Assessing pathogen removal efficiency of microfiltration by monitoring membrane integrity

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Abstract
This study was conducted to investigate the ability of various methods of monitoring membrane integrity to respond to changes in actual membrane integrity imposed by the compromised fibers within the microfiltration unit. In addition, the pilot-scale MF unit was challenged with high concentrations of coliform, Cryptosporidium, and spore, in order to assess the pathogen removal capability of microfiltration. A correlation between the integrity tests and microbial challenge data was also made. The integrity tests investigated in this study were pressure decay and diffusive air flow tests (direct integrity tests), and turbidity and particle counting (indirect integrity tests). Both pressure decay (PDT) and diffusive air flow (DAF) tests were sensitive enough to detect one damaged fiber out of 66,000. The extent of fouling did not affect the sensitivity of the PDT and DAF, showing that PDT and DAF tests are a simple, reliable means to monitor membrane integrity under field conditions. Indirect integrity monitoring using turbidity and particle counting, however, responded poorly to changes in membrane integrity. Microbial challenge study demonstrated that microfiltration was capable of removing various pathogens including Cryptosporidium, at the level required by drinking water regulations, under even adverse operating conditions. Finally, PDT and DAF tests showed a better correlation with actual microbial removal efficiency of microfiltration than turbidity and particle counting. The turbidity and particle counting grossly underestimated the removal of pathogen larger than MF membrane pore size due to poor sensitivity.

Keywords Cryptosporidium; coliform; membrane integrity; microfiltration; pathogen removal

Introduction
Microfiltration has the potential capability to allow water utilities to meet microbial removal requirements set by US EPA’s Surface Water Treatment Rule (SWTR), while avoiding high disinfectant doses (Yoo et al., 1995; Adham et al., 1996). However, small defects in the membrane system can potentially pass objectionably large quantities of pathogenic contaminants (Hong et al., 1999). In order to ensure safe and reliable treatment of surface waters with microfiltration, testing of the integrity of the membranes must be conducted. In addition, a correlation between the integrity test results and the microbial removal efficiency of the membrane must be established to utilize these integrity test results in a meaningful manner.

Broadly speaking, two general categories of integrity tests exist for low-pressure (MF and UF) membranes. These are direct and indirect integrity measurements. Direct testing methods are those which evaluate the integrity of the membrane itself, while indirect methods are those which measure a surrogate parameter, such as turbidity, in order to determine the condition of the membrane. The relative advantages and disadvantages to each method of integrity testing have been well documented by Adham et al. (1995).

The purpose of this research project is to develop the relationship between the microbial removal efficiency and the results of the integrity tests. First, a variety of membrane integrity tests were performed using both new and deliberately damaged membranes, as well as varying the state of fouling of the membrane. Next, the microbial removal efficiency of the microfiltration pilot plant was assessed by a series of microbial challenge experiments conducted at the same operating condition and damage state as the corresponding integrity tests. Cryptosporidium, coliform, and spores were used in these challenge experiments.
Finally, this project attempted to correlate the measured microbial removal efficiency to membrane integrity test results.

Methods and materials

Microfiltration pilot plant

This study was conducted using a pilot-scale microfiltration plant supplied by USF Memcor (Model 3M10C, USF Memcor Research Pty, Ltd., South Windsor, NSW, Australia). This unit has three modules, each containing about 22,000 individual fibers. The fibers are made of polypropylene, with a nominal pore size of 0.2 µm. Each module contains 15 m² of fiber surface area available for filtration. When operated at a flux of 175 L/h/m² (100 gfd), this unit produces about 7,875 L/h (34.7 gpm). This is at the high end of the unit’s rated flux; typical operating fluxes are roughly half this figure. This project conducted all trials at the high flux. The pilot plant was installed in the Tampa Water Department in Tampa, Florida. The source water used for this study was drawn from the Hillsborough River, located in west-central Florida. This is a surface water characterized as having high total organic carbon (TOC) and low turbidity (Reiss et al., 1999).

Membrane integrity monitoring

Two direct integrity monitoring methods, pressure decay and diffused air flow tests, were used to assess the integrity of the MF pilot unit. The pressure decay test (PDT) measures the loss of air pressure due to air flowing under pressure through any defects in the membrane. After pressurizing the permeate side of the membrane to a point just below the bubble point of the membrane by compressed air, the pressure decay rate was recorded. The diffusive air flow (DAF) test, on the other hand, measures the flow rate of liquid displaced by the air flowing under pressure through any defect in the membrane. The air pressure is held constant throughout the course of the test, in contrast to the PDT. In addition, turbidity and particle counting measurements were performed as means of indirect integrity monitoring. Turbidity of the samples was measured for both feed and filtrate samples using a Hach Ratio turbidimeter (Hach, Inc., Loveland, CO, USA). Particle counting was conducted by using a batch particle counter (HIAC Royco 7000, Pacific Instruments, Inc.).

Filtration experiments

In conducting this project, periodic visits to the Tampa Water Department were made. On the days prior to the actual challenges, experimental conditions were set up including a chemical cleaning in all cases, followed by cutting of membrane fibers and/or initiation of fouling conditions, if appropriate. If the unit is to be challenged at an unfouled state, the unit was left to run in normal filtration at roughly 2,269 L/h (10 gpm). This was done in order to restrict biological growth on the membranes. On the day of the challenges, DAF and then PDT tests were conducted according to their respective procedures. Next, the first challenge was conducted as detailed below. Then, the intervening DAF and PDT tests were conducted, followed by the second challenge experiment. After that, the post-challenge DAF and PDT measurements were performed. Thus, for each challenge run, three sets of integrity tests (DAF and PDT) were taken, and two challenge experiments were conducted. Lastly, the CMF unit was restored to its original configuration by conducting a chemical cleaning, and then repairing any fibers that were cut, if applicable.

Microbial challenges

The CMF unit flux was first adjusted to approximately 175 L/h/m² (approximately 34.7 gpm). A backwash was conducted, followed by five minutes of filtration run time in order for the unit to stabilize. Next, a rewet was conducted, with a further two minutes of
filtration run time. A rewet consists of pressurizing the filtrate side of the unit to drive out air trapped within the pore matrix. The pre-challenge membrane integrity tests (DAF and PDT) were conducted according to their respective procedures. The CMF unit was then put into feed side recirculation mode. This directed the flow through the feed side back to the break tank without producing filtrate.

Next, the break tank was filled to between 270 to 290 L, and the raw water feed supply valve was shut. Feed water blank (unspiked) samples were drawn from the sample connection on the raw water supply piping leading to the CMF unit at this time. The challenge spike solutions were added while the CMF unit was in recirculation mode, and allowed to recirculate for ten minutes in order to homogenize the feed. Feed water grab samples were drawn from the break tank. The unit was then switched from recirculation mode to direct filtration mode. This immediately produced filtrate, which was directed out the sample hose. Filtrate samples were collected at this point. The CMF unit continued to produce filtrate until the break tank reached the low level alarm point and the unit shut down.

After the first challenge was completed, the intervening integrity tests were conducted. All of the backwash and stabilization run times were also duplicated as described at the beginning of the protocol. The second challenge experiment was then conducted by repeating the process once again followed by the post-challenge integrity tests. At the conclusion of the challenge experiment and integrity testing cycles, chemical cleaning and fiber repairs were done as needed.

Microbial preparation, spiking and analysis

Coliform. Preparation, spiking and analysis of the *Escherichia coli* (ATCC 25922) spiking solutions was conducted according to the procedures detailed in *Standard Methods for the Examination of Water and Wastewater* (1995) membrane filter technique (section 9222).

Cryptosporidium. Preparation, spiking and analysis of *Cryptosporidium* were conducted by Dr Joan Rose at the University of South Florida. First, dead *Cryptosporidium* oocysts were purchased from Pleasant Hill Farms (Troy, Idaho) at a concentration of $1 \times 10^8$ oocysts/L. Three time averaged 500-mL grab samples were taken on the feed during the conduct of the challenge (beginning, middle, and end), while the filtrate was directed into nine 20 L sterile carboys. Isolation and enumeration of the feed grab samples were accomplished using centrifugation to concentrate the samples, which were then enumerated using Immunofluorescence microscopy with monoclonal antibodies (IFA) specific to the oocyst cell wall. For the filtrate samples, isolation and enumeration of *Cryptosporidium* oocysts utilized vortex flow filtration (VFF). VFF allowed large volumes of water (50–100 L) to be concentrated into approximately 40–50 mL. The concentrate resulting from the VFF was enumerated using the method identical to the feed samples.

Spores. Preparation, spiking and analysis of *Bacillus subtilis* were conducted by the TWD Technical Services Laboratory. Feed and filtrate samples collected were heat shocked and diluted in sterile 0.85% saline to the point necessary to produce 10 to 300 CFU/100 mL by membrane filtration on nutrient agar. Three dilutions were used to bracket the above results each time.

Results and discussion

Membrane integrity monitoring

Direct integrity tests. Figure 1 shows the measured pressure decay rate against the number of cut fibers (out of 66,000 total fibers) and transmembrane pressure (i.e. fouling state). The pressure decay rate was recorded in units of kilopascals per minute (kPa/min). Two things are of note with respect to this chart. First, it is apparent that the pressure decay rate clearly
increased with an increasing level of membrane damage. When no fibers were cut, decay rates of less than 0.5 kPa/min were consistently measured. When one fiber was cut, the decay rate increased to about 1–2 kPa/min. When three fibers were cut, the decay rate increased further to about 4 kPa/min, although more “noise” was evident here as well. The second point of this graph is to demonstrate the relative unimportance of the TMP on the pressure decay rate, indicating membrane fouling did not diminish the sensitivity of PDT to detect membrane defects.

Similar to the PDT results, the key parameter of the DAF test, the flow rate in millilitres per second, is plotted versus the number of cut fibers and fouling states in Figure 2. Note that for clarity, the vertical axis is plotted with a logarithmic scale. Figure 2 demonstrates a clear increase in diffusive air flow rate as the number of cut fibers increased. Specifically, the undamaged membrane flow rate was 0.1–0.01 mL/sec. When one fiber was cut, this flow rate was seen to increase to roughly 1 mL/sec. When three fibers were cut, typical flow rates on the order of 10 mL/sec were measured, showing that the DAF test is very sensitive in detecting small changes in membrane integrity. Note also that in Figure 2, no discernable trend was evident with respect to transmembrane pressure.

Indirect integrity tests. The source water for this project is characterized as having a low mean turbidity of about 2.25 NTU (Reiss et al., 1999). Even when spiked with challenge microbes, the spiked feed turbidities were between 3 and 10 NTU, as seen in Figure 3. Both the feed and filtrate turbidities are differentiated on the basis of fouling state, and are displayed with the y-axis (turbidity) as a logarithmic scale in Figure 3. Filtrate turbidities did not show any significant change when fibers were cut, implying that turbidity is a poor indicator of membrane integrity.

Figure 4 illustrates the spiked feed and filtrate particle counts measured. The feed water concentrations were variable, ranging from 2,200 counts/mL to greater than 40,000 counts/
mL. The filtrate particle counts were similarly variable, ranging from 35–1,700 counts/mL. As with the turbidity results, the salient point of Figure 4 is the lack of any discernable change in permeate concentration with increasing levels of fiber damage.

Comparison of integrity tests with microbial challenges

Microbial challenges were conducted on the MF pilot unit under a variety of transmembrane pressures and levels of damage. The results were shown in log reduction value (LRV). The MF pilot unit showed good rejections (i.e. more than 4 LRV) of Cryptosporidium, coliform, and spores, even under adverse operating conditions, which were high operating flux, high transmembrane pressure, and damaged membranes. The challenge results were compared with those of integrity tests in the following subsections.

Direct integrity tests. Figures 5 and 6 show the challenge LRVs of coliform, Cryptosporidium, and spores versus the corresponding PDT and DAF LRVs, respectively. An ideal 1:1 relationship is defined by the dotted 45° line. It is shown that the measured microbial challenge LRVs were qualitatively related to those of PDT and DAF tests. However, the resulting statistical correlation based on linear regression performed using a commercially available statistics and graphic software package (Microcal Origin™) was rather loose and poor. The coefficient of correlation ($r$) ranged from 0.48 to 0.67. The weak correlation is probably attributable to the scattered microbial challenge data due to inherent difficulties involving microbial analysis. Limited data available also contributed the poor correlation. Despite statistical limitation, the results demonstrated that the PDT and DAF provided more accurate assessment of actual microbial removal by microfiltration than indirect integrity monitoring methods, which are discussed in the following subsections.

Indirect integrity tests. This section represents the attempt to correlate the results of the microbial challenges with the results of the indirect integrity monitoring techniques. The indirect methods used were turbidity monitoring and batch particle counting. As can be expected, a comparison of the rather low removals of turbidity and particle counts with the generally higher logarithmic removals of the model microbes would yield poor results. Indeed, Figures 7 and 8 show just that. Figure 7 plots the turbidity LRVs versus the LRVs for the corresponding microbial challenges. One result is that whatever the actual microbial removal efficiency of the membrane was, the turbidity LRV was about 1–2 log. An accurate predictor of microbial removal would more closely follow the dotted line, which depicts an ideal 1:1 correlation in terms of LRV: LRV. This is attributed to poor sensitivity of turbidity measurements. Likewise, Figure 8 graphs the particle count LRVs versus the corresponding microbial challenge LRVs. The particle count log reductions were variable, but mostly fell within the 1–3 log range. Once again, this shows a lack of correlation between
the particle count LRVs and any of the corresponding microbial removals noted. The poor correlation mainly resulted from low feed particle concentrations.

Conclusions
• Microfiltration showed the ability to remove pathogens such as Cryptosporidium, in excess of the levels required by regulations, even under adverse operating conditions such as high operating flux and transmembrane pressure.
• PDT and DAF tests provided simple, reliable means to monitor membrane integrity under field conditions. Both tests responded well to the presence of even a single cut fiber out of 66,000. In addition, fouling extent marginally affected the sensitivity of the PDT and DAF results.
• Turbidity and particle counting responded poorly to changes in membrane integrity. Relying on either of these integrity monitoring methods may run the risk of overlooking membrane defects, allowing operation to continue when the integrity of the membrane unit is actually compromised.
• PDT and DAF tests showed better correlation with actual microbial removal efficiency of microfiltration than turbidity and particle counting. However, more data collection and refinement of microbial analysis are required for developing a more statistically reliable correlation.

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References