DEMOWARE

Deliverable D2.1

Best practice and trends in membrane integrity monitoring Literature Review

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Abstract	This report is a provides an overview of the present state of membrane integrity testing covering long-known well established procedures as well as new approaches in the stage of research and development. A short description of the different technologies, their characteristics and limitations is given as well as a comparison of important parameters.

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Abbreviations

AIM	Acoustic integrity monitoring
ASV	Anodic stripping voltammetry
CMF	Continuous micro-filtration (Memcor®)
DAF	Diffusive airflow test
DI	Deionized water
FIU	Fluorescence intensity units
ICP-OES	Inductively coupled plasma atomic emission spectroscopy
LED	Light-emitting diode
LIBD	Laser-induced breakdown-detection
MF	Micro-filtration
MI	Membrane integrity
MSP	Magnetically susceptible particles
MWCO	Molecular weight cut-off
NDR	Normalized diffusion rate
NF	Nano-filtration
n. a.	Not applicable
n. s.	Not specified
NTU	Nephelometric Turbidity Unit
PAC	Powdered activated carbon
PDT	Pressure decay test
PFU	Plaqueforming unit
RO	Reverse Osmosis
RT-PCR	Real Time-Polymerase Chain Reaction
SIM™	Spiked integrity monitoring
UF	Ultra-filtration
VHT	Vacuum hold test

Executive Summary

Currently, the standard membrane test in the water treatment industry for the detection of small defects is the pressure decay test (PDT). Particle counting and turbidity monitoring are used to meet regulatory requirements and to detect larger defects. Direct integrity monitoring methods like the PDT are reliable and sensitive, but are time-intensive and, generally, non-continuous. They need to be performed off-line, which requires an interruption of normal operation and, in some cases, a draining of the vessels (Farahbakhsh, et al., 2003). In most cases, weekly tests are required to guarantee a log removal value (LRV) of 4.5 (Pearce, 2007).

Indirect tests like particle counting and turbidity monitoring can be performed continuously and online, but do not show the required sensitivity for micro-filtration (MF) and ultra-filtration (UF) (Troester et al., 2014). They are influenced by the operating conditions and, in some cases possibly air bubbles are leading to false-positive results (Farahbakhsh et al., 2003).

Current integrity tests are not sensitive enough to detect nanometric-size breaches (Gitis et al., 2006). Characteristics of an ideal integrity monitoring technique for low-pressure membranes have been described as low cost, simple, on-line, continuous, reliable and highly sensitive to detect membrane breaches (Guo et al., 2010) as well as applicable to a wide range of membrane configurations, feed water characteristics and operating conditions (Farahbakhsh et al., 2003). Much research has been conducted to this intent. Today, challenge testing with MS2 bacteriophage is considered the best process indicator for enteric virus removal (Antony et al., 2012).

The following review gives an overview of the present state of membrane integrity testing covering long-known well established procedures as well as new approaches in the stage of research and development. A short description of the different technologies, their characteristics and limitations is given as well as a comparison of important parameters.

Many of the methods date back to the 1990's, when a number of patents were issues for the various approaches. They have been designed and developed for membranes in drinking water applications.

New sensitive and applicable on-line approaches are rare and focus on validation of virus or Cryptosporidium retention by membranes, which is an issue when in drinking water treatment. Quite some approaches deal with mimicking virus by nano-scale particles. The rather clean waters treated there also allows for dosing of particles, whilst this might be less effective for water already containing higher amounts of natural particles.

Such investigations are rarer for water reuse schemes, where low pressure membrane integrity is not such a prominent issue. However, some studies aim to relate findings from challenge tests with direct integrity measurements in water reclamation plants. Operators of membrane systems are familiar with applying standard integrity tests such as pressure decay test and vacuum hold test. Such systems and procedures are often developed and delivered by the membrane system providers to directly monitor the membranes as such.

When it comes to indirect methods, we found that much research and development work is dedicated to improve detection of any kind of particle, be it naturally occurring ones or spiked ones. Challenge tests refinements look for improved challenge particles and detection methods that are little affected by background noise. Among those were magnetically susceptible particles, gold, silver or fluorescently labelled (nano)particles (see chapter 2.5.4). Whilst many of these methods are potentially suitable to validate the retention capacity of a membrane system, their continuous online application to monitor membrane integrity still needs to be developed and proven.

This stage has already been achieved for the membrane integrity sensor (see chapter 0). This method does not spike particles but observes a membrane fouling indicator, i.e. changes in pressure differentials caused by increased number of particles passing through an impaired membrane.

1 Introduction

Porous membranes such as microfiltration and ultrafiltration membranes, have become established processes in treatment trains for both water supply and wastewater treatment (Phattaranawik, Fane, & Wong, 2008). Their capacity to retain particles down to the submicron range qualifies them as a reliable barrier for most microbial contamination. In drinking water treatment, they allow for meeting stricter water treatment regulations, limiting the demand of chemical additions in treatment steps and achieving independence of raw water quality. Even in times of strong precipitation impact on the source water quality, 6 log removal of bacteria and cysts can be guaranteed. Due to its tighter particle size cut-off, ultrafiltration additionally reaches the complete removal of viruses (Panglisch, Deinert, Dautzenberg, Kiepke, & Gimbel, 1998).

In wastewater treatment conventional activated sludge systems with subsequent sedimentation can be replaced by membrane bioreactors which separate the biomass by a membrane. That one can be submerged in the biological tank or installed in side-stream in a different tank. MBR effluent followed by disinfection is deemed suitable for many agricultural water reuse application (US, Title 22 and others).

For more advanced applications in water reuse systems, micro- and ultrafiltration can be used as pre-treatment to reduce fouling and damage of downstream nanofiltration (NF) and reverse osmosis (RO) membranes.

The required filtrate quality, though, can be guaranteed only in case of fully integer membrane systems. Membrane integrity can be compromised by various defects that either concern the whole module or the membrane only (Childress et al., 2005). Whilst module leakage may result from defects in connecting elements damages of the membrane are often caused by:

- chemical attack (e.g. oxidation),
- faulty installation and maintenance,
- stress and strain during operation (e.g. backwashing) and
- damage by sharp objects that have not been removed in the pre-treatment (Phattaranawik, Fane, & Wong, 2008).

In hollow fibre membranes, leakages are caused by bursts in the fibres. In spiral-wound modules, pin-hole leakages occur as well as defects at the glue lines. Additionally, leakage can occur at connection elements like O-rings (Nederlof et al., 1997; Antony et al., 2012).

Membrane failure leads to a drop in removal efficiency and a possible contamination of downstream water quality (Phattaranawik et al., 2008). Therefore, effective, continuous and reliable membrane integrity testing is of utmost importance.

1.1 Membrane performance requirements

The need to observe membrane integrity can be motivated differently. There are legal requirements, particularly in drinking water treatment where the focus is on hygiene and where intactness assures the safe removal of pathogens. As an operational requirement, the monitoring of membrane integrity assures proper pre-treatment for downstream processes (e.g. nanofiltration and reverse osmosis steps) and minimise scaling, fouling or other damage.

Where (ultrafiltration) membranes are included in drinking water treatment flow sheets to retain parasites such as *Cryptosporidium* and *Giardia* membrane integrity testing is often made mandatory (DWI, 2002; US EPA 2001). Also in the US compliance with the Long Term 2 Enhanced Surface Water Treatment Rule, requires the continuous retention or removal of particles larger than 1 µm for drinking water production with

membrane filtration (Jackson, 2001). This corresponds to an absolute removal of *Cryptosporidium* and *Giardia* (Guo et al., 2010).

Recently, the German Technical and Scientific Association for Gas and Water (DVGW, 2013) has updated its technical rules for the use of membranes in drinking water production by specification on membrane integrity testing. One of the ambitions is to be able to detect 3 µm defects, which would allow parasites like Cryptosporidium or Giardia to pass.

The increased use of membrane stages in water reclamation schemes also increases interest in more standardised approaches to verify intactness. In principle, the same methods as in drinking water can be applied. However, the range of matrices treated by membranes is much broader. It includes their use in submerged membrane bioreactors, where they are in contact with organic rich mixed liquor suspended solids, or in post-treatment configuration where they receive settled secondary effluent.

Hygienic parameters are also regulated for water reuse applications where water quality standards are either defined as a limit value (e.g. a maximum allowable concentration of a parameter) or a minimum treatment efficiency. Especially the California Title 22 rules refer to ultrafiltration and microfiltration as valid technologies to produce 'filtered water' whose turbidity must not exceed 0.5 NTU at any time.

1.2 Characterising membrane retention capacities

1.2.1 Log Reduction/Retention Value LRV

The LRV gives a direct measure of membrane performance (US EPA, 2005). The pathogen removal efficiency of a membrane in the drinking water industry is often measured as the log removal value (LRV) giving the decimal logarithm of the ratio of the concentration of retained species in the feed to that in the permeate (Bennett, 2008):

$$LRV = log10\left(\frac{c_f}{c_p}\right) \qquad \text{(Equation 1)}$$

c_f: concentration of retained species in the feed

c_p: concentration of the species in the permeate

As is evident from equation 1, in particle-based methods the LRV for intact membranes will take high values when the feed concentration is high. It will also depend on the detection limit for a species (particle), as the permeate concentrations are supposedly low. In particle-based detection methods the sensitivity is also influenced by side-factors as e.g. contamination from the permeate side. For air-based tests both applied pressure and dead volume are influencing factors.

Some more detailed considerations about the calculation of a LRV in membrane applications is given in the Annex.

The sensitivity of a membrane integrity test is described as the maximum log removal value (LRV) that can be reliably verified by integrity tests for a given membrane filtration system. It must be greater than or equal to the required pathogen removal credit and is related to a particular particle size or particle size distribution (US EPA, 2005; Guo et al., 2010).

Required pathogen removal rates in drinking water as stated by the U.S. Environmental Protection Agency (2012) are given in Table 1.

Table 1 Inactivation or removal requirements for pathogens in drinking water (US EPA, 2012) and calculated LRVs

Pathogen	Percentage removal [%]	LRV
Cryptosporidium parvuum	99.00	2
Giardia lamblia	99.90	3
Viruses	99.99	4

2 Membrane integrity testing

2.1 Present state of integrity testing

Currently, the standard test in the membrane water treatment industry for the detection of small defects is the pressure decay test (PDT). Particle counting and turbidity monitoring are used to meet regulatory requirements and to detect larger defects. Direct integrity monitoring methods like the PDT are reliable and sensitive, but are time-intensive and, generally, non-continuous. They need to be performed off-line, which requires an interruption of normal operation and, in some cases, a draining of the vessels (Farahbakhsh, Adham, & Smith, 2003). In most cases, weekly tests are required to guarantee a log removal value (LRV) of 4.5 (Pearce, 2007).

Indirect tests like particle counting and turbidity monitoring can be performed continuously and online, but do not show the required sensitivity for micro-filtration (MF) and ultra-filtration (UF) (Troester et al., 2014). They are influenced by the operating conditions and, in some cases, air bubbles, possibly leading to false-positive results (Farahbakhsh et al., 2003).

Current integrity tests are not sensitive enough to detect nanometric size breaches (Gitis et al., 2006). Characteristics of an ideal integrity monitoring technique for low-pressure membranes have been described as low cost, simple, on-line, continuous, reliable and highly sensitive to detect membrane breaches (Guo et al., 2010) as well as applicable to a wide range of membrane configurations, feed water characteristics and operating conditions (Farahbakhsh, Adham, & Smith, 2003). Much research has been conducted to this intent. Today, challenge testing with MS2 bacteriophage is considered the best process indicator for enteric virus removal (Antony et al., 2012).

2.2 Methods overview

Membrane integrity tests can be broadly divided into direct and indirect testing methods and into on-line and off-line techniques (Krantz et al., 2011). Direct tests are performed directly on the membrane or the module, whereas indirect tests monitor permeate quality along different parameters.

Direct methods include air pressure tests, acoustic sensor test, liquid-liquid porosimetry test and binary gas integrity test. Indirect methods include particle counting and monitoring, turbidity monitoring, and surrogate challenge tests (Guo, Wyart, Perot, Nauleau, & Moulin, 2010).

A classification of various test approaches is illustrated in Figure 1.

On-line and off-line modes specify, if membrane integrity tests are performed during operation or if operation needs to be interrupted. All direct methods and microbial monitoring are performed off-line (Krantz et al., 2011).

The pressure decay test (PDT) and the diffusive airflow test (DAF) are most frequently used in drinking water treatment. They are simple to perform and reliable, require low maintenance and have high sensitivity. Among the indirect methods particle counting, turbidity monitoring and routine microbial analysis are most commonly applied (Crozes et al., 2002).

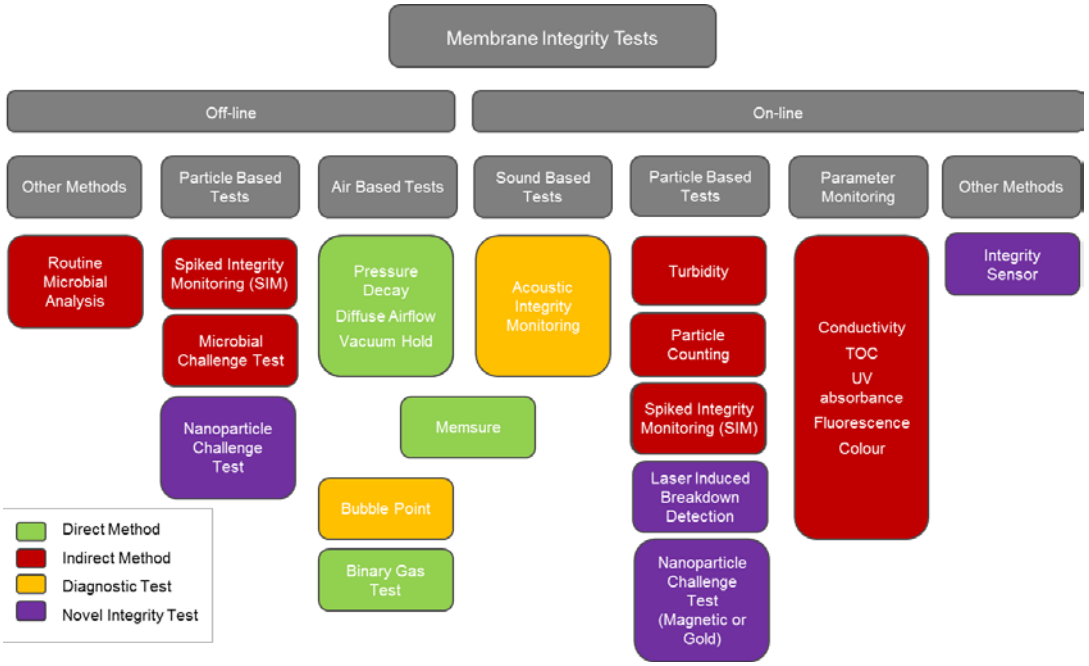


Figure 1 Overview and classification of different types of methods for membrane integrity testing

The main disadvantages of direct testing methods are that they are performed off-line, therefore requiring an interruption of operation, and that they do not measure the water quality of the filtrate. Depending on the testing schedule, in this way, operation might continue even after an impairment occurs (Guo et al., 2010). In contrast to this, Johnson (1997; 1998) states that the "shift in thinking from water quality monitoring to process control" presents an advantage for integrity monitoring. Because direct integrity testing is independent of filtered water quality, impairments may also be detected, if no or before changes in water quality are observed. Another disadvantage of direct methods is the fact that the minimum detectable pore size is limited by the required pressure to displace the water from the pores of a certain size. The pressure needed to detect defects below 2 – 3 µm is out of the operating range of most low-pressure membranes (Farahbakhsh et al., 2003). Indirect methods are performed continuously and on-line, but have low detection sensitivity. They do not meet the requirements for virus removal. In addition, they are often influenced by feed water quality, operating conditions, membrane operation mode, membrane surface area, inside fibre diameter and membrane fouling. Test results are also affected by instrument sensitivity and calibration and need correlation to effective microbial removal (Farahbakhsh et al., 2003, Guo et al., 2010).

Lately, new integrity testing methods have been developed that combine on-line operation with high sensitivity, accuracy, reliability and low costs. Implemented and newly developed test methods are described in this report.

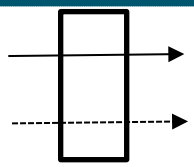
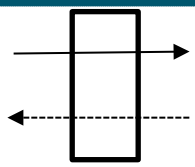
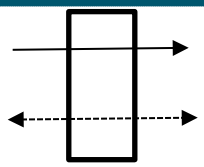
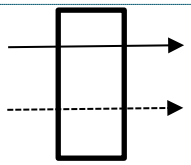
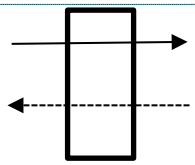
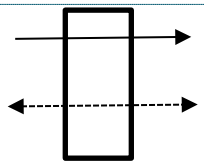
2.3 Air based tests

Air based tests rely on the bubble point principle . The bubble point of a membrane is the point at which the capillary forces in the largest pores are overcome and air flows freely through those pores (Farahbakhsh et al., 2003; Guo et al., 2010).

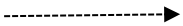
At transmembrane pressures below the bubble point, air passes through a wetted membrane only to a small extent by diffusion through liquid in the membrane pores. In case of a defect (leak), air will pass freely,

if the bubble point pressure of the leak is below the test pressure (Johnson, 1998; Farahbakhsh et al., 2003, Adams & Coté, 2005).

Table 2 Possible configurations for air-based integrity tests (Adams & Coté, 2005)

	Air on feed side	Air on permeate side	Air on both sides
Pressure driven			
Measurement	Pressure decay	Pressure decay / liquid displaced	Pressure decay
Advantages	Airflow direction equal to permeation direction	Usable with shell-less membranes	Same backpressure over the height of membrane modules
Disadvantages	Not usable with shell-less membranes	Airflow direction not equal to permeation direction	Airflow direction not equal to permeation direction / correction for air diffusion necessary
Vacuum driven			
Measurement	Vacuum decay / Air accumulated downstream / Liquid displaced	Vacuum decay	Vacuum decay
Advantages	Airflow direction equal to permeation direction Usable with shell-less (immersed) membranes		Same backpressure over the height of membrane modules
Disadvantages	Transmembrane pressure < 1 atm	Transmembrane pressure < 1 atm; not usable with shell-less membranes	Transmembrane pressure < 1 atm

 Normal water permeation direction

 Air leaking direction

Air-based tests allow quantification and location of defects. They are typically performed once per day requiring the unit to be taken off-line for about 10 -15 minutes. Measurement can be performed directly as pressure decay or indirectly as vacuum decay. In some configurations (see Table 2), it is possible to alternatively measure the liquid displaced (Adams & Coté, 2005). Possible configurations for air-based integrity tests and a description of their main properties are given in Table 2.

Pressure based tests are influenced by temperature, surface area, membrane fouling and other factors like starting test pressure, upstream volume, grade of wetting of membrane and the presence of other leaks in the system (Farahbakhsh et al., 2003). Pressure and vacuum tests are not applicable to flat sheet membrane modules (Krantz et al., 2011). Additionally, pressure based tests are not applicable to ceramic, RO and gravity feed configurations (Antony, et al., 2012).

2.3.1 Bubble point test

The bubble point test is a non-destructive diagnostic test to identify an impaired module and to locate the defect (Farahbakhsh et al., 2003, Guo et al., 2010b Antony et al., 2012).

To perform a bubble point test, the module has to be removed from the rack. The internal shell of the module is drained and pressurized and the membrane wetted uniformly. A dilute surfactant solution is applied to the open ends of the membrane fibers at the end of the module (Guo et al., 2010b). It is then checked, at which pressure a steady stream of bubbles is formed. If bubbles are formed below the bubble point of the membrane, a leak is indicated. A more accurate method for determining the bubble point pressure was described by Farahbahksh and Smith (2004), measuring the airflow rate downstream of a membrane at increasingly higher applied pressures. The resulting diffusion airflow curve is linear up to the point where the bubble point pressure is reached. This method minimizes human errors and the influence of the membrane surface area (Farahbakhsh et al., 2003; Phillips & DiLeo, 1996).

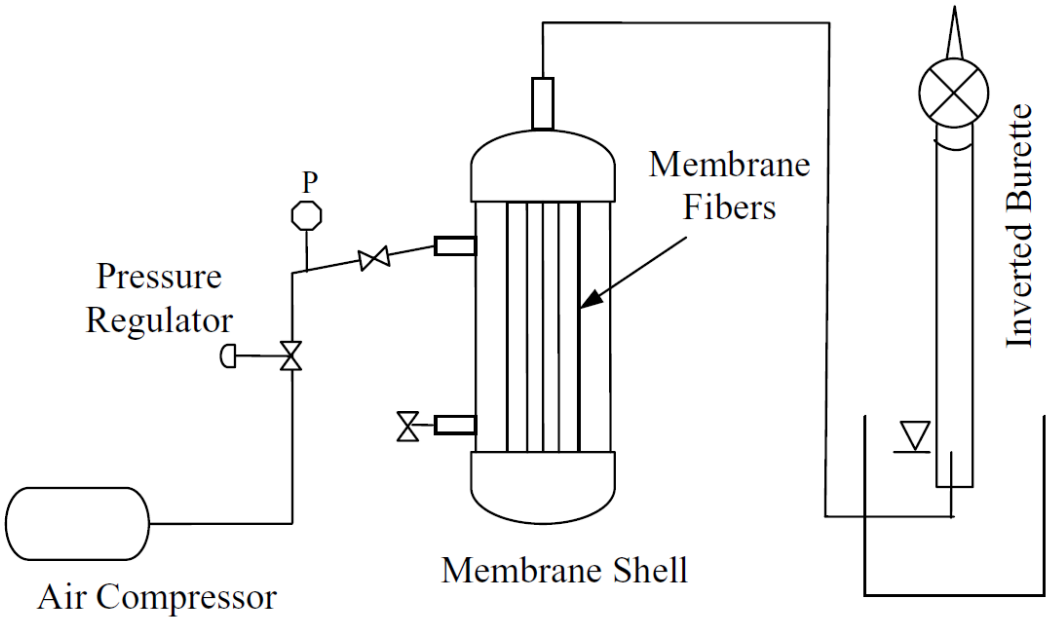


Figure 2 Schematic diagram for determining the bubble point (Farahbakhsh & Smith, 2004)

Bubble point testing is influenced by temperature. A change in water temperature from 20 to 5°C resulted in a drop in diffusive airflow rates of the test gas (nitrogen) of 10% (Hofmann, 1984). The actual bubble point can be masked in case of very small defects, because of difficulties to differentiate the gas flow from the small defects from the gas flow through the unimpaired membrane (Giglia & Krishnan, 2008). Especially in the case of large membrane surface areas (> 1'000 cm^2), the magnitude of diffusive airflow interferes with an accurate determination of the bubble point (Waibel, Jornitz, & Meltzer, 1996).

The bubble point test is presently used in practical operation for direct integrity monitoring and, as part of the Memcor© CMF process, reaches a sensitivity of 6 LRV to guarantee bacterial removal (Randles, 1996). It is not applicable to UF membranes, because the necessary pressure to detect a defect would be impractically high and could lead to membrane compaction and rupture (Guo et al., 2010b). The minimum detectable level is 2-3 µm (Gitis et al., 2006). The bubble point test can be applied to spiral wound and hollow fibre modules and is not applicable to flat sheet, ceramic, RO and gravity feed membrane systems (Antony et al., 2012).

2.3.2 Pressure decay test (PDT)

The pressure decay test investigates the ability of the membrane to hold a pressure against the ambient conditions. To perform a pressure decay test, both sides of a wetted membrane are drained and the pressurized side is isolated as illustrated in Figure 3. Compressed air at a pressure below the bubble point is applied to the membrane and the rate of pressure decay over the membrane is monitored for a specific period of time. The measured pressure decay is compared to the values of an intact membrane (Adams & Coté, 2005; Antony et al., 2012). A defect in the membrane is also indicated by a sharp drop in pressure. PDT tests for hollow fiber membranes are usually set to alarm against parameters based on an absolute size removal (> 4 µm) and a certain log removal value (4–5 LRV).Typical test pressure is 100 kPa during practical operation (20-200 kPa) while the test takes around 10 minutes (Guo et al., 2010b). A daily test frequency is recommended to ascertain a system integrity, which is consistent with the continuous sampling and analysis practice (Naismith, 2005, Guo et al., 2011;).

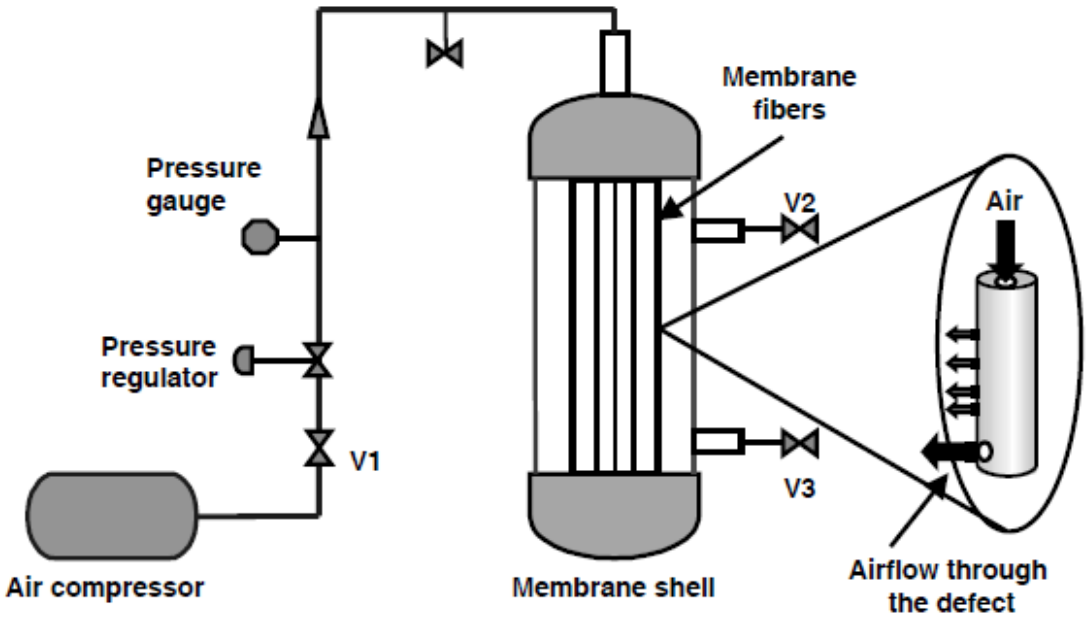


Figure 3 Schematic of a pressure decay system (Farahbakhsh, Adham, & Smith, 2003), V1, V2 and V3 are valves

Air diffusion through intact pores leads to a certain pressure decay. The contribution of air diffusion to pressure decay may produce false-negative results and therefore needs to be estimated and accounted for (Guo et al., 2010b). Especially in full scale plants, air diffusion leads to rapid pressure decay (Antony, Blackbeard, & Leslie, 2012). A way of consistently accounting for air diffusion irrespective of module configuration, surface area and internal hold-up volume has been described by Guibert and Colling (2011). Farahbakhsh and Smith (2004) developed a mathematical model to estimate the contribution of diffusive airflow to the PDT.

The change in pressure decay is directly related to system integrity. An increase in pressure decay by the factor 10 represents a change in LRV of 1 (Johnson, 1997).

The PDT is a very reliable and non-destructive integrity test, which is highly automated and a standard part of many UF and MF systems. It is independent of the filtered water quality and reaches LRV values of 4.5-5 for *Giardia* or *Cryptosporidium* removal (Guo et al, 2010b). Below 1.5 µm, the PDT has low sensitivity for

defects. The pressure needed to detect smaller breaches would be too strong and damage the membrane. The sensitivity can be increased by using citric acid, a common membrane cleaning chemical, which lowers the surface tension. When PDT is applied to a whole rack of membranes, the impact of a single compromised fibre is weakened (Antony, Blackbeard, & Leslie, 2012).

If air diffusion is accounted for, air-to-air configurations (Table 2) are advantageous to air-to-water configurations, because they provide the same backpressure over the entire height of the membrane module and therefore offer a constant leak resolution (Adams & Coté, 2005). Main disadvantage is the fact that it cannot be operated continuously and on-line. Test effectivity is affected by membrane surface area and fluid temperature. False-positive results can result in the case of not-fully wetted membranes (Farahbakhsh et al., 2003, Guo et al, 2010b).

The PDT can be applied to spiral wound and hollow fibre modules and is not well applicable to flat sheet, ceramic, RO and gravity feed membrane systems (Antony et al., 2012). With shell-less membranes it is usable only in water-to-air configuration and if the permeate side is drained (Table 2) (Adams & Coté, 2005).

2.3.3 Diffusive air flow test

Diffusive air flow tests (DAF) are similar to pressure decay tests, except that the airflow is measured instead of the pressure decay (Antony et al., 2012). For DAF the wetted membrane is pressurized with a pressure below the bubble point pressure and the shell side is isolated (V2 closed, V3 open in Figure 3). In contrast to the PDT, the air pressure is maintained. The airflow through the membrane is measured indirectly as displaced air or liquid (Johnson, 1997) and compared to the airflow of an intact membrane (Farahbakhsh, Adham, & Smith, 2003). DAF tests for hollow fiber membranes are usually set to alarm against parameters based on an absolute size removal (> 4 µm) and a certain log removal value (4–5 LRV) (Guo et al., 2010b). Test duration is 15 minutes (Guo et al., 2010b) and is usually performed once per day (Adams & Coté, 2005).

Diffusive airflow is reflecting the total porosity of the filter instead of the size of the largest pores as in the PDT, therefore not providing a direct connection to bacterial retention. To secure integrity test values, a multi-point diffusive airflow curve should be plotted for each membrane and product used. From this curve a correlation factor can be calculated between water and the product bubble point and a maximum allowable value derived. Separate diffusive airflow curves have to be established for pre- and post-filtration membranes (Meltzer, Madsen Jr., & Jornitz, 1999).

The DAF measuring displaced water flow was widely used in water treatment plants and provides easy and accurate measurements (Guo et al., 2010b). Because it is less influenced by air diffusion through the membrane and less sensitive to external air leaks on the lumen side, the DAF is more sensitive than the PDT (Johnson, 1997). It reaches LRV values of above 6. In tests with nitrogen as test gas diffusive flow rates dropped by 10% when water temperature changed from 20 to 5°C (Hofmann, 1984). The sensitivity of air diffusion rates to temperature changes, makes DAF measuring displaced liquid easier and more accurate (Guo et al., 2010b). Test sensitivity is also affected by not fully wetted membranes. In contrast to the PDT, the DAF requires additional pipe work to measure the displaced air or liquid flow rate (Antony, Blackbeard, & Leslie, 2012).

DAF integrity test can be applied to spiral wound and hollow fibre modules and is not applicable to flat sheet, ceramic, RO and gravity feed membrane systems (Antony, Blackbeard, & Leslie, 2012). With shell-less membranes it is usable in water-to-air configurations, if the permeate side is drained (Table 2) (Adams & Coté, 2005).

2.3.4 Vacuum decay test

Vacuum decay test (VDT) works similar to the pressure decay test, except that a vacuum is applied and the vacuum decay rate is measured. Likewise, the testing time is several minutes (Farahbakhsh et al., 2003, Guo et al., 2010b). ().

VDT is suitable for monitoring UF and MF membrane integrity, but is rarely applied in practical operation of membrane drinking water plants. It is useful as a screening procedure, but generally not intended for absolute verification of leaks (Guo et al., 2010b). Farahbakhsh et al. (2003) found VDT sensitive for detection of minor leaks in hollow-fibre UF membranes. Vacuum tests are more commonly used in the RO industry (Antony, Blackbeard, & Leslie, 2012; Farahbakhsh, Adham, & Smith, 2003) and have been standardized to monitor the integrity of RO and NF elements of FILMTEC membranes (Guo et al., 2010b).

According to Antony et al. (2012), the VDT is used to test the integrity of flat sheet membranes, whereas Krantz et al. (2011) state that pressure and vacuum tests are not applicable to flat sheet membrane modules. With VDT, systems can be tested that cannot be pressurized on the filtrate side like spiral-wound and shell-lees (immersed) membranes (Adams & Coté, 2005; Guo et al., 2010b).

2.3.5 Memsure ™

The Memsure™ integrity testing procedure is a method developed to monitor the Memcor™ CMF (continuous microfiltration) process. The Memcor CMF by Siemens consists of a 0.2 µm microfiltration system in side-stream configuration which is operated in outside-in filtration mode (Johnson, 1997).

Essentially, the Memsure™ process uses PDT or DAF (Memsure™ PDT and the Memsure™ DAF) for integrity monitoring and sonic integrity testing (Memcor® Sonic Analyzer) for identification of the impaired module. The sonic analyzer is a sensitive listening device that picks up the distinctive sound made by air leaking from a defect in the membrane and displays it as a sound level (Guo et al., 2010b). After identification, the defective module is isolated with the help of built-in isolation valves, removed and placed inside a housing. Air is applied externally to the fibre bundle to locate the leak. The fibre is sealed using a stainless steel pin (Johnson, 1998). The repair is done on site.

Memsure™ PDT and Memsure™ DAF test generally use a test pressure of 100 kPa. The test duration is about 5 minutes (Guo et al., 2010b). At Joyce Road Water Processing Plant, New Zealand (Antony, Blackbeard, & Leslie, 2012), and at Saratoga treatment plant, California (Randles, 1996), the test results have been correlated to the rejection of microbial challenge organisms.

The Memcor® CMF process guarantees a bacterial log reduction of 6 (Randles, 1996; Johnson, 1997; Johnson, 1998) and has been shown able to detect one broken fibre out of 1 Million (Johnson, 1998) and 12 Million (Randles, 1996) fibres. To ensure this sensitivity, an equivalent of 18 and 100 particle counters respectively would be needed (Johnson, 1998; Randles, 1996).

2.3.6 Binary gas integrity test

In alteration and adaptation of long established and straight forward flow related tests such as (pressure decay test and bubble point method) industry has developed slightly more sensitive tests to verify intactness especially of membranes for virus filtration devices.

The binary gas integrity test has been developed as an improved diffusion test showing a much higher sensitivity to defects in membranes than conventional gas-liquid diffusion tests (Giglia & Krishnan, 2008). It is applied routinely to assess integrity of virus clearance filters for the biopharmaceutical sector. For the time being its adaptation and application to full-scale membrane installations in the water sector is not

something to be expected. The provision of two specific gases in a contained environment seems to be prohibitive for routinely, full-scale application.

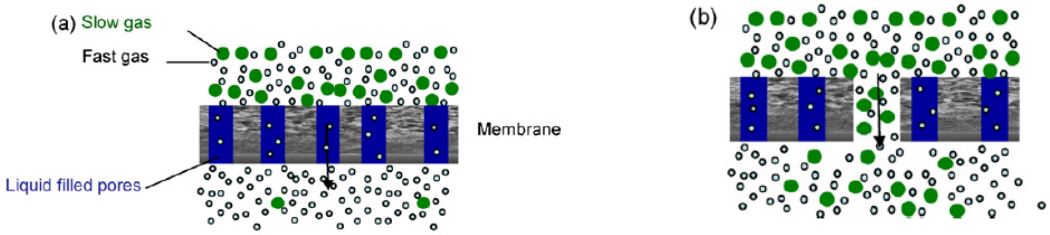


Figure 4 Binary gas diffusion through a wetted membrane: (a) integral membrane and (b) non-integral membrane (Giglia & Krishnan, 2008)

However, the principle is convincing: Two gases with different permeabilities permeate through the liquid layer of a wetted membrane. Downstream, the gas composition is measured and compared to the permeate composition of an intact membrane, which can be predicted based on the transport properties of the gases permeating through the liquid layer and the known operating conditions. An unexpectedly high amount of the slower permeating gas indicates a defect in the membrane as illustrated in Figure 4. Gas composition of feed and permeate gases can be measured by mass spectrometry or FTIR (Giglia & Krishnan, 2008). The test has a high sensitivity (LRV > 6) and is independent of membrane properties, but it needs to be performed off-line. A video illustrating its working principle can be found at: http://www.merckmillipore.com/CH/de/products/biopharmaceutical-manufacturing/downstream-processing/virus-safety/virus-filtration/702b.qB.6KkAAAFAU.BkiQpx,nav

2.3.7 Characteristics of air-based tests

Main disadvantages of air based tests in general are the fact that they are conducted off-line and the minimum detectable pore size. With the usually applied test pressures of 70 – 140 kPa, minimum detectable defects would be around 2 – 3 µm (Farahbakhsh, Adham, & Smith, 2003; Gitis, Haught, Clark, Gun, & Lev, 2006). Test pressures are needed to reach the bubble point of ultrafiltration membranes can exceed 500 psi (about 3.45 MPa), which is both impractical and potentially damaging to the membrane structure (Phillips & DiLeo, 1996). Still, according to Farahbakhsh et al. (2003), direct pressure-driven test are the most accurate tests available for determining the integrity of low-pressure membranes.

Table 3 Characteristics of air-based tests

Method	Mode	Characteristics	Source
Bubble-point test	off-line	LRV 6 in Memcor© CMF processes, detection limit 2-3 µm, not applicable to UF (too high pressure), non-destructive, diagnostic tool, temperature dependent, works well in combination with PDT, for spiral wound and hollow fibre modules	(Antony et al., 2012) (Gitis et al., 2006) (Guo et al., 2010b) (Hofmann, 1984) (Randles, 1996)
PDT Pressure decay test	off-line	LRV = 4.5-5 for Giardia or Cryptosporidium removal, detection limit 2-3 µm, duration 10 min., direct, very reliable, non-destructive, dependent on membrane area and fluid temperature, for spiral wound and hollow fibre modules	(Antony et al., 2012) (Farahbakhsh et al., 2003) (Gitis et al., 2006) (Guo et al, 2010b)
DAF Diffusive air flow test	off-line	LRV>6; non-continuous, easy and accurate, detection limit 2-3 µm, test duration 15 min., widely used in water treatment plants, sensitive to temperature, reflects total porosity of filter →needs validation, for spiral wound and hollow fibre modules	(Antony et al., 2012) (Gitis et al., 2006) (Guo et al., 2010b) (Meltzer et al., 1999)
VDT Vacuum decay test	off-line	for membranes that cannot be pressurized on filtrate side, duration several minutes, currently rarely applied for MF/UF, useful as screening procedure, for spiral wound and immersed membranes	(Adams & Coté, 2005) (Guo et al., 2010b)
Memsure	off-line	LRV = 6 for bacterial removal, for particles > 0.2 µm, duration 5 min., applied pressure 100 kPa, direct, non-destructive, simple, low maintenance, cost-effective, low energy consumption, low use of chemicals, extended life of filtration system, fully-automated, widely used in water treatment plants, not applicable to UF, for CMF systems (0.2 µm PP hollow fibre membranes)	(Guo et al., 2010b) (Johnson, 1997) (Johnson, 1998) (Randles, 1996)

2.4 Sonic sensing analysis

Another option to detect and characterise impaired membrane integrity is to assess the noise created by a membrane during filtration, or more precisely the changes of that noise as created by pressure fluctuations due to a defect (air bubbles passing through a defect). The method is typically applied to whole modules.

2.4.1 Sonic test

Sonic tests monitor the noise created by air bubbles that are passing through a defect and which characteristically changes the noise of a filtrating module in a specific frequency band.

A sound wave sensor, which is attached to headphones and a diode visual display, is manually placed at several locations on the membrane and the difference between intact and defective fibres identified by the operator. It is used as a diagnostic tool to detect the location of defective modules and fibres, which have been identified for example with help of the PDT (Antony et al., 2012).

Manually performed, sonic tests are subject to some degree of subjectivity connected to the interpretation of results by the operator (Laîné et al., 1998). Additionally, they may be affected by background noise and operation mode (Farahbakhsh, Adham, & Smith, 2003). Sonic test are not performed continuously and, in combination with PDT or DAF, they are usually performed offline. They do carry the potential of online and continuous monitoring, though, if the test is automated and computerized as done by Laîne et al. (1998) (Guo et al., 2010b) (see chapter 2.4.2).

At the Kenosha water treatment plant, sonic testing was performed once a month, if the PDT had shown pressure decay rates of 0.5 psi/min or more in two consecutive tests or if the pressure decay rate increased 0.05 psi/min or more compared to the rates of the previous shift. In these tests, sonic testing showed a sensitivity equal to that of the PDT (Landsness, 2001).

This approach has been integrated as diagnostic tool into the Memsure system (Johnson, 1998; Johnson, 1997). Sonic tests are applicable to MF and UF systems in side-stream configuration but cannot be used with submerged systems (Farahbakhsh et al., 2003).

2.4.2 Acoustic Integrity Monitoring

Acoustic Integrity Monitoring (AIM) is an on-line method developed and tested by Laîne et al. (1998) for membranes in side-stream configuration. It is based on hydrophonic sensor technology and continuously measures the noise created by a membrane during filtration.

A defective fibre is creating a distinctive noise signal which is caussed by pressure fluctuation and is detected in a certain frequency range (280–650 Hz). An AIM integrity monitoring system consists of several on-line sensors, which are mounted on each individual membrane module, of several collectors and a processor. One collector is able to treat 12 sensors. The processor compares the signals to a given threshold.

The hydrophonic sensor (Figure 5) works on the principle of piezo-electric ceramic bending. The ceramic is cast in silicon resin, which is in contact with a fluid. An electrical signal is given out, which is amplified and analysed in frequency modulation. In UF pilot scale tests in dead-end mode, one defect fibre caused an increase in acoustic levels of up to 20 dB. In recirculation mode, detection was not possible because of the noise generated by the recirculation pump (Laîné et al., 1998).

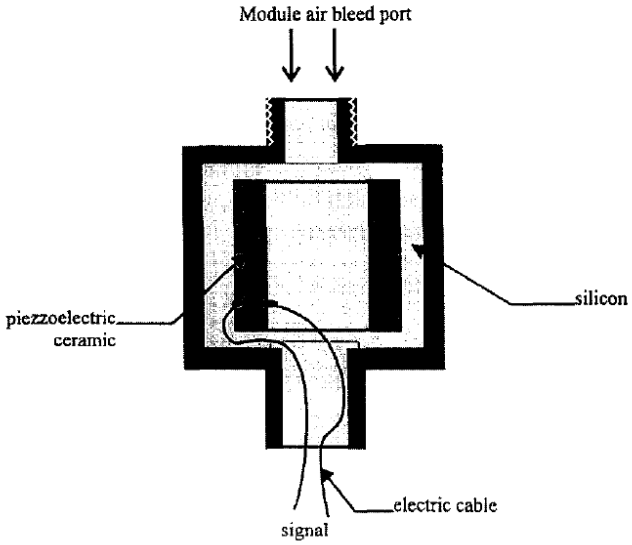


Figure 5 Schematic diagram of a hydrophonic sensor (Laîné et al., 1998)

AIM is an online and direct method, which is independent of feed water quality. A prototype consisting of 28 sensors, 2 collectors and 1 processor, used in dead-end mode, was able to detect a fibre hole of 0.5 mm in a membrane of approximately 771 m^2, which guaranteed a sensitivity of more than 6 LRV for viruses 100% of the time. However, sensitivity is strongly limited by flow rate and background noise caused by valves that control the permeate flow but can be mitigated using a variable frequency drive than using an actuated valve. Higher flow rates enable better acoustic detection since the noise generated by the defect is higher. Compared to particle counting, AIM is supposed to be economically competitive (Laîné et al., 1998). AIM can be applied in side-stream configurations to spiral wound and hollow fibre membranes in MF and UF systems (Antony et al., 2012).

Table 4 Characteristics of sonic sensing analysis

Method	Mode	Characteristics	Source
Sonic test	off-line	LRV 4.5 – 5 for single cut fibres, non-continuous, diagnostic, non-destructive, relatively simple, independent of feed water quality, results may be subjective, for MF and UF, not for submerged systems	(Antony et al., 2012) (Farahbakhsh et al., 2003) (Guo et al., 2010b) (Landsness, 2001)
Acoustic integrity monitoring	on-line	LRV > 6 for viruses 100% of time, continuous, direct, non-destructive, relatively simple, independent of feed water quality, depends significantly on background noise and flow rate, for MF and UF, for spiral wound and hollow fibre membranes	(Antony et al., 2012) (Guo et al., 2010b) (Laîné et al., 1998)

2.5 Particle-based tests

Among the indirect integrity monitoring methods, particle-based tests are a classic to characterise permeate quality. Yet the conclusiveness with respect to membrane damages is strongly depending on the size and type of particles under consideration. Those can be either constituents naturally present in the water or spiked particles.

There are several well-established methods to monitor particles in the above µm range. Currently, more sophisticated approaches are under development to push the detection limits towards the sub-micron range. The methods deliver qualitative or quantitative information to different extents, such as

- the number of particles
- the size distribution of particles
- the chemical characteristics of particles

and is dependent on the exciting wavelength and the light scattering properties of a particle.

2.5.1 Turbidity monitoring

2.5.1.1 Principle

Turbidity is the measure of relative clarity of a liquid. It describes the degree to which water loses its transparency due to the presence of suspended particulate matter. High turbidity means a greater part of light passing through a water sample is being scattered. The intensity of light scattered at 90 degrees (or different angles) is detected by a so-called nephelometer or turbidimeter and expressed as NTU, Nephelometric Turbidity Units (Lenntech, 2014). Turbidity monitoring belongs to the standard requirements for surface water treatment plants and water reclamation schemes (Farahbakhsh et al., 2003)

As intact membranes supposedly retain particulate matter in at least the size range of the pore size of the membrane, membrane defects will be associated with an increase in turbidity of the filtrate/permeate, caused by an increased flux of particles through the membrane. Whilst intact low-pressure membranes can reduce turbidity by more than 90% (depending on feed concentration), to around well below 0.1 NTU, a lower performance indicates a loss in membrane integrity (Guo et al., 2010b).

Turbidity is measured by conventional or laser turbidimeters (Farahbakhsh et al., 2003). A turbidimeter detects the intensity of light scattered at one or more angles to an incident light beam. The main difference between conventional and laser turbidimeters is the light source. Whereas conventional turbidimeters mainly use light-emitting diodes (LED), laser turbidimeters use a laser light source (US EPA, 2005).

2.5.1.2 Characterisation

Turbidity monitoring is an indirect method that is performed online and continuously. It can easily be applied to all membrane filtration configurations in micro- and ultrafiltration (Antony, Blackbeard, & Leslie, 2012). It is a standard in monitoring filtration steps (e.g. sand filter and final effluent quality in wastewater treatment).Though the detected scattered light signal is influenced by the number and sizes of the particle in suspension, it does not give any direct information on these properties (US EPA, 2005; Liu, 2012). Further, it is non-specific, so air entrapment in the filtrate and particle shedding in the plumbing can lead to false-positive results (Guo et al., 2010b (Farahbakhsh et al., 2003; Naismith, 2005).

A maximum LRV of 2 has been reached at a minimum detectable particle size of 1 µm (Liu, 2012). Especially at low particle concentrations and for defects smaller than 1 µm, turbidity monitoring shows low sensitivity. The Water Research Center (WRC) found no correlation between turbidity and particle count for particles between 2 and 5 µm (Hall & Croll, 1996). Other research showed that turbidity stayed constant at 0.45 NTU when particle (2-125 µm) count changed from 400 counts/ml to an average 2000 counts/ml, peaking at

over 4000 counts/ml (Chipps, M. J. et al., 1995). With conventional turbidimeters, particle breakthrough of a filter occurs hours before turbidity breakthrough.

Various investigations showed that turbidity measurement is not well suited for the detection of small membrane damages that actually lead to increased particles concentrations in the permeate (Panglisch et al, 1998; Antony et al., 2012). In a study reported by Liu (2012) the detected turbidity did not correspond to the degree of fibre breach. The experiment was performed on a microfiltration 50 module rack where different numbers of fibres were cut. The feed turbidity varied between 1 and 5 NTU, whilst the permeate quality exhibited mean turbidity values of 0.4 NTU. Crittenden et al. (2012) further exemplified these findings.

With nanometric scale (40 nm) particles, no signal could be detected with turbidity monitoring in an experiment that gave a clear signal with LIBD monitoring method (Troester et al., 2014). Laser turbidimeters have improved detection sensitivity by two orders of magnitude down to 0.001 NTU, but, in pilot scale, have still not reached the sensitivity of particle counters. They proved unable to detect small numbers of defect fibres (Guo et al., 2010b, (Farahbakhsh, Adham, & Smith, 2003)).

2.5.2 Particle counters

2.5.2.1 Principle

Particle counters work on the principal of light blockage or -light scattering. A sensor is used to detect, count and group signals of particles passing through a light beam. Depending on the light source, particles in the size range of 0.5 to 5 µm can be measured (Antony et al., 2012). Other than turbidity measurement, particle counters deliver quantitative results on water purity.

For particle counters with low thresholds, measures have to be taken in order to obtain reliable results. Adham et al. (1995) recommended flow control devices that provide a constant flow and an unchanged particle size distribution (e.g. through flocculation, settlement or contamination) through the particle sensor and a placement of the sensors as close to the sample source or flow controller as possible. Tubes should be made of inert material to prevent the adhesion of particles. Additionally, the sensors should be cleaned and the electronic background noise checked regularly.

Calibration of the sensors should be performed at least once a year (Guo et al., 2010b). To reduce the cost for particle counters, multisensory particle counters have been developed, consisting of 50 sensors, which share one light source, detector and control electronics.

Particle counters can detect LRVs of less than 4, which is mainly due to particle counter performance. To reach higher sensitivities, it is advisable to choose particle counters with a threshold as low as possible (Guo et al., 2010b). However, with consideration of the background concentration of contaminants under field conditions and the non-specificity of particle counting, a measuring range of 0.5–1 µm seems too sensitive to provide reliable data (Adham et al., 1995). Also, the costs for particle counters explode with lower thresholds (Guo et al., 2010b).

2.5.2.2 Application and testing

The method is deemed to reliably count particles larger than 1 µm (Liu, 2012) and may be applied as indirect continuous monitoring method as required by the US American legislation on drinking water production from surface water (according to the LT2ESWTR1 requirements - US EPA, 2005).

[1] Long Term 2 Enhanced Surface Water Treatment Rule. US legal requirement for drinking water quality when produced from surface water esp. aiming at reducing the risk of *Cryptosporidium* infection

The sensitivity of particle counting as a means of integrity monitoring increases with higher feed concentration and smaller membrane surface area. The dilution of permeate from an impaired membrane with permeate from intact membranes always causes a loss in sensitivity (Farahbakhsh et al., 2003; Antony et al., 2012).

Particle counting devices have been in assessed for their suitability to detect membrane damages by Glucina et al. already in 1997 in both pilot and full-scale plants for drinking water production. They tested them on microfiltration and ultrafilltration in different configuration and filtration modes. Experiments were conducted on UF and MF pilot plants (10 and 200 nm pore size respectively) and full-scale UF treatment plants (Aquasource, UF) in dead-end or cross-flow filtration. They found particle counters not sensitive enough in dead-end filtration, but sufficiently sensitive to detect one compromised fibre out of 40'000 in cross-flow filtration.

Also Panglisch et al. (1998) tested particle counters in an ultrafiltration membrane pilot plant (X-Flow UF membranes) on a drinking water facility. From their findings they identified a number of challenges to be overcome for application in full-scale plants, such as

- flow equalisation before the sensor/measuring point
- dependency of sensitivity of particle concentrations in the feed
- this in turn requires either highly sensitive counters or limits the membrane area that can be observed for damages as the dilution of permeate from an impaired membrane with permeate from intact membranes leads to a loss in sensitivity (Farahbakhsh et al., 2003; Antony et al., 2012)
- air entrapments may be detected erroneously as particles

An alternative way of increasing sensitivity of particle counting as a measure to evaluate membrane integrity is spiking the feed with a suitable surrogate (see chapter 2.5.4) whilst a detection of very small sized particles increases the sensitivity for detecting effects of a leak, this may also increase interference with background noise.A summary of devices used and set-up tested as well as detectable damages is given in Table 5.

Table 5 Tested particle counters and their detection ranges

Particle counter / Manufacturer	Detection range [µm]	Scale of testing	Matrix	MI Test	Detected damage	Source
Met One W215; Sensor 211	0.5 – 25	Pilot	Surface water	Particle counting		(Panglisch et al, 1998)
MET ONE 211W; Sensor LS211	0.5 - 25	Pilot & full	Raw surface water	Particle counting		(Glucina et al., 1997)
Klotz LDS 23/25	1 - 250	Lab	Tap water w/ latices	Particle counting		(Panglisch et al, 1998)
HIAC/ROYCO MC-80	0.08 – 0.2 (0.5 - 350)	Pilot	Surface water	Particle counting		(Panglisch et al, 1998)
HIAC/ROYCO 8000 A; Sensor HLRD 150	1.24 - 100	Pilot & full	Raw surface water	Particle counting	1 broken fibre out of 40000	(Glucina et al., 1997)
HIAC/ROYCO 8011 Lab Liquid Particle Counting System	n. s.	Lab	Deionised water, spiked nanoparticles	Challenge		(Gitis et al., 2006)
WPC 2000	>2	Industrial	Water with PAC	Challenge		(Guo et al., 2011)

2.5.3 Laser-Induced Breakdown-Detection

Laser-induced breakdown detection (LIBD) is based on the principle of detecting the dielectric breakdowns of solid matter generated in the high electrical field of a focused pulsed laser beam. The breakdowns can be measured acoustically, optically or by comparing the laser pulse energies before and after passing through the sample. It can be operated in continuous (detection of particles in the filtrate) and non-continuous (determination of particle retention characteristics by determining the particle size distributions) mode (Troester et al., 2014).

Troester et al. (2014) reached a LRV of over 4.5 for a particle size distribution of 50 – 200 nm in discontinuous mode in ultrafiltration lab-scale tests. In continuous mode, nanometric scale particles (20 nm) can be detected at concentrations as low as a few nanograms per liter. LIBD is a highly sensitive, stable and reliable online method, which requires no addition of nano-particles and is easily adjusted to specific process conditions by varying laser pulse energy and total number of laser pulses. The research group found it rather easy to handle and requiring low maintenance. For defect location and characteristics, a combination with a direct testing method is proposed. LIBD fulfils the practical requirements for full-scale operation, but has only been tested in lab-scale so far.

2.5.4 Surrogate challenge tests or marker-based tests

Challenge tests constitute a method to determine the performance of a membrane and its integrity with respect to a target particle or organism. The US EPA (2005) defines a challenge test as "*a study conducted to determine the removal efficiency (i.e., log removal value (LRV)) of a membrane material for a particular organism, particulate, or surrogate*".

It is thus a test method to evaluate the suitability of a barrier for defined pathogens. Using it for membrane integrity testing is rather a transfer of this specific objective to an overall membrane assessment.

The ultimate challenge test for technical systems would use the actual contaminant of concern, e.g. the natural (pathogenic) bacteria or viruses. But because of potential problems with bio-fouling and the penetration of bacteria into the permeate surrogates are used. Those shall represent relevant indicators for the demonstration of pathogen retention by membranes. Surrogates should be mono-dispersed, of well-defined size, easily detectable, non-destructive and reasonably economic (Guo et al., 2010b). Three general types of surrogates are applied:

- alternate microorganisms,
- inert particles and
- molecular markers.

Not all classes of surrogates are appropriate for all membrane filtration systems. For MF and UF systems, generally, particulate surrogates such as alternate microorganisms and inert particles will be appropriate (US EPA, 2005).

Surrogate challenge tests constitute an indirect method based on the relative detection of particles or organisms in the feed and the permeate which reaches higher sensitivities than other indirect methods mainly because of higher (spiked) feed concentrations and better detection technologies. Tests differ with respect to the type and size of surrogate used and the related detection method.

A particular form challenge test are nanoparticle challenge testing. In search for online testing procedures to verify membrane integrity for small pathogens such as enteric viruses various research groups work with nano-scale particles. If properly selected, advantages of nanoscale particles for integrity testing are their low background level, their non-pathogenicity, and their high mono-dispersity.

2.5.4.1 Challenge testing with microbial surrogates

For this type of testing non-pathogenic organisms such as MS2 phages or other challenge organisms are used as surrogates for the rejection target microbial organisms in test systems. The US EPA Guidelines (2005) suggest various microbial surrogates for the parasitic pathogens Giardia and Cryptosporidium as summarised in Table 6. Among the surrogates are bacteria and their spores as well as bacteriophage, representing a size range from few nanometers to around 10 µm.

Table 6 List of microbial surrogates used for challenge testing (US EPA, 2005).

Microorganism	Size Range (µm)	Target Organism	Enumeration Method
Micrococcus l.	7 - 12	Giardia	Standard Methods 9222
Bacillus subtilis (spores)	~ 1	Cryptosporidium	Barbeau et al. (1997)
E. coli	1 - 4	Cryptosporidium	Standard Methods 9222
P. dimunita 0.3		Cryptosporidium	Standard Methods 9222
S. marcessans	0.5	Cryptosporidium	Standard Methods 9222
MS2 bacteriophage	0.01	Enteric virus	Adams (1959)

MS2 is an F-specific RNA bacteriophage with a diameter of 0.024 µm, which is widely used for challenge testing and currently considered the best process indicator for enteric virus removal. Its morphological and structural properties as well as its survival characteristics in aquatic environments are similar to that of enteric viruses. Further advantages are its low isoelectric point (3.5-3.9) and its degree of hydrophobicity, which reduce the probability of adsorption to hydrophilic and negatively charged membranes (Antony et al., 2012).

The surrogates are classically detected and quantified by cultivating methods and expressed in plaque forming units. Though this is time consuming it allows for enumeration of viable organisms. More rapid advanced molecular biological approaches such as PCR methods are routinely customized. Such methods are not influenced by aggregation of particles and count both active and inactivated viral particles (Antony, Blackbeard, & Leslie, 2012), therefore representing a valuable alternative for further research.

The test is highly accurate and sensitive (LRV > 7 for viruses) and the culturing of the microorganisms is relatively easy. Nevertheless, it is rather impractical due to high costs, the effort connected to culturing the organisms and the precautionary handling necessities (Gitis et al., 2002; Antony et al., 2012).

In the context of challenge testing with microbial surrogates to ensure membrane integrity it needs to be considered that fouling and the formation of a cake layer have an increasing impact on virus retention. Whereas the cake layer is removed by back-washing, reversible fouling is only removed by chemical treatment and irreversible fouling leads to a permanent increase in virus retention, possibly over-estimating membrane integrity (Antony, Blackbeard, & Leslie, 2012).

Microbial challenge testing is most often used for validation and improvement of air-based test (Guo et al., 2010b) and not suitable for operational, every day monitoring of full scale plants (US EPA, 2005; Gitis et al., 2002).

2.5.4.2 Challenge testing with fluorescent dye labelled MS2 bacteriophage

To overcome detection drawbacks of viruses by the plaque technique, researcher elaborated an approach using fluorescent-dye-labelled MS2 bacteriophages which are detected by fluorescence spectrometry. The

signal is not influenced by aggregation of bacteriophage or their inactivation and is uncoupled from background noise caused by other similar-sized organisms present in the feed water.

The concentration of the labelled surrogates is determined by fluorometric methods and given in fluorescence intensity units (FIU) (Gitis et al., 2002, Gitis et al., 2006). While the measuring part of this approach is more convenient and faster than cultivation techniques, the preparation of the labelled phages is an ambitious and labour demanding step. Compared to challenge testing with non-biological nanoparticles, it provides much better emulation of viral transport. Its application is limited, though, by its low sensitivity, time and labour demands for cultivation and labelling of the organisms and necessary safety precautions (Gitis et al., 2006).

The approach has been tested in lab-scale experiments with deionised water on intact membranes of different material and at different pH values. Integrity testing with fluorescent-dyed bacteriophage reached LRV between 1.5 and 3, depending on pH, surface charge and MWCO of the membrane. The LRV values decreased with higher pH values. For only one type of PVDF membrane, a permeation of the probe through the membrane was detected. With a relatively high detection limit of 10^6 PFU/ml this method is much less sensitive than challenge tests with other nanoparticles and not yet suitable for full-scale application. To date, the method is limited by the sensitivity of the technique for detection of fluorescence.

2.5.4.3 Spiked integrity monitoring (SIM)

Spiked integrity monitoring (SIM™) was developed as a routine integrity monitoring for ultrafiltration membranes in drinking water production. It basically constitutes an online surrogate challenge test using powdered activated carbon as challenge particulate and particle counting as detection method.

A limited amount of powdered activated carbon (PAC) of particle size 1.7 µm (70% of all particles) is dosed into the feed for a short time. The particle count at the time of the spiking is compared to the count without spiking (van Hoof et al, 2003; Franklin et al., 2000).

SIM™ is a highly sensitive method, which reaches up to 6 LRV (Antony et al., 2012). With application of doses of 14'000 particles/ml to relatively clean feed water systems to identify a single compromised fibre, resulting in a drop in LRV from 5.8 to 5.0 (van Hoof et al., 2003; Franklin, B., et al., 2000). Compared with particle counting SIM™ has increased sensitivity, mainly due to the higher feed concentrations. Nevertheless, it is limited by the non-specificity of the method and the resolution of the detection system. SIM™ can be strongly affected by the background noise created by particles naturally present in water. The establishment of a baseline is relatively difficult.

In non-continuous mode, SIM™ is relatively easy to use, but it may be difficult and expensive applied in continuous mode (Farahbakhsh et al., 2003; Deluhery & Rajagopalan, 2008). With the membrane not being tested directly, SIM™ is an indirect method, whose results are not directly linked to pathogen removal. Problematic can also be the remaining powdered activated carbon in the permeate after spiking (Guo et al., 2010b).

SIM™ is used in full-scale at the IMS Heemskerk Water Treatment Plant for integrity monitoring with PAC particles of ≤ 3 µm, which do not interact with the membrane (Kruithof et al., 2001).

2.5.4.4 Magnetically susceptible (nano) particles method

A variant of the SIM is suggested by Deluhery and Rajagopalan (2008). They spiked magnetically susceptible particles of 1 µm size during lab testing of flat sheet membranes (0.6 µm pore-sized polycarbonate). The feed contained approx. 10^6 particles/mL. They detected the particles based on their magnetic susceptibility using magneto-relaxometry. In order to achieve a decent signal of the sample they concentrated the particles by capturing them in a particle collection column.

The set-up and methods proved capable of quantifying the spiked particles. Their approach is protected by Patent US 7011758 B2 (2006) as "Methods and systems for membrane testing". Further enhancement would be needed in real-case application for membrane integrity testing.

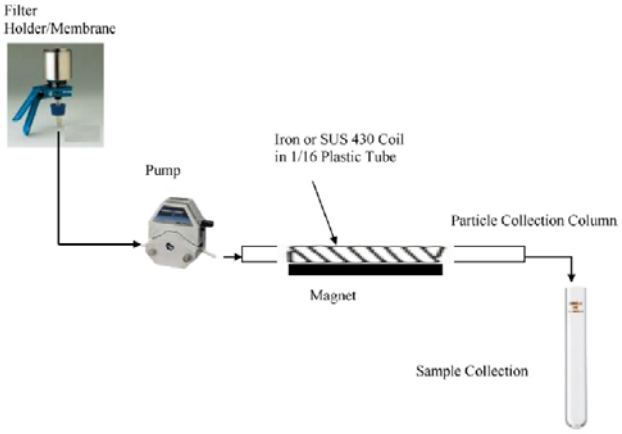


Figure 6 Experiment set-up for membrane integrity testing using magnetically susceptible particles - MSP (Deluhery and Rajagopalan, 2008).

Guo et al. (2010a) developed a similar method where particles were quantified by their magnetic susceptibility without a prior concentration step. There the desired properties of the surrogate particles are high magnetic susceptibility, low density and a particle size, which is greater than the pore size (0.01-0.1 µm). Iron oxide (Fe_2O_3) and magnetite (Fe_3O_4) particles of a mean size around 35 nm (20 – 100 nm) have been successfully used (French Patent No. EP1862791, EP 1862791 A2 2007) (Guo et al., 2010a).

They produced magnetic Fe_3O_4, particles of an average size of 35 nm (20-100 nm) and were able to measure the concentration of these particles by a magnetic susceptibility meter. The methods proved sensitive enough to detect the presence of a 1% fiber breakage rate in 100 fibre Norit X-Flow module. They also ruled out interference of other compounds present in surface water (turbidity) or used during regular water treatment, such as the dosing of ferric chloride ($FeCl_3$) as coagulant and for phosphorous precipitation with the dosed magnetic nanoparticles.

The test was applied in a pilot plant operating industrial scale X-FLOW ultrafiltration modules. Two modules (one intact, one to damage) were tested at the full-scale drinking water plant in Jaunay (Guo et al., 2011). A damage of two 0.6 mm punctures of two fibres was inflicted on one of the modules. A powdered activated carbon dosing (see SIM-test) and particle counting was not able to detect this damage. However, when dosing a sufficiently concentrated Fe_3O_4 nanoparticle solution, increase of the magnetic susceptibility in permeate samples was detectable within 30 seconds (at an hourly flowrate of 2.2 m^3) with one of the tested detection devices.

According to Guo et al. (2010a, 2010b) the magnetically susceptible particles method is a simple, online method that allows accurate and quick detection. It is applicable to low-pressure processes, has a low detection limit and a very low influence on membrane fouling. The nanoparticles are low cost, non-toxic and easily obtainable. The MSP method is highly sensitive (LRV > 6) with or without a concentration step, independent of operational mode (dead-end or cross-flow) and location of fibre. Without a prior concentration step it is possible to instantly detect 1% fibre breakage at feed concentrations as low as 1.2 ppm (lab-scale). Like with particle counting, the detection limit depends on the analytical equipment.

2.5.4.5 Fluorescent silica particles

The concept of this method is based on the use of rhodamine B isothiocyanate (RBITC)-doped silica particles (0.5 - 0.7 µm) as surrogate and their quantification by image analysis. This approach was investigated by Choi et al. (2011) and the procedures of particle labelling and membrane integrity testing are pending for patent (USPA, 2010). In their lab-tests particle concentrations are measured as the intensity of fluorescence emitted under UV excitation. To determine this, samples of the particle containing solutions are filtered through 0.45 µm glassfibre filter, dried and photographed under a UV lamp. The digital images are edited to remove background noise and converted to RGB format. The average RGB per pixel is then calculated and integrated to quantify the overall fluorescence intensity of the particles. The particles mass was estimated by establishing the correlation between particle mass and fluorescence. A schematic of the procedure is depicted in Figure 7.

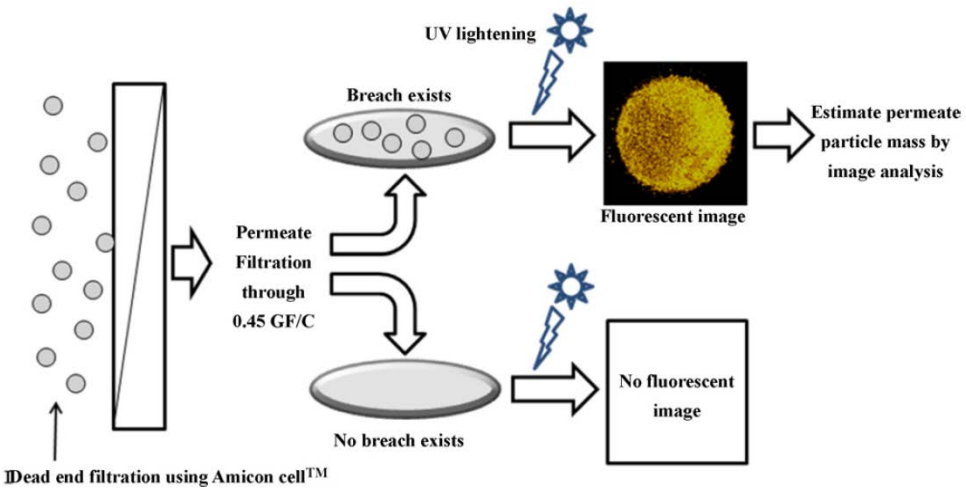


Figure 7 Schematic of the experimental configuration for integrity testing with fluorescent silica particles using image analysis (Choi et al., 2011)

Other than in the use of fluorescence dye labelled MS2 phage, this concept includes a concentration step to capture low particle concentrations from a bigger sample volume on downstream glass fibre filter before analysing the signal. However, the capacity to the filter to collect silica particles was found to be limited to GF/C filter is 7.96×10^{-7} kg/m^2.

They challenged a microfiltration membrane of a nominal pore size of 0.25 µm with particles in the range of 0.5 - 0.75 µm at different feed concentrations and pressures as well as with differently damaged membranes. Based on their results they established a functional relationship between the damaged area and the mass of fluorescent particles detected in the permeate.

Little effect on membrane fouling caused by dosed particles was observed in the lab-tests. Applied concentrations of 10-100 mg/l of particles in the feed solution caused around 10 % flux decline. The linear decline of flux implies that the fouling mainly occurred due to particle cake formation rather than direct pore blocking or internal pore adsorption.

The results indicate that the mass of outflow particles depends on the size of the breach and the particle concentration. However, particle concentration appears to be more influential on the outflow mass compared to the size of the breach.

Whilst the approach is smart it has limited value for membrane integrity testing in operational full-scale plants, due to potential release of labelled particles in the environment. The comprehensive sample treatment and assessment also seems not suitable for online measurements.However, further studies are required to enhance the feasibility of the proposed method for its application in the field. For instance, the synthesis and application of iron-embedded fluorescent particles is being studied in order to retrieve the used particles with a magnetic force while preventing unexpected harmful impacts when they are discharged into the environment (Choi et al., 2011).

2.5.4.6 Challenge testing with gold and silver nanoparticles

Nobel metal nanoparticles are deemed suitable challenge particulates for mimicking virus removal in ultrafiltration applications. The particles are chosen because of their high monodispersity, which facilitates measurement, their non-pathogenicity and extremely low background level in water and chemical inertness.

Gitis et al. performed challenge testing with citrate-stabilized (12 nm) or thiol-stabilized (15 nm) gold nanoparticles in a test cell filtration set-up (Gitis et al., 2006). Photometric methods or electrochemical detection by anodic stripping voltammetry (ASV) have been used to detect the gold nanoparticles. Feed solutions of 5.2 mg/l of gold in deionised water were used for experiments.

The authors found that the particles were retained by the intact membranes, but LRV values depended on pH, surface charge and MWCO of the membrane. They decreased with higher pH values up to a certain point when citrate-stabilized gold were filtered through PVDF-55 membranes (from 1.7 to 1.3 LRV). LRV values did not depend on pH for the same surrogate when filtered through CA-10, CE-20, or PES-15 membranes and were highest with 4.5. Also, thiol-stabilized gold particles did not depend on pH when filtered through PVDF-55 membranes but were retained less effectively (LRV 1.5).

The authors only characterise the retention behaviour of intact membranes and found relatively low LRV, which would make the detection of small damages potentially difficult.

Antony et al., 2014 performed a more comprehensiv testing. They introduced a simple, quick and relatively inexpensive online membrane integrity test using citrate stabilised, spherical, zero-valent silver (60 nm) nanoparticles. They applied them on ultrafiltration hollow fibre membranes (outside-in operated PVDF membranes, 0.04 nm pore size, 0.025 m^2 membrane area, Siemens Water Technologies, Australia). Membrane were compromised either mechanically (pin hole or breach) or chemically (sodium hypochlorite treatment). The challenge particles were provided in different feed concentrations of 12.3 mg/l silver particles, and the membrane was operated at a constant flux of 30 or 50 LMH. Nanoparticles in feed and permeate were quantified by inductively coupled plasma-optical emission spectroscopy (ICP-OES).

At nanoparticle feed concentrations they were able to detect the inflicted membrane damage.

The approach nicely tracked the various degrees of damage as illustrated in Figure 8. The initial damage with one pinhole of 100 µm already cause a drop of the log removal value (LRV) of intact membranes from 2.8 to 1.3, which gradually increased with additional damages.

Also the effect of chemical aging could be depicted in loss of rejection capacity, yet only when applying NaOCl exposure levels of > 10’000 mg/(L x h).

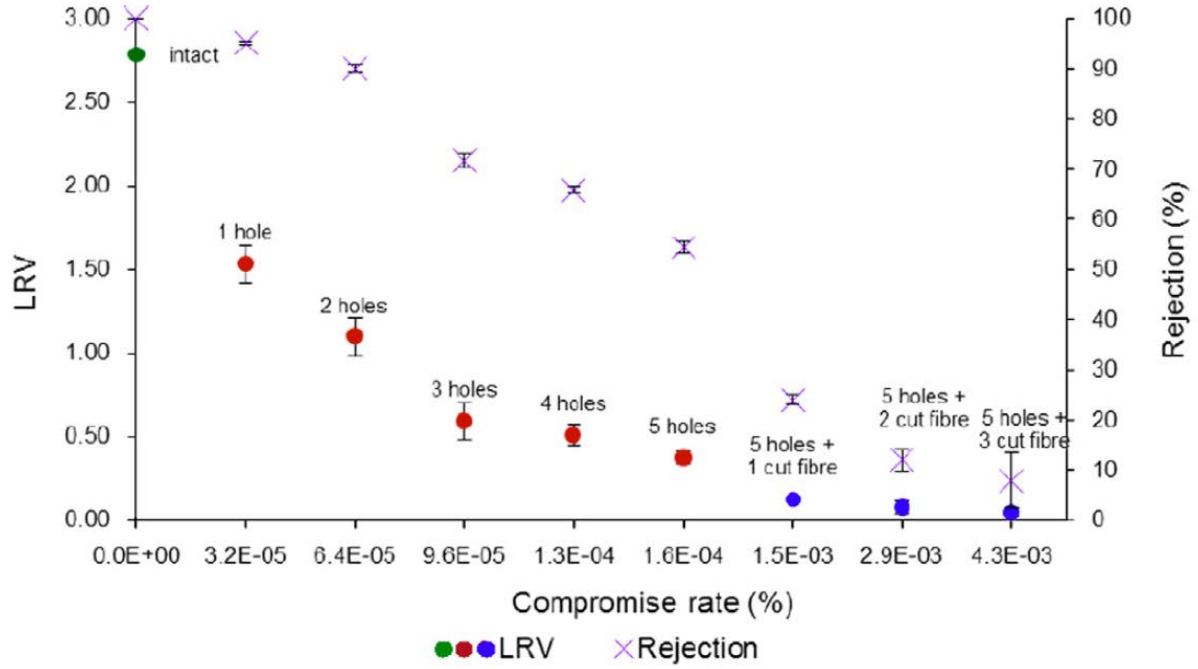


Figure 8 LRV for the physically compromised membranes (compromise rate is the ratio of the compromised area to the total effective membrane area) (Antony et al., 2014)

Although the physical characteristics of gold- and silver nanoparticles are different from viruses and do not consider the surface chemistry, vitality and pathogenesis of the target organism, they show several advantages over bacteriophage as virus indicators. Apart from the higher achieved sensitivity, they are non-pathogenic and safe, require relatively low labour for generation and only minimal personal protective equipment. Since the particles are not microbial, the risk of their contamination is low. For measurement, onsite techniques can be used, guaranteeing a small lead time. Further, the risk of deformation of particles under high pressure, which exists for microbial surrogates, does not exist in the case of nanoparticles (Antony et al., 2014). Gold and silver nanoparticles are highly monodisperse and their use economic.

Challenge testing with gold and silver nanoparticles showed high sensitivity and a low detection limit. With silver nanoparticles LRV values of close to 3 (equalling 3 damaged fibres in 100'000 fibres), with the described gold particles LRV values up to 4.5 were reached for virus-sized particles.

Antony et al. (2014) shortly addressed the problem of nanoparticles being released into the environment as a consequence of the challenge tests, but considered it possible to mitigate. On the one hand, most of the particles will be retained by the membrane and discharged into the backwash. On the other hand, the citrate shell is biodegradable (Vieira, Silva, Santos, & Beppu, 2011) and it is possible to recover the silver by electrochemical techniques (Chen, 2004).

Whilst handling of the challenge particles is deemed favourable for large-scale application, the detection methods are rather elaborated and not easily at hand in operational plants. This may hamper the application for routine membrane integrity testing but poses an opportunity for validation monitoring of installed treatment schemes.

2.5.5 Summary of characteristics of different particle-based tests

Table 7 Characteristics of particle-based

Method	Mode	Characteristics	Source
Turbidity monitoring	on-line	LRV ≤ 2 for particles down to 1 µm, continuous, indirect, non-destructive, less expensive, but also less sensitive than particle counting, non-specific, low sensitivity at low particle concentrations, slow response time, standard equipment of most surface water treatment plants, for all membrane configurations	(Antony et al., 2012) (Farahbakhsh et al., 2003) (Guo et al., 2010b) (Liu, 2012) (Naismith, 2005)
Particle monitoring	on-line	Much lower sensitivity than particle counting, minimum detectable particle size: 1 µm, continuous, indirect, non-destructive, non-specific, not applicable in many cases (e.g. very pure fluids), poor correlations with other integrity tests, for all membrane configurations	(Antony et al., 2012) (Glucina et al., 1997) (Guo et al., 2010b) (US EPA, 2005)
Particle counting	on-line	LRV < 4, minimum detectable particle size: 0.5 µm, continuous, indirect, non-destructive, non-specific, not applicable in many cases (e.g. very pure fluids), poor correlations with other integrity tests, limited by operating conditions, feed particle concentration and membrane surface area, affected by dilution ratio and particle counting equipment, for all membrane configurations (sensitive enough in cross-flow mode)	(Antony et al., 2012) (Glucina et al., 1997) (Guo et al., 2010b)
Laser-induced breakdown sdetection	on-line	LRV > 4.5 for nanometric size particles (discontinuous mode), detection of nanometric size particles at very low concentrations (continuous mode), high sensitivity, reliable, stable, easy handling, low maintenance, higher sensitivity with increased laser pulse energy, laser pulse energy limited by breakdown treshold of particles < MWCO	Tröster et al. (2014)
Surrogate challenge test	on-line	High sensitivity, LRV = 5 - 6, non-continuous, indirect, possibly destructive, depending on surrogate for all membrane configurations	(Guo et al., 2010b)
Microbial challenge test	on-line	LRV > 7 for UF, non-continuous, indirect, may be destructive, highly accurate, easy to perform, but high cost and effort for microorganism cultures, currently best indicator for virus removal, used for validation of air-based tests, not usable for full-scale application	(Antony et al., 2012) (Gitis et al., 2002) (Guo et al., 2010b) (US EPA, 2005)
Fluorescent dye labelled MS2 bacteriophage	on-line	LRV = 1.5 - 3.0, non-continuous, destructive, indirect, destructive, time- and labour-intensive, can cause cake fouling, simpler and more specific	(Gitis et al., 2002) (Gitis et. al., 2006)

Method	Mode	Characteristics	Source
		measurement than PFU analysis, better emulation of viral transport than with non-biological surrogates, convenient tool for background analysis	
SIM™	on-line	LRV ≤ 6, online, indirect, non-continuous, accurate, fast, non-specific, increased sensitivity with increased particle concentration, limited sensitivity due to background noise, no direct link to pathogen removal, applicable to all membrane configurations	(Antony et al., 2012) (Franklin et al., 2000) (Guo et al., 2010b) (van Hoof et al., 2003)
Magnetically susceptible particles method	on-line/ off-line	LRV > 6, non-continuous, simple, accurate, quick, specific, independent of operational mode and location of fibre, low detection limit, sensitivity depends on analytical equipment, permeate flux, injection mode and particle concentration, enhanced sensitivity with concentration step, very low influence on membrane fouling, applicable to all MF and UF membrane configurations	(Antony et al., 2012) (Deluhery & Rajagopalan, 2008) (Guo et al., 2010a) (Guo et al., 2010b) (Guo et al., 2011)
Stabilised gold and silver nanoparticles	on-line	LRV ≤ 4.5 for UF (depending on pH, surface charge and MWCO of the membrane), simple, quick, safe, economic, low labour, different physical characteristics than virus particles, for MF to RO, potential for water reuse systems	(Antony et al., 2014) (Gitis et al., 2006)

2.6 Membrane integrity sensor

The membrane integrity sensor is an online and direct method to observe the integrity of a filtration membrane. It measures the fouling propensity of the permeate from the pre-filtration membrane module. Initially, the system detected the deposition of particles and other foulants from the sample stream onto a sensor membranes and the related change in trans-membrane pressure. It is installed in the side-stream of the regular permeate stream. Any elevated particle concentration in the permeate stream will also pass through the sensor where the transmembrane pressure across a microfiltration membrane in the Integrity Sensor is measured relative to a reference pressure differential (Krantz et al.2010).

An improved sensor was developed by Fane et al. (2010). They replaced the lower membrane by a valve with an adjustable throat diameter and a clearance on the scale of a millimeter. This design eliminates the problem of membrane fouling (Figure 9).

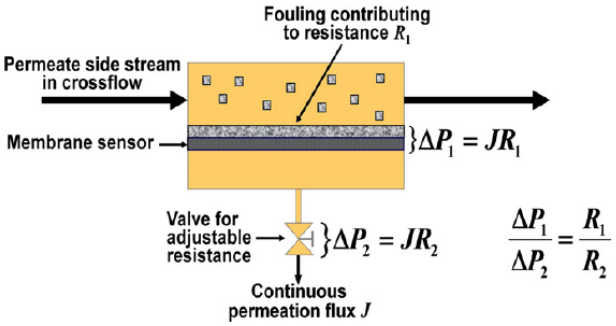


Figure 9 Schematic of new Integrity Sensor design of Fane et al. (2010) with a valve in place of the lower membrane (Krantz et al., 2011)

A so-called π-factor is established as dimensionless metric representing the ratio of the transmembrane pressure to the reference pressure differential. In an unfouled state of the microfiltration membrane the π-factor will take an initial value, which will gradually increase due to fouling. A breech in the integrity of the upstream pre-filtration device is detected by a marked increase in the π-factor (Krantz et al., 2010).

The possibility of backwashing and the adjustability of the valve guarantee long term operation of the sensor with maintained sensitivity. The breaking of merely one of 1500 hollow fibers in a full-scale UF module can be detected reproducibly within 10 min after its occurrence (Krantz et al., 2011).

Following various development steps, membrane integrity sensor is now manufactured by MINT (Figure 10). An instructual video can be found here: https://www.youtube.com/watch?v=EYCuf9IP1dA

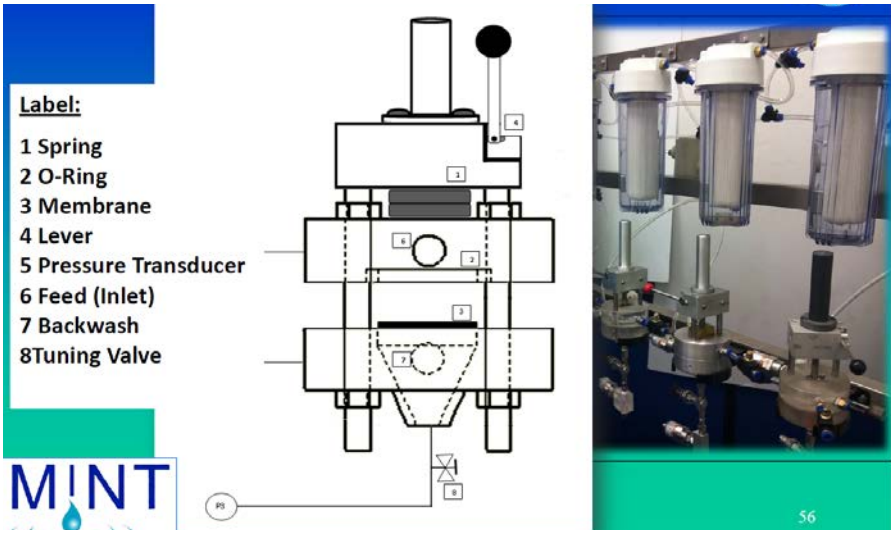


Figure 10 Schematic and view of test stand of the Membrane Integrity Sensor
Source: Presentation of Fane and Chong given at IDA R&D Workshop, March 17-19, 2013, Jeddah "Desalination research centers, achievement and future trends " from 17 – 19 March 2013 at Jeddah Hilton hotel, Saudi Arabia. MINT = Membrane Instruments and Technology private limited

Table 8 Characteristics of membrane sensors (Guo et al., 2010), complemented with information from (Krantz et al. 2010 and 2011) and (Phattaranawik et al., 2008)

Method	Mode	Characteristics
Membrane based sensor (2 membranes)	on-line	reliable, sensitive and low-cost; potential to monitor influent quality to NF and RO processes; independent of permeation rate through membrane; difficult backwashing; decreased sensitivity with increased fouling; no adjustment to maintain sensitivity possible
Membrane Integrity Sensor	on-line	reliable, sensitive and low cost; no fouling problems, rapid response, sensitivity can be maximised by adjusting valve convenience for retrofitting to water-treatment systems, continuous real-time sensing capability, high sensitivity, reliability, robustness, and low cost

3 Full-scale applications

In drinking water treatment, the most frequently used membrane integrity testing methods are the pressure decay test (PDT) and the diffusive airflow test (DAF). They are simple, reliable and highly sensitive and require only low maintenance. PDT is highly automated and a standard part of most MF and UF systems. (Guo et al., 2011). The main drawback is that they are performed off-line. Also, they do not measure the water quality, thereby providing only an indirect indication, whether the necessary water quality is actually reached by filtration (Guo, Wyart, Perot, Nauleau, & Moulin, 2010). Vacuum hold test (VHT) is a standardised method for NF and RO, but rarely used in drinking water treatment (Guo et al., 2011).

Studies on validation of alternative test in full-scale applications are rare and covering different approaches for particle counting and microbial challenge tests as

The most frequently used indirect methods are particle counting, turbidity monitoring and routine microbial analysis (Crozes et al., 2002; Guo et al., 2010). Indirect methods can be conducted online and usually make a comparison of the measured value of a certain parameter of the filtrate to a baseline level. They are applicable to any membrane system, but possess only low detection sensitivity (Guo et al., 2010). A survey in the frame of an AWWARF research project showed that particle counting was used for integrity monitoring of low-pressure filtration systems by almost half of the surveyed plants, whereas particle monitoring was almost not used (Crozes et al., 2002).

The criteria met by currently applied methods reflect the international position, but do not satisfy the regulatory requirements for an absolute removal of more than 1 µm. Nanometric breaches cannot be detected (Guo et al., 2011). Therefore, new on-line tests are needed. Table 9 gives an overview of currently applied methods.

Irwin et al. (2014) report on a full-scale study assessing the effects of cut-fibres on both virus removal and direct integrity testing by pressure decay test.

Table 9 Currently applied membrane integrity testing methods (technical and industrial scale, n.s. not specified)

Technology Developer	Partners	Application field	Reference
Implemented technologies			
SIM™	IMS in Heemskerk Water Treatment Plant	Water Treatment	Kruithof et al., 2001 in Antony et al., 2012
SIM™ NORIT membrane technology	Water Supply Company North Holland & IWW Rhenisch-Westphalian Institute for Water Research	Drinking water supply	Guo et al., 2010
Memsure © Memcor	several hundred CMF plants around the world	From high purity water to primary sewage, often drinking	Randles, 1996
Memsure © Memcor	Saratoga water treatment plant, California, USA	Alternate season (drinking) water supply from high turbidity water (> 250 NTU)	Randles, 1996
Memsure © Memcor	Kenosha Water Utility, Wisconsin, USA	Potable water treatment	Randles, 1996

Technology Developer	Partners	Application field	Reference
Memsure © Memcor	Eraring power station project: Pacific Power (PP) & Hunter Water Corporation (HWC), Australia	Water reuse scheme boiler feed water production	Randles, 1996
Memsure © Memcor	n. s.	Industrial, continuous microfiltration (CMF)	Guo et al., 2010
Memsure © Memcor	Joyce Road Water Processing Plant, New Zealand	CMF	Antony et al., 2012
Turbidity measurement and PDT	San Patricio Municipal Water District, Texas, USA	Wholesale supply of water to cities and industry	Naismith, 2005
Alarm set against pH or turbidity	n. s.	Drinking water treatment plants	Guo et al., 2010
Industrial scale testing			
PDT, AIM, particle counting and monitoring, turbiditiy monitoring	n. s.	6 MF and UF wa-ter treatment plants	Crozes et al., 2002
Microbial challenge testing	n. s.	San Patricio WTP & Appleton WTP	Crozes et al., 2002 Brehant et al., 2010.
Fe_3O_4 nanoparticle challenge test	n. s.	Jaunay drinking water treatment plant; SAUR group (France)	Guo et al., 2011
Hydrophonic sensor	n. s.	La Fillière & Avoriaz UF plants	Laîné et al., 1998
Hydrophonic sensor	n. s.	Vigneux sur Seine UF plant	Laîné et al., 1998
PDT and MS2 phage challenge test		Drinking water treatment plants Validation of a PDT model to detect defects of 3 µm	Brehant et al. 2010
PDT and MS2 phage challenge test	South East Recycled Water Alliance (SERWA, AUS)	Water reclamation plants Validation of 4 LRV of the UF stage	Irwin et al., 2014
Membrane Integrity sensor MINT	PUB (Public Utility Board, Singapore)	Water reclamation plant (NeWater)	

A questionnaire based survey among full-scale water reclamation plants operators also confirmed that turbidity is usually monitored in membrane permeate (3 out of 4 plants). Pressure decay tests are also performed on a regular basis (every 3 to 4 weeks) in half of the plants.

Frequency of other monitoring or testing is adjusted to technical requirements of downstream treatment processes and the sensitivity of the intended water reuse application (indirect potable reuse, urban applications). E.g. the silt density index is observed daily when the ultrafiltration step is followed by reverse osmosis.

4 Overview tables

Table 10 Overview of membrane integrity methods

Method	Mode	Functioning	Characteristics	Reference
PDT	off-line	Measurement of pressure drop on the drained side after pressurising	LRV = 4.5-5 for Giardia or Cryptosporidium, detection limit 2-3 µm, duration 10 min., direct, non-destructive, very reliable, dependent on membrane area and fluid temperature, for spiral wound and hollow fibre modules	(Antony et al., 2012) (Farahbakhsh et al., 2003) (Gitis et al., 2006) (Guo et al, 2010b)
DAF	off-line	Measuring diffused gas or water flow through fully wetted membrane applying constant feed gas pressure below bubble point	LRV > 6; non-continuous, easy and accurate, detection limit 2-3 µm, test duration 15 min., non-destructive, widely used in water treatment plants, sensitive to temperature, reflects total porosity of filter → needs validation, for spiral wound and hollow fibre modules	(Antony et al., 2012) (Gitis et al., 2006) (Guo et al., 2010b) (Meltzer et al., 1999)
VDT	off-line	Like PDT, but with vacuum applied instead of pressure	for membranes that cannot be pressurised on filtrate side, duration several minutes, currently rarely applied for MF/UF, useful as screening procedure, for spiral wound and immersed membranes	(Adams & Coté, 2005) (Guo et al., 2010b)
Bubble-point test	off-line	draining and pressurising module, uniform wetting of	LRV 6 in Memcor© CMF processes, detection limit 2-3	(Antony et al., 2012) (Gitis et al., 2006)

Method	Mode	Functioning	Characteristics	Reference
Bubble-Point Method (gas-liquid porosimetry)		membrane, application of dilute surfactant solution, observing pressure at which a steady stream of bubbles is reached	µm, non-destructive, diagnostic tool, temperature dependent, works well in combination with PDT, not applicable to UF, for spiral wound and hollow fibre modules	(Guo et al., 2010b) (Hofmann, 1984) (Randles, 1996)
Memsure	off-line	(1) Memsure™ PDT or Memsure™ DAF; (2) identification of leaks using Memcor® Sonic Analyser; (3) isolation of faulty modules (4) localisation of leak (5) repair on site	LRV = 6 for bacteria, for particles > 0.2 µm, duration 5 min., direct, non-destructive, simple, low maintenance, cost-effective, low energy consumption, low use of chemicals, extended life of filtration system, fully-automated, widely used in water treatment plants, not applicable to UF, for CMF systems (0.2 µm PP hollow fibre membranes)	(Guo et al., 2010b) (Johnson, 1997) (Johnson, 1998) (Randles, 1996)
Sonic test	off-line	A sound wave sensor is attached to headphones and a diode visual display and is manually placed at several locations on the membrane; difference between intact and defective fibres is identified by an operator	LRV 4.5 – 5 for single cut fibres, non-continuous, diagnostic, non-destructive, relatively simple, independent of feed water quality, results may be subjective, for MF and UF, not for submerged systems	(Antony et al., 2012) (Farahbakhsh et al., 2003) (Guo et al., 2010b) (Landsness, 2001)
Acoustic integrity monitoring	on-line	Hydrophonic sensor technology; noise signal (= pressure fluctuation) is continuously measured and compared to a threshold; distinctive noise signal is created by a defect fiber	LRV > 6 for viruses 100% of time, continuous, direct, non-destructive, relatively simple, independent of feed water quality, depends significantly on background noise and flow rate, for MF and UF, for spiral wound and hollow fibre membranes	(Antony et al., 2012) (Guo et al., 2010b) (Laîné et al., 1998)

Method	Mode	Functioning	Characteristics	Reference
Turbidity monitoring	on-line	Measurement by conventional or laser turbidimeters; comparison of turbidities of feed water and filtrate	LRV ≤ 2 for particles down to 1 µm, continuous, indirect, non-destructive, less expensive, but also less sensitive than particle counting, non-specific, low sensitivity at low particle concentrations, slow response time, standard equipment of most surface water treatment plants, for all membrane configurations	(Antony et al., 2012) (Farahbakhsh et al., 2003) (Guo et al., 2010b) (Liu, 2012) (Naismith, 2005)
Particle monitoring		Principle of light obstruction; measurement of fluctuations in the intensity of a narrow light beam sent through the fluid relative to a baseline; result given as a water quality index	Much lower sensitivity than particle counting, minimum detectable particle size: 1 µm, continuous, indirect, non-destructive, non-specific, not applicable in many cases (e.g. very pure fluids), poor correlations with other integrity tests, for all membrane configurations	(Antony et al., 2012) (Glucina et al., 1997) (Guo et al., 2010b) (US EPA, 2005)
Particle counting	on-line	Laser-based light scattering principle; a sensor is used to count and group particles in the size range of 0.5 to 5 µm	LRV < 4, minimum detectable particle size: 0.5 µm, continuous, indirect, non-destructive, non-specific, not applicable in many cases (e.g. very pure fluids), poor correlations with other integrity tests, limited by operating conditions, feed particle concentration and membrane surface area, affected by dilution ratio and particle counting equipment, for all membrane configurations (sensitive enough in cross-flow mode)	(Antony et al., 2012) (Glucina et al., 1997) (Guo et al., 2010b)

Method	Mode	Functioning	Characteristics	Reference
Laser-induced breakdown-detection	on-line	Acoustic, optical or comparative detection of the dielectric break-downs of solid matter generated in the high electrical field of a focused pulsed laser beam; ad-justment to a specific UF pro-cess by varying laser pulse en-ergy and total number of laser pulses	LRV > 4.5 for nanometric size particles (discontinuous mode), detection of nanometric size particles at very low concentra-tions (continuous mode), high sensitivity, reliable, stable, easy handling, low maintenance, higher sensitivity with increased laser pulse energy, laser pulse energy limited by breakdown threshold of particles < MWCO	
Surrogate challenge test	on-line	Spiking the feed with monodispersed, easily detectable surrogate material of defined size, measuring and comparing surrogate material in the feed and the permeate	High sensitivity, LRV = 5 - 6, non-continuous, indirect, possibly destructive, depending on surrogate for all membrane configurations	(Guo et al., 2010b)
Microbial challenge tests	on-line	Dosing of MS2 or other challenge organisms into the feed, measurement in the permeate by PFU analysis or RT-PCR method	LRV > 7 for UF, non-continuous, indirect, may be destructive, highly accurate, easy to perform, but high cost and effort for microorganism cultures, currently best indicator for virus removal, used for validation of air-based tests, not usable for full-scale application	(Antony et al., 2012) (Gitis et al., 2002) (Guo et al., 2010b) US EPA, 2005
SIM™	on-line	Surrogate challenge test with particulate activated carbon (PAC); PAC dosing into the feed and particle measurement in permeate	LRV ≤ 6, online, indirect, non-continuous, accurate, fast, non-specific, increased sensitivity with increased particle concentration, limited sensitivity due to background noise, no direct link to pathogen removal,	(Antony et al., 2012) (Franklin et al., 2000) (Guo et al., 2010b) (van Hoof et al., 2003)

Method	Mode	Functioning	Characteristics	Reference
			applicable to all membrane configurations	
Magnetic Susceptible particles	on-line / off-line	Spiking of superparamagnetic particles into the feed; detection in the permeate with magnetic sensors; with or without concentration step induced by applying a magnetic field	LRV > 6, non-continuous, simple, accurate, quick, specific, independent of operational mode and location of fibre, low detection limit, sensitivity depends on analytical equipment, permeate flux, injection mode and particle concentration, enhanced sensitivity with concentration step, very low influence on membrane fouling, applicable to all MF and UF membrane configurations	(Antony et al., 2012) (Deluhery & Rajagopalan, 2008) (Guo et al., 2010a) (Guo et al., 2010b) (Guo et al., 2011)
Fluorescent dye labelled MS2 bacteriophage	on-line	Spiking of fluorescent dye labelled MS2 bacteriophage into the feed, detection of feed and permeate concentration by fluorometric method, LRV as indicator of system performance	LRV = 1.5 - 3.0, non-continuous, destructive, indirect, destructive, time- and labour-intensive, can cause cake fouling, simpler and more specific measurement than PFU analysis, better emulation of viral transport than with non-biological surrogates, convenient tool for background analysis	(Gitis et al., 2002) (Gitis et. al., 2006)
Stabilised gold and silver nanoparticles	on-line	Spiking of nanoparticles at high concentrations into the feed for a certain time, measurement of their concentration in feed and permeate by photometric or electrochemical methods	LRV ≤ 4.5 for UF (depending on pH, surface charge and MWCO of the membrane), simple, quick, safe, economic, low labour, different physical characteristics than virus	(Antony et al., 2014) (Gitis et al., 2006)

Method	Mode	Functioning	Characteristics	Reference
			particles, for MF to RO, potential for water reuse systems	
Periodic testing	on-line	Periodic measurements of electrical conductivity, TOC, colour, sulfate, UV absorbance, specific ion concentration	Better results than turbidity/particle monitoring (esp. colour and TOC)	
Membrane Integrity Sensor	on-line	Similar to sensor with 2 membranes; second membrane is replaced by a valve	no fouling problems, rapid response, sensitivity can be maximised by adjusting valve	
Binary gas integrity test	off-line	Different permeabilites of two gases through liquid layer of wetted membrane; measurement of downstream gas composition: unexpectedly high amount of slower permeating gas indicates defect	LRV > 6; independent of membrane properties; non-destructive; sensitivity dependant on separation selectivity of gases; only realised in lab-scale so far	
Liquid-liquid porosimetry	off-line	Two immiscible fluids; flow rate ratio between total flow through membrane (Qtot) and flow (Qds) through membrane pores ≥ solute particle diameter (ds) at specific operating pressures gives percentage of total flow through membrane pores accessible to a given size particle	high sensitivity; non-destructive; independent of membrane surface area and geometry; relatively simple to perform; possible to measure pore size distribution, low pressure necessary (compared to PDT) → economically and practically feasible	

4.1 Methods – Advantages and Disadvantages

Table 11 Advantages and disadvantages of proposed methods

Method	Advantages	Disadvantages	Source
Pressure decay test	• LRV = 4.5-5 for *Giardia* or *Cryptosporidium* removal • very reliable, non-destructive • low maintenace • directly related to log removal capacity • standard part of many UF/MF systems • highly automated • independent of feed and filtered water quality • advance warning possible • sensitive to demonstrate integrity of a plant	• Off-line, non-continous • Low sensitivity for defects < 1.5µm • No measurement of filtrate quality • Dependent on fluid temperature and membrane area • False-positive results with not fully wetted membrane • Rapid pressure decay via diffusion in full scale plants • Dilution effect • LRV calculated at a given time for a given membrane integrity (≠ continuous measurements)	(Antony et al., 2012) (Farahbakhsh et al., 2003) (Guo et al., 2010b) (Johnson, 1997) (Johnson, Predicting log removal performance of membrane systems using in-situ integrity testing, 1998)
DAF	• LRV > 6 • Non-destructive • Easy and accurate • Low maintenance • Independent of feed and filtered water quality • Advance warning possible • Widely used in water treatment plants	• Off-line, non-continous • Not sensitive enough for nanoscale breaches • No measurement of filtrate quality • Reflects total porosity (needs validation) • Sensitive to temperature • False-positive results with not fully wetted membrane • Needs additional pipe work	(Antony et al, 2012) (Farahbakhsh et al., 2003) (Guo et al., 2010b) (Meltzer et al, 1999)
VDT	• Non-destructive • Independent of feed and filtered water quality • Possible to test membranes that cannot be pressurised on the filtrate side	• Off-line • No measurement of filtrate water quality • Not as widely used as PDT • Not fully developed and proven • More difficult to conduct	(Farahbakhsh et al., 2003) (Guo et al., 2010b)
Bubble point test	• LRV 6 in Memcor® CMF processes • Non-destructive • Standard procedure • Easy to conduct	• off-line • no measurement of filtrate quality • time-consuming and labour-intensive • practically unattainable for UF membranes	(Farahbakhsh et al., 2003) (Giglia & Krishnan, 2008) (Guo et al., 2010b)

Method	Advantages	Disadvantages	Source
	• Reliable and easy to interpret • Independent of feed and filtrate water quality • Identification of individual compromised fibres	• not for monitoring an entire system or module • possibly subject to operator error • use only complementary to other methods	
Memsure	• LRV = 6 log for bacterial removal • Non-destructive • Fully-automated • Reliable, simple, low maintenance • Independent of feed and filtrate water quality • Utilises existing low-pressure air backwashing step • Advanced warning possible • Low use of energy and chemicals • Extended service life of modules (5-7 yrs)	• Off-line, non-continous • Not applicable to UF • Same as PDT and DAF	(Antony et al., 2012) (Guo et al., 2011) (Johnson, 1997) (Johnson, 1998) (Randles, 1996)
Sonic test	• Can identify compromised modules (and fibres) • Relatively easy to use	• Not automated • Affected by background noise and operation mode • Results may be subjective • Time-consuming and labour-intensive • Not for submerged systems	(Farahbakhsh et al., 2003) (Guo et al., 2010b)
Acoustic integrity monitoring	• LRV > 6 for viruses • on-line • non-destructive • simple • economically competitive • independent of feed water quality • identification of compromised module • competitive	• strongly affected by background noise, flow rate and mode of operation • no measurement of filtrate quality	(Guo et al., 2010b) (Laîné et al., 1998)
Turbidity monitoring	• on-line, continous • non-destructive • less expensive than particle counting • Independent of membrane configuration • standard equipment in most surface water treatment plants • Convenient for routine qualitative monitoring	• LRV ≤ 2 • low sensitivity for defects < 1 µm • non-specific • slow response time • Strongly depends on feed concentration and operating conditions	(Antony et al., 2012) (Farahbakhsh et al., 2003) (Guo et al., 2011) (Naismith, 2005) (Phattaranawik et al., 2008)

Method	Advantages	Disadvantages	Source
	• increased sensitivity (x100) with new laser units • increased sensitivity (x100 of laser turbidimeter) with %RSD (relative standard deviation) as measure (→ independent of calibration)	• false-positive results possible due to microbubbles in filtrate or particle shedding in plumbing • requires regular maintenance and calibration	
Particle monitoring	• on-line and continuous • non-destructive • Significantly lower cost than particle counters • More sensitive than turbidity monitoring • Independent of membrane configuration • sensitivity increases with particle concentration • independent of membrane configurations • No calibration required	• Low detection sensitivity • Minimum detectable particle size: 1 µm • Non-specific • Strongly depends on feed concentration and operating conditions • Results harder to interpret than particle counters • Requires regular cleaning of sensor • Seldom used in the water industry	(Adham et al., 1995) (Farahbakhsh et al., 2003) (Guo et al., 2011)
Particle counting	• on-line and continuous • non-destructive • higher sensitivity than turbidity and particle monitoring • Independent of membrane configuration • widely used as standard equipment • sensitivity increases with particle concentration • independent of membrane configurations • can be used in multiple-channel configuration to save cost	• LRV < 4 (due to particle counter performance) • Minimum detectable particle size: 0.5 µm • non-specific • Strongly depends on feed concentration and operating conditions • loss of sensitivity due to dilution effect • not sensitive enough in dead-end mode • poor correlation with challenge tests • Relatively high costs • requires difficult and frequent calibration, regular maintenance and cleaning of sensor • Flow control devices are recommended before the sensor	(Adham et al., 1995) (Antony et al., 2012) (Farahbakhsh et al., 2003) (Glucina et al., 1997) (Guo et al., 2011) (Landsness, 2001) (Liu, 2012)
Laser-induced breakdown-detection	• high sensitivity, reliable, stable, no need to spike nanoparticles and chemicals • detection of nanoscale breaches at very low concentrations (few ng/l) • easily adjustable to a specific process → low background noise • easy to handle, low maintenance	• indirect method • not possible to localise and determine size of defect	(Troester et al., 2014)

Method	Advantages	Disadvantages	Source
	• fulfils practical requirements for operation in full-scale UF processes		
Surrogate challenge test	• LRV = 5 - 6 • on-line • depending on surrogate for all membrane configurations	• indirect, non-continuous • Possibly destructive • Relation to virus removal capacity must be verified	(Guo et al., 2010b)
Microbial challenge tests	• LRV > 7 for UF • on-line • highly accurate • easy culturing of microorganisms • PFU analysis well established	• non-continuous • no real-time monitoring possible • not applicable to full-scale systems • substantial logistics problems • high cost and effort for microorganism cultures • possible over-estimation of membrane retention • difficult differentiation between physicochemical retention and biological inactivation	(Antony et al., 2012) (Farahbakhsh et al., 2003) Gitis et al., 2002) (Guo et al., 2010b) (US EPA, 2005)
SIM™	• LRV ≤ 6 • on-line • relatively easy to use • accurate, fast • no interaction of particles with membrane • high sensitivity compared to simple particle counting	• non-continuous • non-specific • high background noise • relatively difficult to establish baseline value • PAC in permeate • results not directly linked to pathogen removal • accuracy limited by monitoring devices • particle size not constant during test • large variations in particle size distribution • no testing protocols or standards available	(Antony et al., 2012) (Farahbakhsh et al., 2003) (van Hoof et al., 2003) (Guo et al., 2011) (Kruithof et al., 2001)
Magnetic susceptible particles	• LRV > 6 • both on- and off-line • simple, accurate and quick • specific • low detection limit (Fe_2O_3) • independent of operational mode and location of the broken fiber • nanoparticles: low cost, non-toxic, easily obtainable	• non-continuous • lower detection achievable with higher dilution factor • particles may adhere to membrane surface	(Antony et al., 2012) (Deluhery & Rajagopalan, 2008) (Guo et al., 2010a) (Guo et al., 2010b) (Guo et al., 2011)

Method	Advantages	Disadvantages	Source
	• very low influence on membrane fouling (Fe_3O_4) • variety of detection technologies • no concetnration step needed at industrial scale • appears suitable for large scale drinking water plants • applicable to all MF and UF membrane configurations		
Nanoscale probes	• highly sensitive • on-line • small holes (20 nm) could be detected • real-time detection • low background level (gold) • nonpathogenic, safe to use • high monodispersity • direct relationship between measured data and removal efficiency • level of membrane damage can be predicted • different particle sizes possible	• possible fouling due to pore blocking • physical characteristics different from virus particles • neglecting surface chemistry, infectivity, pathogenesis • high application costs • difficulty to detect particles in permeate (latex) •	(Antony et al., 2012) (Choi et al., 2011) (Guo et al., 2011) (Farahbakhsh et al., 2003) (Gitis et al., 2006)
Fluorescent dye labelled MS2 bacteriophage	• better emulation of viral transport than with non-biological surrogates • simpler and more specific measurement than PFU analysis • uncoupled from background noise • convenient tool for background analysis	• low sensitivity (LRV < 3) • non-continuous • destructive • time- and labour-intensive • necessary safety precautions • sensitivity limited by analytical equipment	(Gitis et al., 2002) (Gitis et. al., 2006)
Stabilised gold and silver nanoparticles	• High sensitivity (LRV ≤ 4.5 for UF) • Simple, quick, safe • Economic • Low labour requirments • Particles may be recovered • Extremely low background level • Possibility to use onsite techniques • Potential for use in water reuse systems • Applicable to MF to RO	• different physical characteristics than viruses • surface chemistry, vitality and pathogenesis of the target organism not considered • may cause cake fouling • needs validation at industrial scale	(Antony et al., 2014) (Gitis et al., 2006) (Chen, 2004)

Method	Advantages	Disadvantages	Source
Membrane Integrity Sensor	• low cost, sensitive, compact • on-line • rapid response • permits adjusting in order to maximise sensitivity • no fouling problems	• stability and sensitivity strongly dependent on membrane characteristics and operating parameters	(Krantz et al., 2011)
Binary gas integrity test	• LRV > 6 • non-destructive • independent of membrane properties • higher sensitivity than bubble point and liquid-liquid porosimetry for small numbers of small defects	• off-line • only realised in lab-scale so far	(Giglia & Krishnan, 2008) (Guo et al., 2010b)
Liquid porosimetry technique	• highly sensitive • reliable and non-destructive • low intrusion pressures necessary • independent of membrane and fluid properties • easy to flush out • accepted reagents for pharmaceutical applications • pre- and post-use membrane integrity • economically and practically feasible • measurement of only one intrusion phase flow rate at one specific transmembrane pressure necessary	• off-line	(Guo et al., 2010b) (Phillips & DiLeo, 1996)

4.2 Sensitivity of particle-based tests

Table 12 Sensitivity of integrity monitoring techniques

Method	Measuring equipment	Membrane	Mode	Detection limit*/sensitivity	Detection time	Source
Turbidity monitoring	Laser turbidimeter	---	---	0.001 NTU	---	(Farahbakhsh et al., 2003)
	Laser turbidimeter plus % RSD (relative standard deviation of turbidity)	Pall / Ahshadi PVDF	---	100 x more sensitive than laser turbidity; 1 of 6'400 fibres	< 45 min.	(Naismith, 2005)
	HACH 1720 C	Aquasource	cross-flow	1 of 4’000 with PAC added to the feed	---	(Glucina et al., 1997)
Particle counting	HIAC/ROYCO MC-80	X-Flow	dead-end	1 of 114'000	---	(Panglisch et al., 1998)
	HIAC-ROYCO (8000A; HLRD 150) / MET ONE (211W; LS211)	Aquasource	cross-flow	1 of 40’000 with PAC added to the feed	---	(Glucina et al., 1997)
Challenge tests						
MS2 Phage		Aquasource	dead-end	No holes < 60–200 µm		(Brehant et al., 2010)
MS2 fluorescent-dye-labeled bacteriophages (28nm)	PFU counting	Flat-sheet : Spectrum CA-0.5, CA-5, CA-10, CE-20, FuMA-Tech PVDF-55, PES-15, Sterlitech PES-20	---	Min. 10^6 PFU/ml	<< 1 min.	(Gitis et al., 2006)
Citrate-stabilised gold nanoparticles (12 nm)	HIAC-ROYCO 8011 (not sufficiently sensitive)			Min. 0.2 µg/l		
Thiol-stabilised gold nanoparticles (15 nm)						
Paramagnetic particles (MSP)	Particle collection column	0.6 µm PC track-etched	---	6.3 x 10^5 particles/ml	---	(Deluhery & Rajagopalan, 2008)

Method	Measuring equipment	Membrane	Mode	Detection limit*/sensitivity	Detection time	Source
Magnetic nanoparticles	Magnetic susceptibility meter Kappabridge (MFK1-FA sensor)	Norit		0.001 x 10^-6 SI unit --> one broken fibre in 5'000'000 hollow fibre		(Guo et al., 2010a)
	Magnetic susceptibility meter Bartington (MS2B sensor)			0.2 - 0.3 ppm Fe3O4 --> 1 broken fibre in 500'000 hollow fibres		

* Detection limit always dependent on resolution of particle counter

Table 13 Number of particle counters necessary per membrane area

Membrane manufacturer	Filtration mode	Feed concentration	Membrane area/particle counter	Source
X-Flow	dead-end, inside → out	7’000 particles/mL	22 m²	(Panglisch et al., 1998)
X-Flow	dead-end, inside → out	50'000 particles/mL	154 m²	(Panglisch et al., 1998)
X-Flow	dead-end, inside → out	200'000 particles/mL	616 m²	(Panglisch et al., 1998)
X-Flow	dead-end, inside → out	500'000 particles/mL	1’562 m²	(Panglisch et al., 1998)
Memtec	dead-end, outside → in	200'000 particles/mL	ca. 18 m²	(Adham et al., 1995)
Aquasource	dead-end, inside → out	200'000 particles/mL	385 m²	(Glucina et al., 1997)
Aquasource	cross-flow, inside → out	1’000 particles/ml	ca. 620 m²	(Glucina et al., 1997)
Aquasource	cross-flow, inside → out	10’000 particles/ml	>> 1’540 m²	(Glucina et al., 1997)

According to Panglisch et al. (1998), the retention R of particles dependent on the ratio of defect and intact capillaries kd can be expressed as

$$R = exp(-5.74 \cdot k_d)$$ Equation 2

for an X-Flow membrane. This expression is independent of trans-membrane pressure, feed concentration and locality of defect.

5 References

DVGW (2013) Technische Regel – Arbeitsblatt DVGW W 213-5 (A) Oktober 2013. Filtrationsverfahren zur Partikelentfernung; Teil 5: Membranfiltration

Adams, N., & Côté, P. (2005). An Evaluation of Membrane Integrity Monitoring Methods for Micro- and Ultrafiltration Systems. Proceedings of the 10th AMK (16th-17th March 2005, Aachen), 11-22.

Adham et al. (1995). Assessing integrity. Journal AWWA 87(3), S. 62.

Adham, S., Jacangelo, J., & Laîné, J.-M. (1995). Low-pressure membranes: assessing integrity. Journal - American Water Works Association 87(3), S. 62-75 .

Antony, A., Blackbeard, J., & Leslie, G. (2012). Removal Efficiency and Integrity Monitoring, Techniques for Virus Removal by Membrane Processes. Critical Reviews in Environmental Science and Technology, 42, S. 891–933.

Antony, A., Blackbeard, J., Angles, M., & Leslie, G. (2014). Non-microbial indicators for monitoring virus removal by ultrafiltration membranes. Journal of Membrane Science 454, S. 193–199.

AWWARF. (2000). Assessment and development of low-pressure membrane integrity monitoring tool. Denver: RFP 2681, AWWA Research Foundation.

Bennett, A. (2008). Drinking water: Pathogen removal from water – technologies and techniques. Filtration & Separation 45(10), S. 14–16.

Brehant, A., Glucina, K., Moigne, I. L., & Laine, J.-M. (2010). Risk management approach for monitoring UF membrane integrity and experimental validation using Ms2-phages. Desalination 250, S. 956–960.

Chen, G. (2004). Electrochemical technologies in waste water treatment. Separation Purification Technology 38, S. 11-41.

Childress A, Le-Clech P, Daugherty JL, Chen C, Leslie GL (2005) Mechanical analysis of hollow fiber membrane integrity in water reuse applications. Desalination 180 (2005) 5-14

Chipps, M. J. et al. (1995). Rapid gravity filter management. Symposium on Sensors & Sensitivity: Water Quality Control through Instrumentation and Sensors. Chartered Institution of Water & Environmental Management (CIWEM)/SWIG.

Choi, S. H., Yang, J., Suh, C., & Cho, J. (2011). Use of fluorescent silica particles for checking the integrity of microfiltration membranes. Journal of Membrane Science 367, S. 306–313.

Coté, P and Adams, N. (2005). An Evaluation of Membrane Integrity Monitoring Methods for Micro- and Ultrafiltration Systems. In: Proceedings of 10th Aachen Membrane Colloquium, March 16-17, 2005, Aachen, Germany, 11-22

Crittenden JC, Trussell RR, Hand DW, Howe KJ, Tchobanoglous G (2012) Chapter 12-8 Process Design (Membrane filtration). in MHW's Water Treatment: Principles and Design. 3rd Edition, John Wiley & Sons, Inc. Hoboken, New Jersey.

Crozes, G. F., Sethi, S., Mi, B., Curl, J., & Mariñas, B. (2002). Improving membrane integrity monitoring indirect methods to reduce plant downtime and increase microbial removal credit. Desalination 149, S. 493-497.

Deluhery, J., & Rajagopalan, N. (2008). Use of paramagnetic particles in membrane integrity testing. Journal of Membrane Science 318, S. 176–181.

DWI (2002) CRYPTOSPORIDIUM IN WATER SUPPLIES: GUIDANCE ON LOW-PRESSURE MEMBRANE INTEGRITY MONITORINDWI. Information Letter 3/2002. Letter of the Drinking Water Inspectorate to the Board Level Contacts in Water Service Companies and Water Companies in England and Wales.

Fane, A., Krantz, W., Tan, E., Sim, V., Nyut, W., Wong, F.-S., et al. (2010). Membrane Integrity Sensor. US patent application No. 61/408753 filed on November 1, 2010.

Farahbakhsh, K., & Smith, D. (2004). Estimating air diffusion contribution to pressure decay during membrane integrity tests. Journal of Membrane Science 237 (1–2), S. 203–212.

Farahbakhsh, K., Adham, S. S., & Smith, D. W. (2003). Monitoring the Integrity of Low-Pressure Membranes. Journal American Water Works Association 95(6), S. 95-107.

Franklin, B., et al. (2000). Using Membranes as a Cryptosporidium Barrier for the World's Largest Municipal Ultrafiltration Plant. Proceedings of the AWWA Annual Conference. Denver.

Giglia, S., & Krishnan, M. (2008). High sensitivity binary gas integrity test for membrane filters. Journal of Membrane Science 323, S. 60–66.

Gitis, V., Adin, A., Nasser, A., Gun, J., & Lev, O. (2002). Fluorescent dye labeled bacteriophages—a new tracer for the investigation of viral transport in porous media: 1. Introduction and characterization. Water Research 36(17), S. 4227–4234.

Gitis, V., Haught, R. C., Clark, R. M., Gun, J., & Lev, O. (2006). Nanoscale probes for the evaluation of the integrity of ultrafiltration membranes. Journal of Membrane Science 276, S. 199–207.

Glucina, K., Do-Quang, Z., & Laîné, J. M. (1997). Assessment of a particle counting method for hollow fiber membrane integrity. Desalination 113, S. 183-187.

Guibert, D., & Colling, A. (2011). Direct membrane integrity testing: Impact of parameter selection on log removal value calculations. Desalination 272, S. 174–178.

Guo, H., Wyart, Y., Perot, J., Nauleau, F., & Moulin, P. (2010a). Application of magnetic nanoparticles for UF membrane integrity monitoring at low-pressure operation. Journal of Membrane Science 350, S. 172–179.

Guo, H., Wyart, Y., Perot, J., Nauleau, F., & Moulin, P. (2010b). Low-pressure membrane integrity tests for drinking water treatment: A review. Water Research 44, S. 41-57.

Guo, H., Wyart, Y., Perot, J., Nauleau, F., & Moulin, P. (2011). Magnetic nanoparticles for UF membrane integrity: Industrial scale. Membrane Water Treatment 2(1) , S. 00-00.

Hall, T., & Croll, B. (1996). The UK approach to Cryptosporidium control in water treatment. Boston, Massachusetts, USA: American Water Works Association Water Quality Technology Conference.

Henderson, R. K., Baker, A., Murphy, K. R., Hambly, A., Stuetz, R. M., & Khan, S. J. (2009). Fluorescence as a potential monitoring tool for recycled water systems: A review. Water Research 43, S. 863-881.

Hofmann, F. (1984). Integrity testing of microfiltration membranes. Journal of Pharmaceutical Science and Technology 38(4), S. 148-159.

Irwin R, Cooper N, Tsuchihashi R, van Herk M, Smith D (2014) Validation of UF Membrane plants for water recycling in Victoria, Australia. Proceedings of the AWWA/AMTA Membrane Technology / Conference & Expositions, 10-14 March 2014, Las Vegas 281-299 (

Jackson, C. (2001). Review of the adequacy of existing proposals for membrane integrity monitoring. DWI ref 43/2/159.

Johnson, W. T. (1997). Automatic monitoring of membrane integrity in microfiltration systems. Desalination 113, S. 303-307.

Johnson, W. T. (1998). Predicting log removal performance of membrane systems using in-situ integrity testing. Filtration & Separation 35(1), S. 26-29.

Kirby, R., Blakeley, P., & Spencer, D. (1998). The development of a fine particle monitor based on the method of dynamic light extinction. Filtration & Separation 35(1), S. 73–77.

Krantz WB, Yeo A, Sim V, Fane AG (2010) The Integrity Sensor – A Novel Instrument for Assessing the Performance of Pre-Filtration Membranes. Proceedings of 2010 AIChE Annual Meeting, 8-12 November 2010. Originally presented on: 11/9/2010 13:30:00 - 13:50:00. Available at: http://www3.aiche.org/proceedings/Abstract.aspx?ConfID=Annual-2010&GroupID=1503&SessionID=15553&PaperID=205749

Krantz, W. B., Lin, C. S., Sin, P. C., Yeo, A., & Fane, A. G. (2011). An Integrity Sensor for assessing the performance of low pressure membrane modules in the water industry. Desalination 283, S. 117–122.

Krause, S. (2012). Ultrafiltration für kleine Trinkwasseraufbereitungsanlagen. München: Oldenbourg Industrieverlag.

Kruithof, J., Kamp, P., Folmer, H., Nederlof, M., & van Hoof, S. (2001). Development of a membrane integrity monitoring strategy for the UF/RO Heemskerk drinking water treatment plant. Water Science and Technology: Water Supply 1, S. 261-271.

Laîné, J., Glucina, K., Chamant, M., & Simonie, P. (1998). Acoustic sensor: a novel technique for low pressure membrane integrity monitoring. Desalination 119, S. 73-77.

Landsness, L. B. (2001). Accepting MF/UF technology—making the final cut. San Antonio, Texas (USA): Paper presented at the American Water Works Association Membrane Technology Conference.

Langlet, J., Gaboriaud, F., & and Gantzer, C. (2007). Effects of pH on plaque forming unit counts and aggregation of MS2 bacteriophage. Journal of Applied Microbiology 103, S. 1632–1638.

Liu, C. (2012). Integrity Testing for Low-Pressure Membranes. American Water Works Association Denver, USA. ISBN 1-58321-793-2

Meltzer, T. H., Madsen Jr., R. E., & Jornitz, M. W. (1999). Considerations for Diffusive Airflow Integrity Testing. PDA Journal of Pharmaceutical Science & Technology 53(2), S. 56-59.

Naismith, J. (2005). Membrane integrity - direct turbidity measurement of filtrate from MF membrane modules at an operating potable water treatment plant. Desalination 179, S. 25-30.

Nederlof, M. M., Folmer, H. C., Verdouw, J., & Heijman, B. (1997). Integrity of membrane elements, vessels and systems. Desalination 113, S. 179-181.

Panglisch, S., Deinert, U., Dautzenberg, W., Kiepke, O., & Gimbel, R. (1998). Monitoring the integrity of capillary membranes by particle counters. Desalination 119, S. 65-72.

Pearce, G. (2007). Water and wastewater filtration: Membrane module format. Filtration & Separation 44(4), S. 31–33.

Phattaranawik, J., Fane, A. G., & Wong, F. S. (2008). Novel membrane-based sensor for online membrane integrity monitoring. Journal of Membrane Science 323, S. 113–124.

Pipe-Martin, C. (2010). Challenge Testing Water Recycling Plants - Making Sure We Get the Bugs Out. 4th Annual WIOA NSW Water Industry Engineers & Operators Conference, (S. 99-105). St Stanislaus' College, Bathurst.

Radziemski, L. J., & A.Cremers, D. (1989). Laser Induced Plasmas and Applications. New York, USA: Marcel Dekker.

Rajagopalan, N., Qi, S., & J.P. Pickowitz. (2001). Field evaluation of ceramic microfiltration membrane in drinking water application. In Proceedings of the American Water Works Association 2001 Membrane Technology Conference. Texas.

Randles, N. (1996). Large scale operating experience in membrane for water and waste water reclamation . Desalination 108, S. 205-211.

Schroeder, H., & DeLuca, P. (1980). Theoretical Aspects of Sterile Filtration and Membrane Integrity. Pharmaceutical Technology 4(11), S. 80-85.

Troester, M., Lipp, P., Sacher, F., Brauch, H.-J., & Hofmann, T. (2014). Laser-Induced Breakdown-Detection for reliable online monitoring of membrane integrity. Journal of Membrane Science 466, S. 313–321.

US EPA (2001) Low pressure membrane filtration for pathogen removal: application, implementation and regulatory issues

US EPA (2005). Membrane Filtration Guidance Manual. EPA 815-R-06-009, US Environmental Protection Agency Office of Water.

USPA (2010) Method for detecting membrane module flaw with the application of fluorescent particles. United States Patent Application, Pub. No US 2010/0060887 A1, March 2010

US EPA (2012). Drinking Water Pathogens and Their Indicators. Von US Environmental Protection Agency: derived on 7 July 2014 from: http://www.epa.gov/enviro/html/icr/gloss_path.html#giardia abgerufen

van Hoof, S. v., Broens, L., Nahrstedt, A., Panglisch, S., & Gimbel, R. (2003). Development of a new integrity testing system. Water Science & Technology: Water Supply 3(5), S. 101–108.

Vieira, M. G., Silva, M. A., Santos, L. O., & Beppu, M. M. (2011). Natural-based plasticizers and biopolymer films: A review. European Polymer Journal 47(3), S. 254–263.

Waibel, P., Jornitz, M., & Meltzer, T. (1996). Diffusive Airflow Integrity Testing. Journal of Pharmaceutical Science and Technology 50(5), S. 311-316.

6 Annex

The impact of a liquid leak on the performance of a membrane can be calculated as (Giglia & Krishnan, 2008):

$$\Delta LRV = LRV_I - \log\left(\frac{c_f V_T}{c_p V_I + c_f V_d}\right) \quad \text{(Equation 3)}$$

LRV_I: LRV of the integral (defect free) portion of the membrane

V_I: volume of feed passing through integral part of membrane

V_d: Volume passing through defect

V_T: total volume of feed passing through the filter

c_p: permeate concentration

c_f: feed concentration

To accurately correlate and predict particle passage without influencing the permeate stream by interruption of operation or spiking of appropriate-sized particles to increase test sensitivity, the log removal value can be approximated by the following equation (Phillips & DiLeo, 1996):

$$LRV \approx c_f \cdot \left(\frac{Q_{tot}}{Q_{ds}}\right) \quad \text{(Equation 4)}$$

Q_{tot}: total amount of flow through membrane sample under a given set of operating conditions

Q_{ds}: amount of flow through membrane pores equal in size or larger than d_s

c_f: feed concentration

Equation 3 holds under the assumption of rigid, spherical particles of diameter ds and unhindered transport of solutes through membrane pores (Phillips & DiLeo, 1996).

Under the assumption that the particles of interest are completely rejected by the membrane and can pass freely through leaks, the sensitivity of a membrane system can be estimated by equation 4 (Adams & Coté, 2005):

$$LRV_e = \log_{10}\left(\frac{Q_p}{CF \cdot Q_{breach}}\right) \quad \text{(Equation 5)}$$

LRV_e: estimated log removal value

Q_p: permeate flow rate of the membrane unit

Q_{breach}: flow through breaches

CF: concentration factor (particles of interest in membrane tank relative to feed water).

If pressure decay or vacuum decay are measured, equation 4 can be adapted using a correction for diffusion and the Hagen-Poiseuille model (for laminar flow) as conversion to a water flow rate to give a reliable estimate of the LRV (Adams & Coté, ???):

$$LRV_e = \log_{10}\left(\frac{Q_p}{CF \cdot Q_{air}} \cdot f_1 \cdot f_2\right) \quad \text{(Equation 6)}$$

with

$$Q_{air} = PDR\ (or\ VDR) \cdot \frac{V_{system}}{P_{atm}} \quad \text{(Equation 7)}$$

$$f_1 = \frac{\mu_{water}}{\mu_{air}}$$ (Equation 8) and

$$f_2 = \frac{P_{u,test}^{\ 2} - P_{d,test}^{\ 2}}{2 \cdot P_{atm} \cdot TMP}$$ (Equation 9)

Q_{air}: volumetric air flow rate corrected for diffusion
f_1: air-to-water conversion factor
f_2: transmembrane pressure conversion factor
μ_{water}: viscosity of water during filtration [Pa s]
μ_{air}: viscosity of air during the test [Pa s]
$P_{u,\,test}$: upstream pressure, average test pressure for pressure test or P_{atm} for vacuum test
$P_{d,test}$: downstream pressure, average test pressure for vacuum test static head must be considered for both pressure and vacuum test, if water is on downstream side
P_{atm}: atmospheric pressure
TMP: transmembrane pressure