPa filtration pressure was essentially constant through the first 71 h of operation. The flux decline evidenced through the next 504 h of operation indicates the probable formation of a subsurface membrane. Immediately after increasing the filtration pressure to 280 KPa, the flux began to increase and reached a maximum of 5.03 $L/min \cdot m^2$ after 76 h at this pressure. The flux remained essentially constant for the remainder of the run. Filtrate assays indicate background levels of gross alpha for the duration of the test. The rejection and filtrate flux data are shown in Fig. 7.

The data obtained at a filtration pressure of 140 KPa indicate that a filtration area of 3.7 m^2 (40 ft^2) could produce 378,500 L (100,000 gal) of filtrate over a 682-h period. At a filtration pressure of 280 KPa, 400 h would be required to produce the same volume of filtrate. These data indicate that the flux decay that occurs at a filtration pressure of 280 KPa is less severe than at 140 KPa. Therefore, based on this study, 280 KPa is the recommended filtration pressure.

Table 4. Filtrate flux as a function of time at filtration pressures of 140 and 280 KPa

Time (h)	Flux ($L/min \cdot m^2$)
140 KPa filtration pressure	
24	5.55
71	5.63
95	3.83
170	2.91
215	2.18
315	1.76
484	1.35
575	1.01
Average	2.50
280 KPa filtration pressure	
576	2.52
604	3.63
651	5.03
671	4.50
1000	4.30
Average	4.30

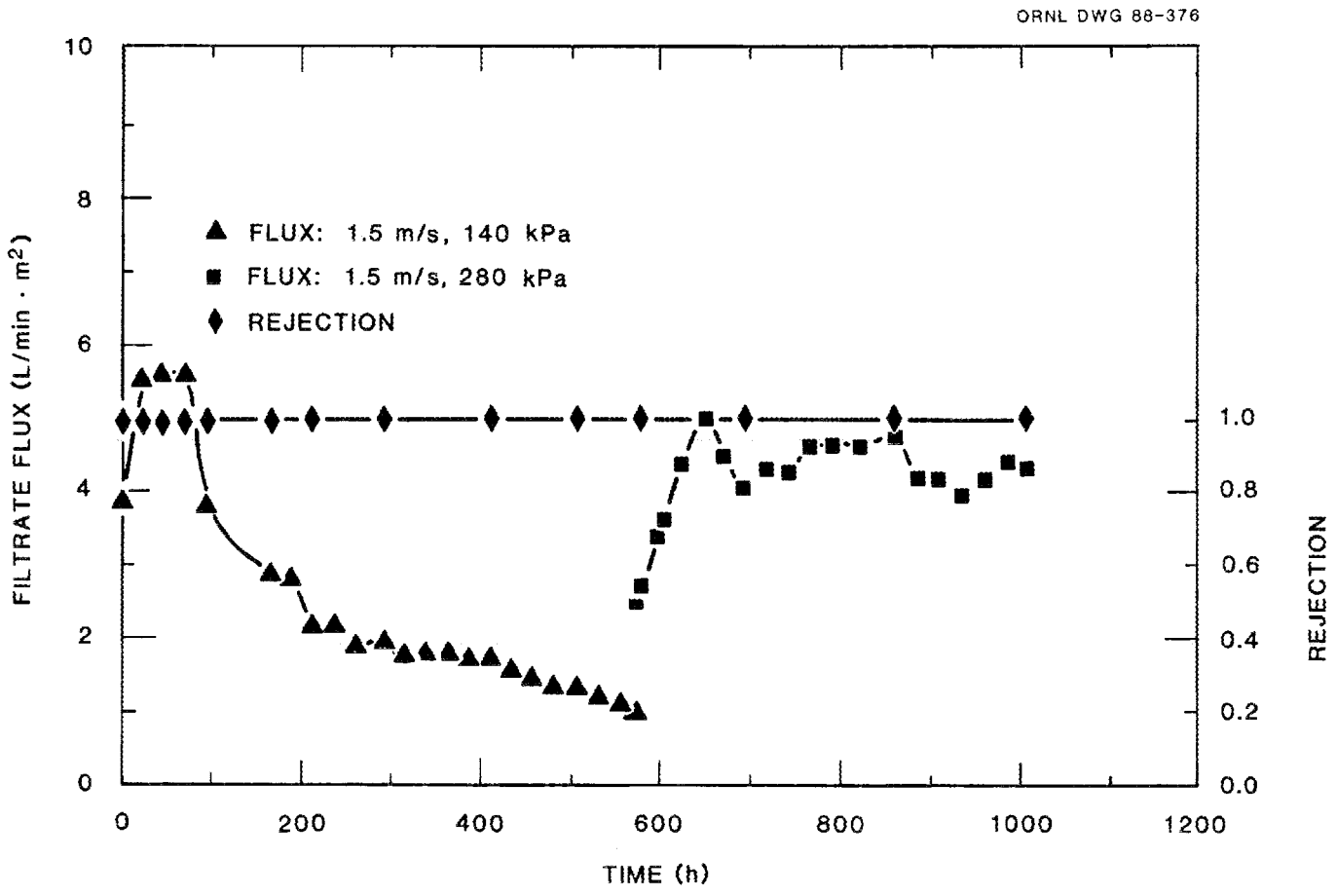

Fig. 7. Long-term test: filtration of Melton Valley Storage Tanks waste with a Mott 0.5-mm filter element; flux and rejection as a function of filtration pressure with respect to time.

After 1000 h of operation, the system was placed in a volume reduction mode to determine filtration efficiency with respect to an increase in TSS content of the feed. Operating parameters were not changed. No problems were encountered in achieving a volume reduction of 85% over a time span of 4 h. The average flux obtained during this time span was 4.36 $L/min \cdot m^2$ (data shown in Fig. 8). This volume reduction increased the TSS content of the feed from 0.5 to 3.3 wt % and the gross alpha content from 490 to 3250 nCi/mL. Based on studies with simulated waste, no problems would be encountered with cross flow filtration of a slurry containing up to 50 wt % TSS.[2] At the termination of this test, the gross alpha content of one composite filtrate sample was at background levels, indicating essentially 100% rejection.

4.3 CLEAN WATER TESTS

Clean water tests (using process water at the naturally occurring pH of 7.6) were performed on each new filter to obtain baseline filtrate rates. Additional clean water tests were performed periodically to determine the extent of filter element fouling. A dispersant dye (0.2-μm nominal size) was added to the water as an indicator of filtrate quality in several tests.

4.3.1 Baseline Data

The CARRE, Inc., filter was operated for 100 min at a filtration pressure of 270 KPa and with a linear feed velocity of 2.8 m/s through the tube. The average filtrate flux for this period was 0.52 $L/min \cdot m^2$. Backwashing of the filter tube was not employed as there was no evidence of a decline in flux.

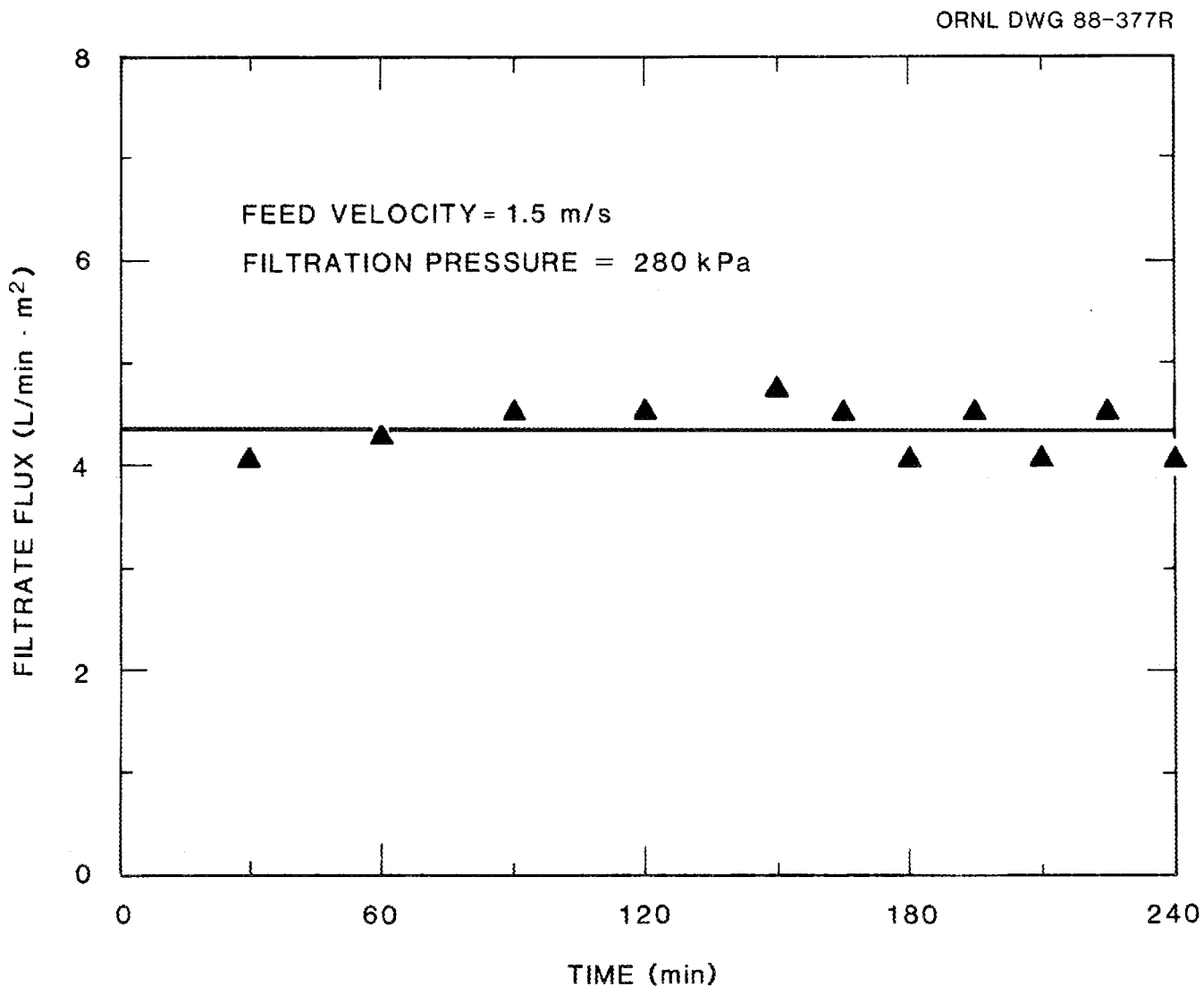

Fig. 8. Volume reduction test: filtration of Melton Valley Storage Tanks waste with a Mott 0.5-mm filter element; flux as a function of an 85% reduction in feed volume.

The 0.5-μm Mott filter used in the scouting tests was operated for 135 min at a filtration pressure of 180 KPa and with a linear feed velocity of 1.5 m/s. Backwashing occurred at 15-min intervals for a duration of 0.4 s using air at 90 psig. The average filtrate flux was 13.4 L/min·m^2. The 0.5-μm Mott filter used for the long-term test was operated at the same conditions but for a 75-min period. The average filtrate flux for this time period was 13.2 L/min·m^2.

A filtration pressure of 280 KPa was used to obtain baseline data for the 2.0-μm Mott filter. The linear feed velocity remained at 1.5 m/s. The filtrate flux 14 min after startup was 49 L/min·m^2, a factor of 2.1 times greater than that produced by the 0.5-μm filter for the same time period. This rate decreased to 13 L/min·m^2 during the next 15 min. During the final 30 min of operation, the average flux for the 2.0- and 0.5-μm filters was 11 L/min·m^2. These data are compared in Fig. 9 and indicate that suspended particles in the process water penetrate the filter tube walls to form a subsurface membrane that effectively reduces the nominal pore size of both filters to $\leq$0.5 μm. Average dye rejection values were 91 and 11%, respectively, for the 0.5- and 2.0-μm filters.

4.3.2 Final Clean Water Tests

Clean water tests of ~2-h duration with both the 0.5-μm Mott and CARRE filters followed the MVST filtration scouting studies. A dispersant dye of 0.2-μm nominal particle size was added to the water to quantify the filtrate quality with respect to dye concentration. Operating conditions were consistent with the initial clean water tests. Both filters were tested with process water at a pH of 7.6, the natural pH of the water, which was also consistent with the initial tests. Then NaOH was used to

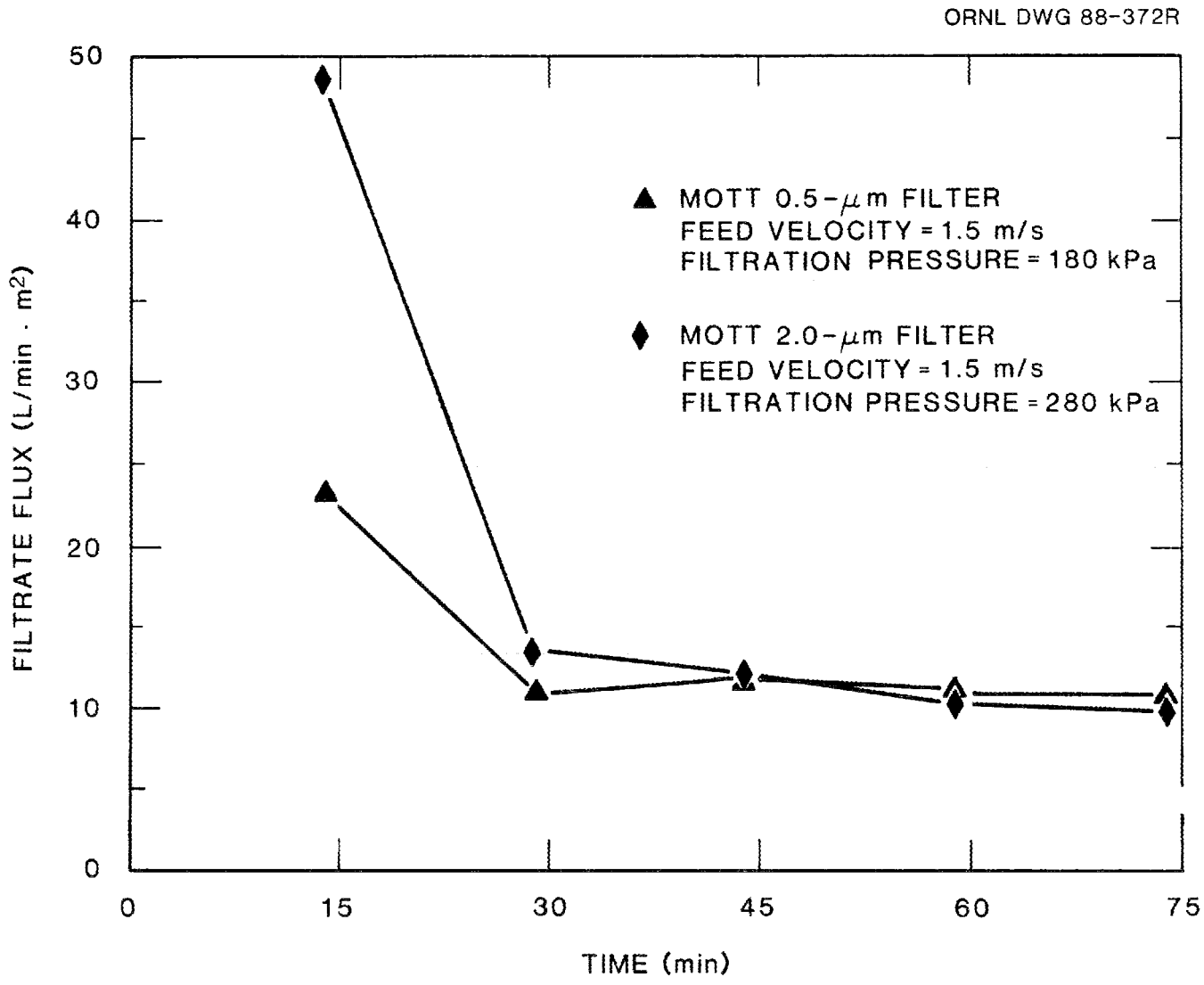

Fig. 9. Mott initial clean water test; flux as a function of pore size.

adjust the pH to 13.9,and the water was retested. This pH adjustment was made to determine the filtrate flux and dye rejection as a function of pH. The flux data is compared to the initial baseline data in Fig. 10. These data indicate that the flux is not affected by pH at the two pH values tested. The CARRE flux data indicate essentially no change from the as-received filter. The Mott data from the postfiltration tests indicate a flux decrease to ~20% of that obtained with the as-received filter. As discussed earlier, this is consistent with the formation of a subsurface membrane in a bare-tube filter element. Based on the flux data, it is not evident that any dissolution of the CARRE ZOSS membrane occurred during the scouting studies.

The effect of water pH with respect to dye rejection was significant. Analysis of the filtrate by spectrophotometer for dye content indicates a rejection of 86 and 91% for the CARRE and Mott filters, respectively, at a pH of 7.6. Rejection decreased to 38 and 29% for the Mott and CARRE filters, respectively, at a pH of 13.9. These data are shown in Fig. 11. Unfortunately, dye was not used in the initial clean water tests, so this data does not permit evaluation with respect to dye rejection of the as-received filters. This rejection data is also inconclusive as to the extent of dissolution of the CARRE ZOSS membrane. There is no incontrovertible explanation for the increased passage of the dye through the filters at pH 13.9; abnormally low filtration rates are normally associated with improved particle rejection. It can only be assumed that the membrane or subsurface layers are pH sensitive. Resolution of the mechanisms behind these system behaviors is likely to require a major

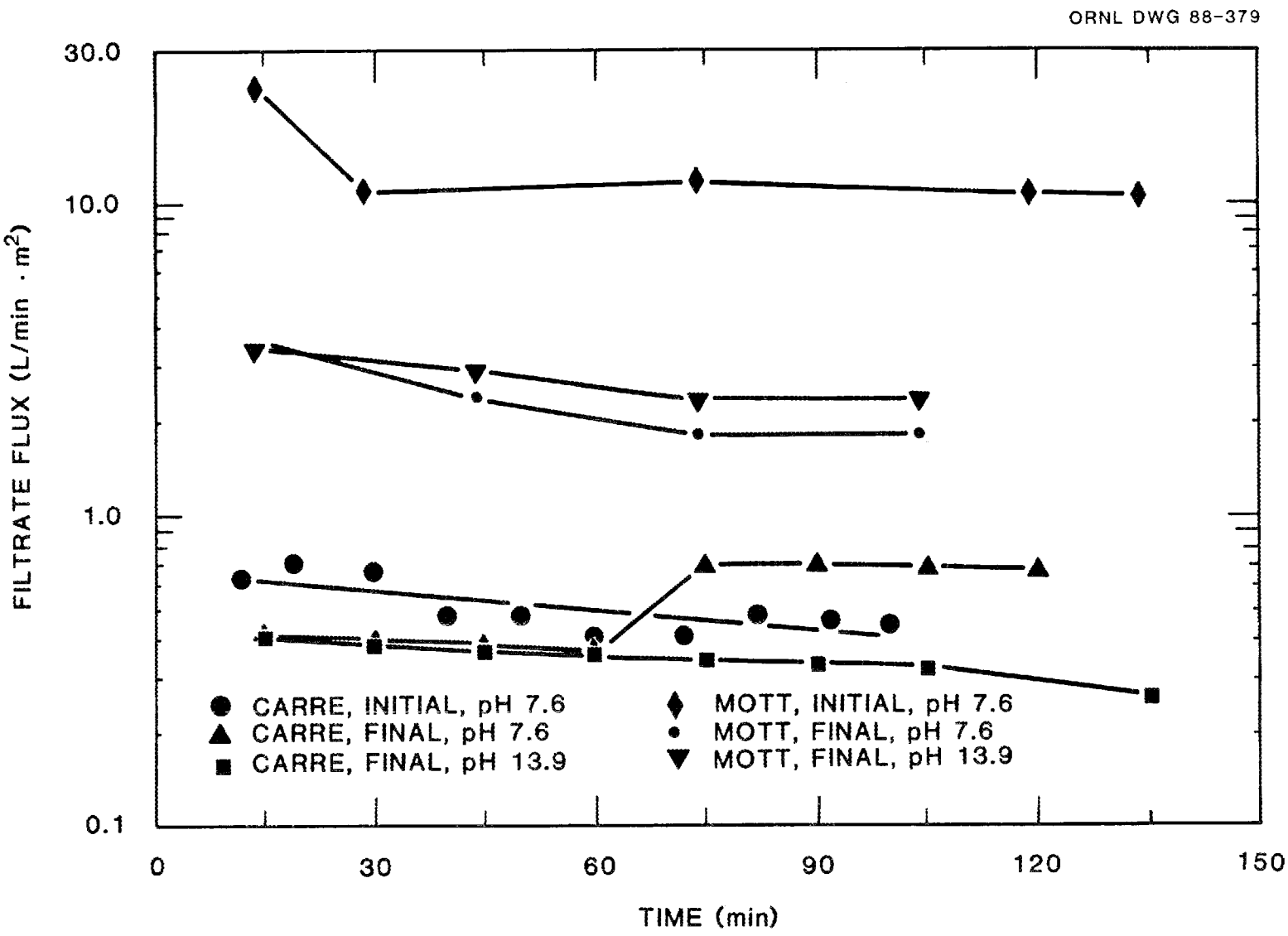

Fig. 10. Clean water test; flux as a function of water pH.

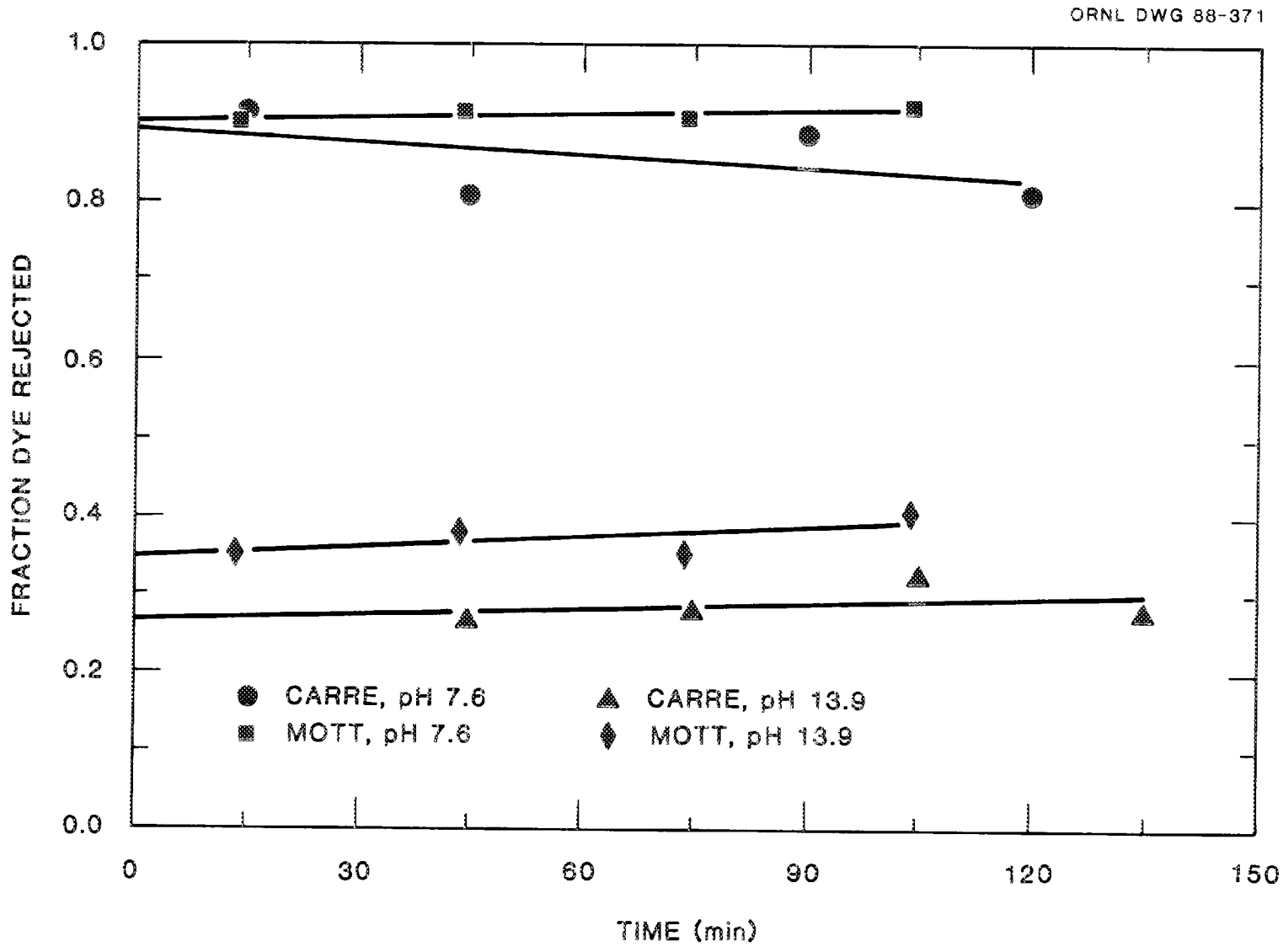

Fig. 11. Final clean water test; dye rejection as a function of pH.

effort, and this is difficult to justify when all operating conditions to date have produced filtrate with gross alpha concentrations of <1 nCi/mL.

Clean water tests, acid cleaning, and final clean water tests were performed with the 2.0-μm Mott filter element used in the 6-h scouting test and the 0.5-μm Mott filter used in the long-term test. A dispersant dye (50 mg/L, 0.2-μm nominal particle diameter) was added to the water for these tests to determine filtrate quality.

The following operating parameters were used for these tests:

	Mott filter element	
	0.5 μm	2.0 μm
Linear feed velocity (m/s)	1.5	1.5
Filtration pressure (KPa)	180	280

The elements were backwashed as in all previous tests: 15-min intervals, 0.4-s duration, and 90-psig pressure. The run time for each of the clean water tests was 75 min.

Following the previously described clean water tests, nitric acid was added to the feed tank to produce a feed water of 1.7 M H^+ concentration. Both filters were then operated at the conditions listed for the 2.0-μm Mott filter element in the preceding spot table for a period of ~8 h. Following the acid-cleaning step, samples of the acid were analyzed for gross activities, and the assays indicated that the acid solution contained 0.3% of the gross alpha, 1.5% of the gross beta, and 2.1% of the gross gamma activities that were contained in the original MVST feed

solution. It cannot be determined from this data if the acid-cleaning step removed all the activity trapped within the porous filter tube walls, but it is clear that 1.7-M nitric acid will dissolve at least a portion of the particulate activities. The clean water tests (as in the preceding spot table) were repeated after the acid-washing tests, the average flux and dye rejection values obtained are shown in Table 5. The data from the baseline clean water tests are included for comparison.

Table 5. Average flux and rejection values obtained with the Mott filter elements using process water and 1.7 M H^+ as feed

Filter condition	Filter element			
	0.5 μm		2.0 μm	
	Flux[a] ($L/min \cdot m^2$)	Rejection[b]	Flux[a] ($L/min \cdot m^2$)	Rejection[b]
Prefiltration[c]	13.2	0.91	21.2	0.11
Postfiltration[c]	2.9	0.84	6.5	0.56
Acid cleaning[d]	191.1	----	260.9	----
Postacid[c]	101.9	0.11	249.8	0.07

[a]Flux values are averages for the duration of the tests.
[b]Rejection values are reported as fraction of dye removed by filtration and are averages for the duration of the tests.
[c]Process water at the normally occurring pH of 7.6.
[d]Process water adjusted with HNO_3 to 1.7 M.

As shown in Table 5, the flux rates obtained during the postacid tests were factors of ~8 and ~12 higher (for the 0.5- and 2.0-μm filters, respectively) than were obtained with the as-received filters. The dye rejection capabilities of the 0.5- and 2.0-μm filters were reduced to 63 and 12%, respectively (following acid cleaning), compared to the rejection values of the as-received filters.

The data presented in Table 5 suggest that metal oxides or other materials in the Mott filter tube matrix were dissolved during the acid-cleaning procedure, thus effectively increasing the pore size. This increase in pore size then resulted in extremely high filtrate rates and low rejection of the dispersant dye. One reference reports that a set of plugged (20–25% of the original filtrate rate) 0.5-μm Mott filter elements was soaked in a nonagitated 15% nitric acid (2.6-M) bath for 60 h.[6] When returned to service, approximately 75% of the original filtrate flow rate was recovered. The cleaning procedure recommended by Mott Metallurgical personnel is to soak the filter tubes in an unagitated solution of 5% HNO_3 for 20 to 30 min.[7]

To determine the metals dissolved from the filter by cleaning with strong nitric acid, the following test was conducted. Three liters of 2.0-M nitric acid was prepared using deionized water. This acid solution was circulated through the tube walls of a new 0.2-μm nominal porosity Mott bare-tube filter element for 48 h. All components of this circulation loop were of plastic materials to prevent inadvertent introduction of any metals to the acid solution. Samples of the initial and final acid solutions were submitted for ICP analyses. The results of these assays indicate that 69 mg Fe, 11 mg Cr, 12 mg Ni, and 1.4 mg Mo were dissolved from the filter element. These assays are consistent with the composition of the material of filter fabrication (i.e., type 316L powdered stainless steel), so these data clearly indicate that partial dissolution of the Mott filter element did occur under the conditions used for this cleaning test. It is reasonable to assume that the filter

nominal pore size was enlarged and caused an increased flux with a corresponding decrease in rejection capabilities, as shown in Table 5.

Any future cleaning efforts should be confined to the use of HNO_3 in concentrations not to exceed 5%. The recommended procedure for filter cleaning in the HCTL is to pump only the volume of acid (5% strength) required to fill the inside diameter of the filter tube—not to force the acid through the walls of the tube, as was done in this test. After a 30-min contact period, use process water to flush the acid from the system before reusing the filter.

5. SUMMARY AND RECOMMENDATIONS

Three cross flow filter elements (0.5- and 2.0-μm Mott bare-tube filters and a 0.5-μm CARRE, Inc., ZOSS membrane filter) were used to filter a sample of MVST W-29 waste containing 0.5 wt % solids and 492 nCi/mL of gross alpha contaminants. All three filter tubes tested in this study produced filtrate containing <1 nCi/mL of gross alpha for an alpha rejection >99.8%. The three filters also rejected >95% of the ^{60}Co, ^{90}Sr, and europium isotopes that were contained in the feed. Based on the alpha rejection values, the alpha particles are apparently larger than 2.0 μm. Therefore, a bare-tube filter element of $\leq$2.0-μm nominal porosity is adequate to produce filtrate essentially free of TRU isotopes.

Filtration pressures used in these studies were in the range of 140 to 280 KPa. Linear feed velocities through the filter tube ranged from 1.5 to 3.0 m/s. The average filtrate flux obtained with the Mott 0.5-μm filter was a factor of 30 greater than that obtained with the CARRE ZOSS membrane filter and 1.4 times greater than the 2.0-μm Mott filter element.

Optimum operating conditions for the Mott 0.5-μm filter appear to be at a filtration pressure of 280 KPa and a linear feed velocity of 1.5 m/s. The filtrate flux at these conditions was 72% greater than at 140 KPa.

Tests were not carried out to determine the exact service life of the Mott 0.5-μm filter, although they did show that the service life is certainly greater than 1000 h. Assuming a 1000-h service life for a filter with a filtration area of 3.7 m^2, these studies indicate that ~1.1 million L of MVST waste could be processed during this time span, producing ~950,000 L of "non-TRU" filtrate (a volume reduction of ~86%) before the filter must be cleaned or replaced. This calculation assumes that feed with an increased solids concentration does not reduce the filtration rate significantly. This study indicates that a 0.5-μm bare-tube filter is adequate to treat the LLLW currently stored in the MVSTs.

Because the composition of the waste in the eight MVSTs varies widely, it is recommended that additional studies be conducted to determine the alpha rejection and service life of these filters with waste from each of the tanks. Tests in this study were conducted with waste from W-29, which contained only ~500 nCi/mL of gross alpha and ~0.5 wt % TSS. Other tanks are reported to contain up to ~6000 nCi/g gross alpha.[1] Tests should also be conducted with a starting TSS content of ~100 mg/mL, which would result in a higher gross alpha content of the feed. These tests should be used to evaluate the effect of a higher TSS content, particularly during volume reduction studies, with respect to filtrate flux and gross alpha rejection.

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