Evaluation of biologic and non-biologic methods for assessing virus removal by and integrity of high pressure membrane systems

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Abstract
This paper describes pilot-scale studies that examined three integrity test methods for: (1) quantifying virus removal by nanofiltration (NF) and reverse osmosis (RO) membrane systems when arranged in single element unit and two-stage system configurations, and (2) determining change in virus removal capability of such systems when subject to different types of membrane/o-ring compromisation and fouling. The three methods evaluated included one biologic type (MS-2 phage), that has been employed previously; and two, new non-biologic types (24-nanometre polystyrene fluorescent dyed microspheres and fluorescent Rhodamine WT [R-WT] dye, molecular mass 496 daltons). All three surrogates were employed in a manner intended to show a minimum of 4-logs removal by the NF and RO membranes selected for test. Methods of compromisation included a pinhole induced through one membrane leaf in the spiral wound NF/RO element, and both cracking of and removal of sections from one of the permeate tube o-rings. Testing was conducted on two source waters, representing brackish surface water and effluent categories: a microfiltered secondary effluent and a river water. The river water is characterized by low to moderate TDS and high TOC and was treated with conventional alum coagulation, flocculation, sedimentation and granular media filtration for subsequent membrane processing.

Keywords
Fluorescent microspheres; membranes; MS-2 phage; nanofiltration; reverse osmosis; Rhodamine WT dye; virus removal

Introduction
The Surface Water Treatment Rule (SWTR), promulgated in 1989, required the control of viruses, bacteria, and protozoa (e.g. Giardia lamblia) in drinking water treatment (EPA, 1989). This regulatory requirement has triggered an increasing interest in the application of membrane processes, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO), to provide an effective barrier for pathogenic microorganisms in drinking water supplies. Although RO membranes have been applied to remove microorganisms in water treatment as early as 1972 (Sorber et al., 1972), high-pressure (75 to 1,200 psi) membrane processes such as RO and NF have seen only limited use in the treatment of surface waters compared to low-pressure (5 to 50 psi) membrane processes such as MF and UF, mainly due to their relatively high capital and operational costs. However, the recent promulgation of the Disinfectants/Disinfection By-product Rule (D/DBP) and the interim Enhanced Surface Water Treatment Rule (ESWTR) have generated new interest in the use of high-pressure membrane processes because RO and NF membranes can provide high removals for both pathogens and disinfection by-product precursors (Gagliardo et al., 1997; Adham et al., 1998a,b).

The successful application of high-pressure membrane processes as effective barriers for pathogenic microorganisms requires the implementation of proper methods for monitoring and controlling membrane integrity during process operation. However, integrity methods currently used in full-scale RO and NF systems are generally limited to
conductivity and total organic carbon monitoring, approaches that can only assess pathogen removals up to maximum values of approximately 99 percent (2 logs). Given the capability of RO and NF membranes to provide much greater removals for viruses, *Cryptosporidium* oocysts, and *Giardia* cysts, it is critical to develop more reliable tools for assessing the actual pathogen removal capacity and integrity of high pressure membrane systems during process operation.

The main objective of this research is to determine the integrity of RO and NF membrane systems with respect to microbial passage using both microbial- and non-microbial surrogates. The focus of this paper is to present the results from pilot-scale tests. The bench-scale portion of the study was presented in an earlier publication (Colvin *et al.*, 2000).

**Experimental approach**

**Materials**

Bacteriophage MS2 (referred to as MS2 in this manuscript), a virus that utilizes *Escherichia coli* as a host, was used to assess the integrity of membranes because this microorganism is similar in shape, size, and nucleic acid composition to enteric viruses. Orange-colored fluorescent-dyed carboxylate-modified microspheres (referred to as microspheres in this manuscript) (Molecular Probes, Eugene, OR) and Rhodamine WT (referred to as R-WT in this manuscript) were used as non-microbial surrogates. The microspheres had a size of $24 \pm 5$ nm, comparable to the size of MS2, and could be measured at concentrations as low as approximately 50 ng/L, thus allowing determination of rejections greater than 99.99 percent (4 logs) using relatively low feed concentrations (approximately 1 mg/L). R-WT was chosen because it was a non-reactive tracer chemical approved by EPA for use in water treatment plants, and expected to be rejected well by both RO and NF membranes considering its molecular weight of 480 Daltons. R-WT could be quantified at levels as low as 10 to 20 ng/L in permeate allowing detection of 4- to 5-log removal at a feed concentration of 0.1 to 1 mg/L. A minimum detection level of 4-log removals for the non-microbial surrogates was targeted in order to demonstrate the 4-log removal requirements for viruses in the SWTR and ESWTR. Pilot experiments were performed with two types of spiral-wound elements: Hydranautics Model ESPA1-4040-UHT RO element (referred to as ESPA1 RO in this manuscript) (Hydranautics, Oceanside, CA) and Koch Fluid Systems Model TFCS-4040S NF element (referred to as TFCS NF in this manuscript) (Koch Membrane Systems, Wilmington, MA). Both elements had a diameter of 4 inches, and a length of 40 inches.

**Pilot-scale membrane treatment facilities**

Pilot-scale experiments were conducted at two sites: Water Factory 21 (WF-21) at the Orange County Water District (OCWD) in Fountain Valley, CA, and the Northwest River Water Treatment Plant (NWR-WTP) in the City of Chesapeake, VA. The source waters used were micro-filtered secondary effluent (MFSE) for the WF-21 tests, and conventional process-treated river water (CTRW) for the NWR-WTP tests. Waters from WF-21 and NWR-WTP were selected to reflect two generic types of source waters, i.e., surface waters and reclaimed wastewater effluent, for which disinfection or removal credit for pathogens by RO and NF would most likely be required.

Two units were employed for testing in WF-21: a dual-element test unit (DETU) (Figure 1), and a multi-stage test unit (MSTU) (Figure 2). The DETU consisted of two pressure vessels arranged in series, operated in single-pass mode with no concentrate or permeate recycled to the feed. The ESPA1 RO element was operated in the lead vessel and the TFCS NF element in the trailing vessel. The test surrogates were injected into the feed line using an electromagnetic dosing pump. Pressure was measured in the feed and concentrate of each
vessel using a single pressure gauge coupled to a 4-way selector valve. Permeate flow rate in each vessel and the final concentrate flow rate were measured using in-line flowmeters. Samples were collected from the feed (from an in-line sample port), the permeate from each vessel, and the final concentrate. The MSTU setup consisted of two separate flow streams each in parallel with two vessels in series (i.e. four lead vessels) followed by combined flow with a series of two vessels in series (Figure 2). Each vessel was designed to hold three spiral-wound elements. Permeate flows were regulated by a valve located in the final concentrate line. Permeate and concentrate flow rates were measured using in-line flowmeters. A single pressure gauge coupled to a four-way selector valve was used to measure the pressure difference between feed and concentrate at different locations (i.e. vessel 1 and 2, 3 and 4, 5 and 6). Samples were collected from the feed, final concentrate, individual vessel permeates and combined permeate through in-line sample ports.

A multi-vessel test unit (MVTU) was used for the testing at NWR-WTP (schematic not shown). The MVTU contained three parallel pressure vessels, each capable of holding three spiral-wound membrane elements. It was configured so that the feedwater flow, to which the surrogates were dosed, was directed to two of the three vessels, with one vessel containing three ESPA1 RO elements and the other three TFCS NF elements. The MVTU was operated in a single-pass mode with no recirculation of either permeate or concentrate.

**Experimental procedures**

**Experimental matrix.** The DETU experiments at WF-21 and MVTU experiments at NWR-WTP employed elements in the following matrix of membrane and o-ring conditions:
i) intact membrane and intact o-rings; ii) compromised membrane and intact o-rings; and iii) intact membrane and compromised o-rings. Target flux and recovery in each DETU experiment was 10.4 gallons per square feet per day (gfd) and 15 percent, respectively. The low flux was selected as representative of full-scale RO/NF element operation on MFSE. Target flux and recovery in each MVTU experiment was 12 gfd and 45 percent, respectively. The MSTU experiments included testing of a single compromised ESPA1 RO element or o-ring installed at different locations within the vessel array of the MSTU. For the compromised membrane tests, the 17 remaining RO elements installed were not compromised. Target flux and target recovery in each MSTU experiment was 10.4 gfd and 75 percent. The impact of the location of the compromised membrane or o-ring on removals was also investigated in MVTU.

Each experiment included the dosing of microspheres, MS2 phage and R-WT in series with respect to time. The target feed concentrations for microspheres, MS2 phage and R-WT were 1 mg/L, $10^7$ plaque forming units/ml (pfu/ml) and 1 mg/L, respectively. The R-WT was dosed last to avoid the positive interference of the fluorescent dye molecules on the measurement of the fluorescent microspheres, particularly in permeate samples. The stock solutions of microsphere, MS2 phage and R-WT were prepared in distilled de-ionized water (DDW).

Membrane integrity compromisation. The integrity of the membrane elements was compromised by inducing a pinhole with a needle on the surface of the membrane. The tape wrap on the outside of the spiral-wound element was first removed and the membrane leaves and associated feed spacer were unrolled to access the membrane surface. The needle was then used to penetrate the membrane surface, taking care of perforating all membrane structure layers but without damaging the membrane on the facing side of the leaf. Following imperfection inducement, the leaves and feed spacer were rolled back into the original position and secured with tape wrap.

O-Ring integrity compromisation. O-rings, installed in the adaptor used to connect the membrane element to the end cap of the head of the vessel, were compromised either by cracking their inner surface or by cutting and removing 1, 2, or 4 mm sections.

Membrane fouling and cleaning. Compromised ESPA1 RO and TFCS NF elements were fouled in both the DETU and MVTU by continuously operating for prolonged periods (up to 2 weeks). For DETU, the elements were operated with MFSE at 15.8 gfd with 15% recovery for 10 days when the feed pressure increased to 1.3 times the initial pressure at constant permeate flux. The factor of 1.3 was chosen as the criterion typically used in full-scale treatment for chemical cleaning of membrane elements. The MVTU was operated with CTRW at flux rates as high as 16 gfd for 13 days when feed pressure increased to 1.15 times the initial pressure. Once the membrane-compromised elements were fouled, the experiments designed to investigate the impact of membrane fouling on surrogate removal were conducted. After completion of the compromised/fouled membrane experiments, the fouled elements were cleaned. At WF-21, a solution of sodium tripolyphosphate and sodium dodecylbenzene sulfonate at pH 11 was used for cleaning. A similar cleaning approach was employed at the NWR-WTP except that a commercially available high pH cleaning solution (Ecolab, Inc., St. Paul, MN) was used as the cleaning agent. Detailed information regarding fouling and cleaning procedures can be found elsewhere (Lozier et al., 2002).

Analytical methods

R-WT concentrations in the samples were measured on-site using a TD-700 laboratory
fluorometer (Turner Designs, Sunnyvale, CA). An excitation wavelength of 550 nm and an emission wavelength of 570–700 nm were used. The minimum quantification level for the NF/RO permeate was 20–25 ng/L. Microsphere concentrations were determined with a Bowman Series 2 Luminescence Spectrometer (SLM Aminco, Rochester, NY). An excitation wavelength of 533 ± 4 nm or 533 ± 8 nm (depending on concentration) and an emission wavelength of 566 ± 8 nm were used to analyze all microsphere samples. The minimum quantification level for this method was about 50 ng/L. MS2 (ATCC, 15597-B1) was prepared in high concentration stocks (>10^{11} pfu/ml) using *E. coli* (ATCC, 15597) as a host. The MS2 concentration in the samples was determined by a plaque assay procedure. For the DETU, MSTU and MVTU experiments, conductivity, total dissolved solids (TDS), pH, and temperature of the samples were measured onsite during each experiment using a single meter that had separate probes for conductivity/TDS, pH and temperature (Ultrameter 6P, Myron L company, Carlsbad, CA). The meter was calibrated for pH using a pH 7 standard buffer solution, and for conductivity/TDS using a 0.01 M KCl standard solution. More detailed information regarding all analytical techniques employed in this research can be found elsewhere (Lozier et al., 2002).

**Results and discussion**

**ESPA1 RO membrane testing with DETU and MVTU**

Figure 3 shows the log R-WT removals observed as a function of operating time with ESPA1 RO elements in the DETU with integrity compromised by various means, and at different degrees of membrane fouling. For the intact membranes and o-rings, 4-log removal was achieved at the target feed concentration of 1 mg/L. R-WT removals generally decreased over the 45-min sampling period, although only slightly for the DETU, while more significantly with the MVTU which ranged from 3.5 to 5.3 logs (data not shown). This decreasing trend was more pronounced when the initial removals were relatively higher (>4 logs). It is hypothesized that the initial higher removal was due to adsorption of the R-WT molecule into the membrane structure, and the subsequent decrease in rejection resulted from gradual saturation of the adsorption capacity of the membrane. Despite the differences observed, the removal of R-WT by the ESPA1 RO membrane was similar for the two feed waters tested. In contrast with the observation for R-WT, the removal
observed for MS2 with both DETU and MVTU did not vary much with operating time for the 45 min duration of the tests (Figure 4 for DETU).

Figure 4 shows the log removals of MS2 obtained from DETU experiments. Almost complete removal was achieved (>log 6.8) with the intact ESPA1 RO membrane in DETU, consistent with bench-scale testing results (Colvin et al., 2000). In contrast, the removals achieved in MVTU experiments with the intact element were in the order of approximately 5 logs (data not shown).

The removal of microspheres observed for the intact ESPA1 RO elements was at least 4 logs with both DETU and MVTU (data not shown). The microsphere removals observed with this membrane in the bench-scale experiments were at least 5–6 logs (Colvin et al., 2000), when performing experiments at the higher feed concentration of 10 mg/L. Unfortunately such high concentration could not be used in the pilot scale tests because of budget limitations.

**Effect of membrane integrity compromisation.** Compromising the integrity of the ESPA1 RO element with a pinhole resulted in a significant decrease in the removal of R-WT, from 4 to 2.2 logs with both DETU (Figure 3) and MVTU (data not shown). The similar decrease in the removal levels obtained with both units suggested that the sizes of the induced pinholes were similar. It is noteworthy that the removal efficiency increased significantly when the compromised ESPA1 element was switched from the lead position (~2.2 log removal) to the trailing position (~4.3 logs initially, declining to ~3.8 logs after 45 minutes) in the MVTU experiments. In the trailing position, the element would be subjected to a higher concentration of R-WT (~1.5 times feed concentration versus ~1.1 times for lead position), but a lower pressure differential across the pinhole.

Compromising the ESPA1 element with a pinhole also significantly decreased the log removal of MS2 from 6.8 to 2.9 logs in DETU (Figure 4). The MS2 removal in MVTU decreased from 5.3 to 2.3 logs when the compromised membrane element was placed in the lead position and from 5.3 to 4.2 logs in the trailing position. Degrees of reduction were comparable to those observed for R-WT. Microsphere removal decreased from at least 4.0 logs to about 2.2–2.3 logs due to membrane compromisation in both DETU and MVTU.
experiments with lesser decrease observed for the compromised element in the trailing position compared to the lead position in MVTU. These results are also in good agreement with the results for R-WT.

The similar removal observed for the various surrogates due to membrane integrity compromisation suggested that the pinholes induced on the ESPA1 RO elements allowed passage relatively independent of surrogate type and physicochemical characteristics. This further suggested that the passages of enteric viruses through the membrane imperfections (i.e. a pinhole) may be predicted by biological and non-biological surrogates as used in this study, as long as the size of imperfection is large enough not to cause a steric hindrance to passage of surrogates. This is an important finding, in that a non-particulate surrogate such as R-WT, with its advantages of low cost and ease of use, could be used as a surrogate for particles such as viruses.

Effect of membrane fouling. For both DETU and MVTU experiments, fouling of the compromised ESPA1 RO element resulted in significantly improved R-WT removals, reaching values even somewhat higher than those obtained with the intact membrane (Figure 3). Fouling of the compromised ESPA1 element also resulted in enhanced MS2 (Figure 4), and microsphere (data not shown) removals for both DETU, and MVTU experiments (data not shown).

This finding suggests that fouling the elements may effectively clog the imperfections by possibly a combination of pinhole filling and cake layer formation. Consistent with this hypothesis, chemically cleaning the compromised and fouled ESPA1 elements resulted in lower removals. However, the level of reduction in log removals was different for the DETU and MVTU. Cleaning the element in DETU decreased the log removals back to levels comparable for those observed for the compromised element, while the cleaning was less effective with MVTU. This discrepancy could be indicative of differences in the characteristics of the fouling layers, which in turn could be the result of differences in the quality of the different feed waters used.

Effect of o-ring compromisation. Cracking of the o-ring did not result in measurable increase of the passage of R-WT, MS2 or microspheres in DETU experiments (Figure 3 for R-WT). Apparently, the o-ring crack was completely sealed when pressurized inside its groove. On the other hand, removing sections from the o-ring did result in decreased log removals in both DETU and MVTU tests. As expected, the larger the cut, the greater the reduction in removal. For the DETU experiment with the o-ring with a 4-mm section removed, the removals were in the order of 0.6 logs, indicating that more than 10% of the overall permeate flow of 0.65 gpm (~0.07 gpm) was derived from the passage of feedwater across the o-ring gap. The o-ring with the 2-mm removed section, providing intermediate levels of R-WT rejections, was used for additional experiments conducted with the MSTU and MVTU. Interestingly, removing a 1-mm section from the o-ring resulted in decreased R-WT removal (Figure 3), but it did not affect the rejection of MS2 (Figure 4). Apparently, the smaller dye molecules were able to penetrate the partially compromised o-ring, but the orifice was sufficiently small to reject particles of the size of MS2. For MVTU tests, the 2-mm cut o-ring in the lead adaptor position significantly decreased the removal of microspheres. As was observed for R-WT, locating the cut o-ring in the trailing position had a much lower impact on removal (from 4.4 to 3.8 logs) compared to the lead position (to 2.7 logs). These results suggest that the degree of physical compromisation to o-rings must be significant to impact microbial integrity.

TFCS NF membrane testing with DETU and MVTU. Figure 5 shows the removal of R-WT by TFCS NF membranes in DETU at different experimental conditions. For the intact
TFCS NF element, the removal ranged from 3 logs to 2.75 logs. For MVTU, removals ranged from 4 logs initially to 2.9 logs at 45 minutes (data not shown). As observed for the ESPA1 element, the log removals of R-WT decreased with time, consistent with transient dye adsorption. However, the reductions in removal efficiency with respect to time were more significant with the TFCS NF membrane than with ESPA1 RO membrane, suggesting that the TFCS NF membrane might adsorb more R-WT. This finding was further supported with a separate test in which R-WT concentrations in permeate of both ESPA1 RO and TFCS NF membranes were monitored for 24 hours after R-WT spiking was discontinued. R-WT was observed in the permeate from both elements during the entire 24-hour flushing period, but the R-WT mass released from the TFCS NF element was higher than that for the ESPA1 RO element.

In contrast to the ESPA1 RO elements, which provided 6.8 log (in DETU) and 5.4 log (in MVTU) removals of MS2, the TFCS NF elements in DETU showed incomplete MS2 removals ranging from 4.3 to 5.3 logs (Figure 6). Similarly, the TFCS NF element used in
MVTU removed only about 4–4.2 logs of MS2. Microsphere removals by the TFCS NF membrane in DETU were found to be 2.5–3.1 logs (data not shown), which were low compared to those observed with the intact ESPA1 RO element. Microsphere removals by the intact TFCS NF element in MVTU were at least 4 logs.

Effect of membrane integrity compromisation. Compromising the integrity of the TFCS NF membrane used in DETU resulted in a decrease in R-WT removal, from 3.0 to 1.5 logs (Figure 5). The results obtained for MVTU appeared to indicate that the degree of TFCS NF membrane integrity compromisation (induced by the pinhole) was not sufficient as supported by the R-WT removals being only slightly lower compared with those obtained with the corresponding intact TFCS element. In DETU, while the impact of membrane compromisation on microsphere rejection was similar to that observed with R-WT, the impact was somewhat greater for MS2. The removal of MS2 decreased to 1 log versus 1.5 logs for R-WT and spheres. Consistent with the results for R-WT, the rejection of MS2 was not affected by compromising the TFCS membrane in MVTU.

Effect of membrane fouling. Although fouling of the compromised TFCS NF elements resulted in increasing R-WT removals for both DETU and MVTU, the extent of increase was not as large as that observed during the corresponding tests with the ESPA1 RO membranes. Especially for DETU, where compromisation had the greatest impact, membrane fouling did not increase the R-WT log removals to levels observed for the intact element. The lack of removal efficiency recovery by membrane fouling could be attributed to a lesser amount of fouling on the TFCS NF elements compared to that on the ESPA1 RO elements. Digital pictures taken of the surface of both elements after fouling showed more abundance of fouling matter on the ESPA1 RO membrane than on the TFCS NF membrane. Similar removal trends were observed for the test performed with MS2 and microspheres for both DETU and MVTU. Chemically cleaning the compromised and fouled TFCS NF elements decreased the log removals back to the levels observed with the compromised TFCS NF element. Unlike the ESPA1 MVTU experiment, where cleaning did not reverse foulant impacts, it appeared that the lesser mass of fouling matter present on the TFCS NF membrane was more readily removed by chemical cleaning.

Effect of o-ring compromisation. Compromising the o-ring (1-mm cut) decreased the R-WT removal in DETU from about 4.5–5.0 to 2.5 logs. With the MVTU, the 2-mm cut rings caused slightly greater loss of R-WT removal when installed in the lead versus the trailing position.

Rejection of conductivity and TDS
Rejection of conductivity and TDS during R-WT injection was constant for all ESPA1 RO and TFCS NF experiments and was not different from that observed prior to R-WT dosing. Similar results were also found for MS2 phage and spheres, indicating that the presence of low concentrations of the surrogates did not impact on membrane inorganic ion rejection.

As would be expected, conductivity rejection was greater for the intact ESPA1 RO elements than for the corresponding TFCS NF elements under all experimental conditions. Rejections of 98.2–98.4% were achieved with the intact ESPA1 RO elements during the DETU testing of all three surrogates. Rejection by intact ESPA1 RO elements during MVTU tests was somewhat lower, between 95.7 and 96.6%. For the intact TFCS NF element used with DETU, conductivity rejections of 95.3–95.7% were measured. As with ESPA1 RO experimental results, lower rejections, 88.4–90.5%, were observed during
MVTU tests for the intact TFCS NF elements. Rejection was anticipated to be higher for DETU tests given that the ratio of divalent to monovalent ions in the MFSE was significantly higher than in the CTRW and that RO, and especially NF, membranes reject divalent ions at a greater rate than monovalent ions.

Intact ESPA1 RO and TFCS NF elements in DETU rejected 98.4–98.6% and 96.1–97.0% TDS, respectively, comparable to conductivity rejection. This degree of rejection was <2 logs and clearly illustrates that inorganic ion rejection, as measured by either conductivity or TDS, did not provide sufficient sensitivity to be used as an accurate measure for viral passage.

Both membrane and o-ring compromisation produced consistent decrease in both conductivity and TDS rejections, except for the case of the cracked o-ring. As observed with the other surrogates, the decrease in inorganic ion removal efficiency caused by membrane compromisation indicated that a substantive amount of dissolved salts passed through the pinhole via advective flow. Further, deposition of a fouling layer on the surface of the compromised membranes increased inorganic ion rejection, demonstrating that such fouling effectively plugged the pinhole to an extent that ion passage could be prevented. Cleaning of the compromised/fouled elements with standard membrane cleaners effectively removed the foulant layer and allowed increased passage of inorganic ions.

**MSTU experiments**

When MSTU was operated for 60 min without surrogate dosing to assess the integrity of each of the “intact” vessels, permeate conductivity and TDS values for these vessels were very consistent with the results of the QA/QC test conducted before the start of compromisation experiments, confirming integrity of the vessels. Log removals of the particle surrogates (MS2 and microspheres) in MSTU were slightly greater than for DETU while R-WT removals were lower.

The impact of location of compromised element or o-ring on the removal of surrogates in a multiple-stage RO system was also investigated through MSTU experiments. The potential impact of position of compromised membrane and/or o-ring within the train of membrane system on overall integrity can be related to: (1) change in net driving force \( (NDF) \) pressure; and (2) change in surrogate feed concentration along the different locations. \( NDF \) pressure is defined as:

\[
NDF = P_f - \Delta P_{fc}/2 - \Delta \Pi - P_p
\]

where:
- \( P_f \) = system feed pressure, psig
- \( \Delta P_{fc} \) = difference in pressure between feed side of first stage and concentrate side of last stage, psi
- \( \Delta \Pi \) = difference in osmotic pressure across the membrane or between feed/concentrate and permeate, psi
- \( P_p \) = permeate side pressure, psig

As concentrations of contaminants, both soluble and particulate, increase from feed inlet of first stage to outlet of final stage due to rejection by membrane, the imperfection at the trailing element would experience higher concentrations of contaminants and lower \( NDF \) pressure.

The impact of the location of compromised o-ring. Based on the results obtained from DETU experiments, a 2-mm cut from the o-ring was selected as a method of compromisa-
tion in the MSTU tests. This level of compromisation provided a medium level of surrogate passage through o-ring imperfection enabling a better assessment of the impact of compromised o-ring location on surrogate removal. The compromised o-ring was installed in the lead adapter of vessels 1, 3, 5 or 6 (one position per experiment). For any given experiment, all other o-rings and all membrane elements were intact. Figure 7 shows the impact of the location of the vessel containing the compromised o-ring on the overall R-WT log removal efficiency. In general, log removals of all surrogates for the combined permeate (data for MS2 and microsphere not shown) decreased as the location of the compromised o-ring was moved from lead to trail (i.e. from vessel 1 to 6). For example, when the compromised o-ring was installed in vessel 6, the log removals of R-WT, MS2 and microspheres were decreased from 3.7 to 2.2, 7.0 to 2.7, and 4.2 to 2.3, respectively. These results suggest that the impact of increasing surrogate concentration in the feed water from lead vessel to trailing vessel was greater than that of decreasing NDF pressure. Further, the lower NDF pressure may have actually contributed to increased flow across the o-ring cut in the downstream vessels. This impact was observed in the DETU tests, for which the permeate conductivity increased with decreasing feed pressure for the same compromised o-ring (2-mm cut). Apparently, larger NDF pressures (pressure differential from feed water to permeate) provided more compression on the o-ring closing the gaps resulting from the removed section of the o-ring. Since the NDF pressure (and feed/permeate pressure differential) decreased in MSTU with vessel number, a negative impact on log removals would be anticipated with increasing vessel number. Thus, the combination of the two factors, decreasing NDF pressure (causing higher flow across the cut) and increasing feed concentrations, combined to produce a greater concentration of each surrogate in the combined permeate with increasing vessel number.

The impact of the location of compromised membranes. The element containing the compromised membrane was installed as the lead element in vessels 1, 3, 5 or 6 (one position per experiment). While the trends for the o-ring experimental results were consistent for all surrogates, significant differences were found depending on whether the surrogate is particulate or dissolved (data not shown). For MS2 and microspheres, the log removals for the combined permeate showed an increase with increasing vessel number (with an exception

![Figure 7](https://iwaponline.com/ws/article-pdf/3/5-6/81/419317/81.pdf)
for the MS2 data from the experiment performed with the compromised membrane placed in vessel 1). This trend suggested that, for the particle-based surrogates, the rate of passage of the particles through the membrane imperfection was controlled more by NDF pressure than by feed concentration. Unlike the imperfections in the o-ring, it is not expected that differences in NDF pressure would impact the size of the imperfection. However, it may be possible that a higher concentration of particles could more effectively occlude the imperfection, thereby reducing the rate of advective flow through the imperfection.

The removal of R-WT, TDS and conductivity for the combined permeate was essentially constant, again except for vessel 1. In contrast to the particles, the dissolved molecules would not be expected to cause occlusion and reduce advective flow.

Conclusions
This research suggested the importance of implementing proper methodology for membrane integrity testing in full-scale applications in order to comply with stringent regulations on viral removals in drinking water treatment. This research suggested the potential for using R-WT as a practical surrogate for detecting imperfections in both RO and NF membranes with respect to virus removal, as supported by accompanying experiments performed with other types of particulate surrogates, such as MS2 phage and microspheres. Fluorescent microspheres were also demonstrated to be a suitable viral surrogate. However, their cost, based on current manufacturing methods, is considered prohibitive for full-scale plant integrity testing. Discussions with microsphere suppliers suggest the real potential to reduce costs for such an application in the future.

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