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Review

Low-pressure membrane integrity tests for drinking water treatment: A review

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ABSTRACT

Low-pressure membrane systems, including microfiltration (MF) and ultrafiltration (UF) membranes, are being increasingly used in drinking water treatments due to their high level of pathogen removal. However, the pathogen will pass through the membrane and contaminate the product if the membrane integrity is compromised. Therefore, an effective on-line integrity monitoring method for MF and UF membrane systems is essential to guarantee the regulatory requirements for pathogen removal. A lot of works on low-pressure membrane integrity tests have been conducted by many researchers. This paper provides a literature review about different low-pressure membrane integrity monitoring methods for the drinking water treatment, including direct methods (pressure-based tests, acoustic sensor test, liquid porosimetry, etc.) and indirect methods (particle counting, particle monitoring, turbidity monitoring, surrogate challenge tests). Additionally, some information about the operation of membrane integrity tests is presented here. It can be realized from this review that it remains urgent to develop an alternative on-line detection technique for a quick, accurate, simple, continuous and relatively inexpensive evaluation of low-pressure membrane integrity. To better satisfy regulatory requirements for drinking water treatments, the characteristic of this ideal membrane integrity test is proposed at the end of this paper.

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1. Introduction

Low-pressure membrane systems, including microfiltration (MF) and ultrafiltration (UF) membranes, are being increasingly used for the drinking water treatment. The global installed volume of low-pressure membranes has grown at an impressive rate during the last 10 years, and 60% of applications are for drinking water (David et al., 2008). This rapid growth of water treatment low-pressure membranes, both in terms of capacity and number of installations, is mainly due to their high level of pathogen removal, such as viruses, bacteria and protozoa cysts (*Giardia* and *Cryptosporidium*). Complete removal of coliform bacteria, *Giardia* spp. and *Cryptosporidium* spp. by MF and UF membranes has been demonstrated by many researchers (Adham and Jacangelo, 1994; Edwards et al., 2001; Freeman et al., 1996; Hirata and Hashimoto, 1998; Jacangelo et al., 1991). Compared to MF, UF technology is able to remove viruses due to its low cut-off and thus it can take the place of the disinfection step. In addition to concerns over microbial contaminants, low-pressure membranes are becoming more attractive for the drinking water industry due to some other reasons, such as stricter regulatory requirements, operation easiness, minimum staffing requirements, competitive cost and independence of water source quality.

However, pathogens may pass through membranes and enter the public water supply system if the membrane system integrity is compromised (e.g., broken fiber, fiber degradation, O-ring failure, etc.). Failure of membrane fibers or sheets results from four main reasons: (1) chemical corrosion such as oxidation; (2) faulty installation and maintenance; (3) membrane stress and strain from operating conditions, such as backwashing or excessive movement due to vigorous bubbling; and (4) damage by sharp objects not removed by pretreatment. Zondervan et al. (2007) concluded that the fouling status of a membrane, the number of applied back pulses and the combination of these two factors are significant aging factors and responsible for the membrane failure. Accurate and efficient integrity tests of the membrane system can guarantee the quality of filtered products. Therefore, it is important for a membrane plant to perform an effective membrane integrity monitoring as well as reachable flux, membrane retention and membrane permeability recovery. An increasing number of regulatory agencies in North America and Europe (e.g., U.S. EPA in United States, DWI in United Kingdom, etc.) require membrane water treatment utilities to conduct membrane integrity monitoring on a regular basis.

Membrane integrity tests are specific for the membrane type and depend on the membrane manufacturer and membrane system supplier. Generally, membrane integrity monitoring techniques are divided into two main groups: direct methods and indirect methods. Direct methods refer to tests directly applied to the membrane or the membrane module, i.e., pressure decay test (PDT) (Adham et al., 1995; Johnson, 1997, 1998), diffusive air flow (DAF) test (Adham et al., 1995; Johnson, 1997, 1998), bubble point test (Randles, 1997), acoustic sensor test (Adham et al., 1995; Laîne et al., 1998), liquid–liquid porosimetry test (DiLeo and Phillips, 1994, 1995; Phillips and Dileo, 1996; Gekas and Zhang, 1989) and binary gas integrity test (Giglia and Krishnan, 2008). Indirect methods refer to tests applied to water quality parameters in the permeate solution, i.e., particle counting (Adham et al., 1995; Landsness, 2001; Panglisch et al., 1998), particle monitoring (Adham et al., 1995), turbidity monitoring (Adham et al., 1995; Banerjee et al., 1999, 2001), and different surrogate challenge tests such as spiked integrity monitoring system – SIM™, microbial challenge tests and some new surrogate challenge tests (Van Hoof et al., 2001; Kruithof et al., 2001; Brehant et al., 2008; Gitis et al., 2006a,b; Trimboli et al., 2001; Moulin, 2008; Rajagopalan et al., 2006; Deluhery and Rajagopalan, 2008). Among these methods, the PDT and DAF test are the most frequently used in drinking water treatments due to their advantages of simplicity, low maintenance, reliability and high sensitivity to detect membrane breaches. In addition, particle counting, turbidity monitoring and routine microbial

analysis are the most frequently used indirect methods (Crozes et al., 2002). All these methods have their own advantages and disadvantages. For example, the PDT and DAF test are sensitive enough to meet the regulatory requirements of drinking water treatment, but they are conducted off-line and provide no measurement of filtrate water quality. It should be noted that on-line monitoring is very important to guarantee the filtered quality of the membrane system. Plants that only rely on pressure integrity tests (e.g., PDT and DAF test) are likely to continue their operations beyond the point where their membrane integrity is compromised. In contrast, indirect methods, such as particle counting and turbidity monitoring, are very convenient for routine qualitative monitoring. Changes in filtrate quality are monitored by comparing measured data with a previously established baseline level. These indirect tests are able to detect membrane integrity in continuous and on-line mode. In addition, the same indirect methods and testing instruments can be applied to any membrane system, independent of membrane manufacturers, system configurations, and other parameters intrinsic to the system. They are also likely to remain applicable to any new system that is developed. But the main problem of these two techniques is their low detection sensitivity. It has been reported that these two tests failed to detect the individual fiber pin-holes in membrane system occurring as a result of challenging operating conditions (Walsh et al., 2005). Furthermore, current membrane integrity tests in practical drinking water treatments are not sufficiently sensitive to detect nanometric breaches for capital and operational reasons. Then they are not effective to meet removal requirements for viruses (~20 nm). The absence of a reliable, sensitive, and on-line detection method for monitoring membrane integrity is currently hampering more significant virus removal credits of UF membranes. Therefore, it is necessary to develop an alternative on-line detection technique for an accurate, quick, simple, and affordable evaluation of the UF membrane integrity for drinking water plants.

In this paper, different methods for monitoring low-pressure membrane integrity are reported. In addition to commonly used tests (pressure-based tests, acoustic sensor test, particle counting, particle monitoring, turbidity monitoring) which have been summarized in some previous reports (U.S. EPA, 2001, 2003), other integrity monitoring technologies which are mostly in lab-scale study so far are also presented here (liquid–liquid porosimetry, binary gas integrity test, new membrane-based sensor and different surrogate challenge tests). Pointing out advantages and disadvantages of these methods, it can be realized that the development of a new membrane integrity test up to the level of industrial applications is still important and urgent.

2. Sensitivity evaluation for membrane integrity tests

According to the DWI requirements, the continuous removal or retention of particles greater than 1 μm should be obtained during the membrane operation for drinking water treatments (Jackson, 2001). This can be interpreted as an absolute removal, which means that *Giardia* (~6–20 μm) and *Cryptosporidium* (~4–6 μm) are required for a complete removal. Considering that it is not practical for a 100% guaranteed removal, generally the drinking water industry measures the pathogen removal efficiency in terms of the log removal value (LRV), which is defined as Eq. (1) (Bennett, 2008):

$$\mathrm{LRV} = \log_{10}\left(\frac{C_f}{C_p}\right) \tag{1}$$

where C_f is the concentration of the retained species in feed solution and C_p is the concentration of the species in permeate solution. Of course, a stated LRV is related to a particular particle size or particle size distribution. When there are membrane failures, the impact of a liquid leak on the retention can be calculated as Eq. (2) (Giglia and Krishnan, 2008):

$$\Delta\mathrm{LRV} = \mathrm{LRV}_1 - \log_{10}\left(\frac{C_f V_T}{C_p V_p + C_f V_d}\right) \tag{2}$$

where LRV_1 is the LRV of the unimpaired portion of membranes, V_p is the feed volume passing through the unimpaired portion of the membrane, V_d is the feed volume passing through the defect and V_T is the total feed volume passing through the membrane. Here, it is assumed that the defect does not partially retain the considered species.

The United States Environmental Protection Agency has specified pathogen removal (or inactivation) rates (*Cryptosporidium*, *Giardia* and viruses) through the enhanced surface water treatment rule (ESWTR), as shown in Table 1 (Faber and Pearce, 2004).

In general, the purposes of membrane integrity tests include verification of high filtered water quality, demonstration of regulatory compliance and detection of equipment/filtration problems. Considering that membrane integrity tests are function of the different membrane suppliers, the American Society for Testing and Materials (ASTM) sub-committee designed the Standard Practice for Integrity Testing of Water Filtration Membrane Systems (ASTM – D6908-03), which describes four integrity test methods (PDT, vacuum decay test, soluble dye test (SDT) and total organic carbon monitoring test (TOCMT)) that can be applied to all membrane systems, regardless of application (Moch and Paulson, 2003).

The sensitivity of a membrane integrity test is very important to guarantee the pathogen removal credit. According to the membrane filtration guidance manual (MFGM) (U.S. EPA, 2003), the sensitivity of membrane integrity tests refers to the maximum log removal value that can be reliably verified by integrity tests associated with a given membrane filtration system. It is expressed in terms of a LRV,

Table 1 – Inactivation/removal requirements for pathogen in portable water(Faber and Pearce, 2004).

Pathogen	Log removal value (LRV)	Percentage removal (%)
Cryptosporidium parvum	2	99.00
Giardia lamblia	3	99.90
Viruses	4	99.99

Based on ESWTR – adapted from Faber and Pearce (2004).

which must be equal to or greater than the required pathogen removal credit. The sensitivity of some other tests can be calculated in different forms. For pressure-based tests, the sensitivity can be expressed as Eq. (3):

$$\mathrm{LRV} = \log\left(\mathrm{VCF}\frac{Q_p}{Q_{breach}}\right) \quad (3)$$

where Q_p is the designed filtrate flow of the membrane unit, Q_{breach} is the flow from the breaches associated with the smallest integrity test response that can be reliably measured and VCF is the volume concentration factor (i.e., the ratio of concentration in the retentate to concentration in the influent). For surrogate challenge tests, the sensitivity can be expressed as Eq. (1). Based on the practical results, Johnson (1998) compared the sensitivity of different integrity tests (PDT, DAF test, particle counting and turbidity monitoring) and ranked them as shown in Fig. 1.

3. Low-pressure membrane integrity tests

3.1. *Pressure-driven tests*

Pressure-driven tests are non-destructive for membrane systems. All pressure-driven membrane integrity monitoring tests, including PDT, DAF test, vacuum decay test and bubble point test, are theoretically similar since they are based on the bubble point pressure concept but they differ in operating protocols and measured parameters. Here, bubble point is typically defined as the minimum pressure required to overcome the capillary forces and surface tension of a liquid in a fully wetted membrane filter and force air flow through the filter pores. Briefly, for a wetted membrane, liquid can be forced out of the filter pores by applying gas pressure. The removal of liquid from the largest pores creates a passage way through which bulk air flow takes place. The minimum pressure at which this bulk flow through the membrane is detected is referred to as the bubble point. The bubble point is related to the diameter of the largest pore or defect, which can be estimated by the capillary equation as Eq. (4) (Farahbakhsh and Smith, 2004):

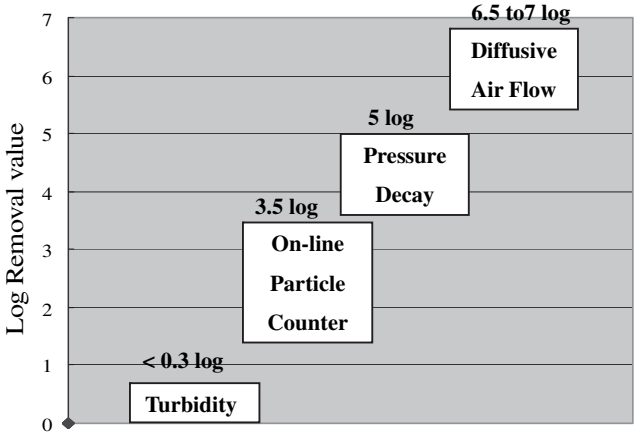

Fig. 1 – Relative sensitivity of various monitoring methods (Johnson, 1998).

$$P = \frac{4k\cos\theta}{d}\sigma \quad (4)$$

where k is the correction factor for the largest pore shape, d is the diameter of the largest pore, θ is the contact angle between the liquid and the membrane and σ is the surface tension of liquid. It can be seen that the air pressure required to force liquid from the pores is inversely proportional to the pore diameter. The larger the pore is, the lower the pressure is required. If the membrane has no defect, any air pressure below the bubble point cannot force water from the membrane pores. In contrast, defects such as holes or broken fibers have a comparatively low bubble point. Assuming that the pore shape factor k is 1 and the liquid/membrane contact angle θ is close to 0 it is possible to calculate the maximum bubble point pressure (Bennett, 2005). For UF membranes (pore size <0.1 μm), the bubble point is generally in the range of 3000–30,000 KPa. For a hole in the membrane comparable to a cyst at 4 μm mean diameter, the bubble point of the membrane is 73 KPa. This means that any hole with a diameter larger than 4 μm can be identified if the test pressure is above 80 KPa. For current PDT and DAF test, a typical test pressure of 100 KPa, which can identify a defect of 2.8 μm, guarantee a complete barrier against pathogens such as *Cryptosporidium* and *Giardia* but not against viruses.

Diffusive air flow through a wetted membrane can be expressed as Eq. (5) modified from the Fick's law of diffusion (Giglia and Krishnan, 2008):

$$Q = \frac{A\varepsilon DS(P_f - P_p)}{\tau L} \quad (5)$$

where Q is the diffusive air flow rate, A is the membrane area, ε is the membrane porosity, D is the gas diffusivity in the liquid, S is the gas solubility coefficient, P_f and P_p are the feed and permeate side pressures respectively, τ is the pore tortuosity and L is the liquid thickness in the membrane. A measured gas flow rate in excess of that predicted by Eq. (5) or higher than a flow rate empirically established for an unimpaired membrane indicates the presence of defects in the membrane. As shown in Eq. (5), the amount of diffused air through a wetted membrane at a given applied pressure is a function of the membrane porous surface area (εA). During a pressure decay test, an oversized defect or hole contributes to a more diffusive air flow because of an increase in membrane porous surface area. It has been demonstrated that the dilution effect observed during pressure decay tests on membranes with large surface can be mainly attributed to air diffusion through intact pores (Giglia and Krishnan, 2008; Farahbakhsh and Smith, 2004). This may produce false–negative results. In other words, membrane breaches will not be detected until the defect is significant enough to produce a noticeable pressure decay rate above that resulting from diffusion. Therefore, accounting for and estimating the air diffusion contribution to pressure decay during a pressure decay test would produce much more reliable and sensitive results.

3.1.1. *PDT*

The main principle of the PDT is based on the measurements of pressure drop in the feed side after draining and pressurizing. PDT can be performed with PDT-filled or PDT-drained

mode. An outline of the PDT-filled involves: (1) draining the water from one side of the membrane; (2) applying pressurized air to the drained side of the fully wetted membrane at a predetermined level below the bubble point; and (3) holding the pressure for a specific time duration (2–10 min) and monitoring the pressure decay. The predetermined pressure, ranging from 20 to 200 KPa (typically 100 KPa during practical operation), directly relates to the defect size under investigation. It also determines the smallest hole that can be detected by the PDT. During the test, a small decrease (e.g., 0.5–1.5 KPa per minute) is considered acceptable due to air diffusion across the intact membrane structure. A faster pressure decrease in excess of a critical threshold indicates a faulty membrane. PDT-drained is similar to ''PDT-filled'', but in this case the module is drained on both sides of the element so that only the membrane is ''wetted''. After the detection of membrane defects, the bubble test or sonic test can be used to locate defects. Bubble tests are performed with a specific test pressure (e.g., 30–50 KPa) in water filled basin or using soap on the outer ends of a membrane element. Bubbles will appear from any defective fiber. Then, the fiber can be isolated and repaired. Sonic tests for defect location will be presented in the Memsure™ process. Generally, the PDT is a standard part of most UF and MF membrane systems and it is highly automated during the operation.

PDT is a reliable method for membrane integrity monitoring but it cannot be operated on-line and continuously. PDT appears to be very sensitive to detect leaks and integrity breaches. Adham et al. (1995) reported that a considerable loss of pressure was observed due to a needle puncture of 0.6 mm internal diameter in the lumen wall of one out of over 22,000 fibers in a membrane module. Johnson (1998) determined that this method is able to detect a single broken fiber in a membrane array containing over one million fibers. PDT is able to detect changes in the membrane integrity at a level up to 4.5–5 of *Giardia* or *Cryptosporidium* log removal, independent of feed water quality and without relying on filtered water quality monitoring such as turbidity and particle counters. However, the sensitivity of PDT is limited by the minimum detectable excess air flow. It has been shown that the PDT effectiveness for the detection of membrane integrity changes is affected by the membrane surface (U.S. EPA, 2001). This phenomenon becomes more important as the membrane surface increases. It has been reported that, for a rack of 90 modules of the same type of membrane tested by Adham et al. (1995), PDT results have exhibited significant variation from the case of intact membrane system until six fibers were cut (Landsness, 2001). Hence, the greater the number of fibers tested, the more sensitive the pressure transducers need to be so as to differentiate between background noise spread out over a larger membrane area and losses due to integrity breaches. Additionally, PDT may sometimes cause membrane breaks during the test and may yield false–positive results due to a non-fully wetted membrane (U.S. EPA, 2003).

3.1.2. DAF test

DAF test is fundamentally similar to the PDT. However, rather than measuring pressure decay rate, DAF test measures the diffused gas filtrate flow or displaced water flow through the fully wetted membrane pores when applying a constant feed side gas pressure below the bubble point of the selected whole size. The most commonly cited DAF tests refer to air diffusion measurement (Cheryan, 1998). But one associated difficulty is the sensitivity of air diffusion rates to temperature, which will directly affect the air viscosity. Seasonal variations in temperature along a year may cause fluctuations in the results of this test. The other DAF test, which measures the displaced water flow, has been described in a 1997 report by the American Water Works Association Research Foundation (AWWARF) (Jacangelo et al., 1997). Due to its advantages of easiness and accuracy of measurements, the DAF test measuring displaced water flow was widely used in practical membrane water treatment plants. Overall, the DAF test shows more sensitivity to detect changes in membrane integrity than the PDT (Trimboli et al., 2001). It is able to detect integrity changes at levels >6 LRV. Despite of its increased sensitivity, the DAF test requires some additional pipe work and fittings in order to measure displaced liquid flow rate. The DAF test is subject to the same disadvantages as the PDT. In addition, it is not included as part of the standard equipment in most MF and UF systems and thus it has not been automated.

3.1.3. *The Memsure™ process*

As a typical example of application of PDT and DAF test, the Memsure™ process, which was developed by Simens–Memcor for industrial application, is an integrity monitoring technique for continuous microfiltration (CMF) system (Johnson, 1997 and 1998). This technique involves three key steps: (1) monitoring the integrity using the Memsure™ PDT or the Memsure™ DAF test; (2) identification of leaks using sonic analysis (The Memcor® Sonic Analyzer); and (3) isolation of faulty modules using module isolating valves for later repair. Generally, the lumen is pressurized at 100 KPa during the Memsure™ PDT and the Memsure™ DAF test, the test duration being around 5 min. One significant advantage of this PDT test is that it requires no additional equipment and it makes use of the CMF air backwash system. Thus the test can be automatically conducted by the control system, including the logging of results and generating an alarm when results are beyond limits. The Memcor® Sonic Analyzer is a sensitive listening device tuned to the sound made by bubbles escaping through fiber defects or leakages. Air leaking through defects creates a distinctive sound that can be picked up by the device and displayed as a sound level on the front of the unit. In this way the identified modules can be isolated using the in-built module isolation valves at the top and bottom of each Memcor® CMF module to be removed and repaired on site later on.

3.1.4. *Bubble point test*

A bubble point test is a test designed to determine the bubble point pressure of the membrane. Theoretically, the bubble point pressure decreases with the presence of breaches in the membrane. When a bubble point test is conducted, the module to be tested is first removed from the rack. The internal shell of the module is then drained and pressurized. The membrane must be wetted uniformly. A dilute surfactant solution is applied to the open ends of the membrane fibers at the end of the module. When reaching the bubble point pressure, the liquid is expelled from one or more

passageways, establishing a path for the bulk flow of air. As a result, a steady stream of bubbles should be seen in the surfactant solution. Explicitly, the formation of vigorous bubbles in the surfactant solution can be traced to locate specific leaking fibers.

In a bubble point test, the excessive gas flow originating from a single or very small defect may not be identified from that coming from the unimpaired part of the membrane, thereby masking the actual bubble point. Another limitation of this test is that the pressure required to reach the bubble point may be impractically high for UF membranes, even with the use of low surface tension fluids. For example, the air–water bubble point for a membrane with a pore size of 20 nm will be in excess of 3500 KPa (Giglia and Krishnan, 2008). Such high bubble point pressure is generally unattainable in practical operation and is likely to lead to membrane compaction and rupture. Therefore, this method is not suitable for UF membrane integrity monitoring. Large-scale operating experience showed (Randles, 1997) that Memcor® membrane integrity test, which is based on the bubble point test, was very effective for monitoring membrane integrity of the Memcor® CMF process. It is able to detect a single compromised membrane fiber in around 12 million fibers and it offers high sensitivity to guarantee bacterial removal at levels of 6 LRV. So far, the bubble point test is generally employed in conjunction with the PDT rather than as a separate and independent gauge of the membrane integrity. In other words, once a membrane integrity problem is detected, the bubble point test can be applied to identify the compromised fiber(s) following its removal from the rack. Used in this manner, the LRV of the test is limited to that of the PDT.

3.1.5. *Vacuum decay test*

The vacuum decay test is a variation of the PDT where a vacuum is applied to the drained side of a fully wetted membrane and the vacuum pressure decay rate is monitored. This method can be used to monitor UF and MF membrane integrity but it is rarely performed in practical operation for membrane drinking water plants. So far, this method, based on ASTM Standard D3923-2 and D6908-3, is used for FILMTEC membranes to detect leaks or confirm the integrity of FILMTEC RO and NF elements after they have been in operation (FILMTEC Membranes). A vacuum decay test is able to identify leaking elements or O-rings within a short time and it is a non-destructive test. This test is useful as a screening procedure and generally it is not intended as a mean of absolute verification of leaks.

3.2. *Binary gas integrity test*

Assuming that the gas is completely mixed on both sides of the membrane when two gases permeate through a membrane, the composition of the permeate gas can be calculated from the ratio of diffusive flow rates of the two components and the inlet side composition (Weller and Steiner, 1950). The composition of the permeate gas is independent of membrane thickness, tortuosity, porosity and area. It is also independent of the pressure difference across the membrane but dependent on the pressure ratio. In addition, the permeate composition depends on the feed side composition and the permeability ratio of these two gases. In order to maintain a constant feed side composition, a constant sweep flow has to be applied. A binary gas integrity test for low-pressure membranes has recently been developed by Giglia and Krishnan (2008). This test uses a binary gas mixture and is based on the gas permeability difference between the two components of a gas mixture through the liquid layer of a wetted membrane. In contrast to the single gas diffusion test, the binary gas test primarily relies on the measurement of downstream gas composition rather than downstream flow rate. The presence of membrane defects results in an elevated concentration of the slower permeating gas in the permeate stream. In an unimpaired membrane, the permeate composition can be predicted from known operating conditions and the transport properties of gases permeating through the liquid layer. A change in gas composition across the membrane indicates the presence of defects or opened pores. The sensitivity of the binary gas test mainly depends on the selectivity (permeabilities ratio) of these two gases through the liquid layer. High selectivity of the two gases results in high detection sensitivity. In addition, the selection of gas concentration in the mixture is influenced by a number of factors, including easiness of composition measurement, gas flow rate through the membrane and economic considerations. Naturally, the permeability of gases through the liquid layer – generally water – is independent of the membrane type. Due to the high selectivity of CO_2/C_2F_6 pair through water of about 1000, a 90/10 CO_2/C_2F_6 molar concentration was selected to test the membrane integrity under a pressure of 345 KPa by Giglia and Krishnan (2008). At this level of concentration, the high permeability of the CO_2 enables a convenient flow and composition measurement even for relatively small membrane areas as small as 3 cm^2. Evidently, C_2F_6 is toxic and the storage of C_2F_6 is not so simple during practical operation. Because the binary gas test has lower sensitivity to membrane porosity, liquid layer thickness and membrane area, integral devices exhibit a relatively narrow range of test values, resulting in a superior defect signal-to-noise ratio. As a result, the binary gas test, which can provide an LRV assurance greater than 6, shows higher defect detection sensitivity than the single gas test. However, this method is conducted off-line and reported just in a lab-scale study.

3.3. *Acoustic sensor test*

An acoustic sensor analysis was at first conducted manually by applying an accelerometer (an instrument used to detect vibration) in one or two locations on each membrane module (Adham et al., 1995). Using headphones, an operator listens to vibrations generated by leaking air. This analysis is effectively administered by a skilled and experienced operator and it is to some extent more subjective than other forms of integrity tests. Yet, it cannot monitor membrane integrity continuously. In practical operation, acoustic sensor analysis is usually used to identify the impaired module and the breaches location which have been detected by the PDT. However, this method has the potential to eliminate the subjectivity and to be developed into an on-line and continuous membrane integrity test if it is automated and computerized.

Such an automated acoustic system to monitor membrane integrity was described by Laîne et al. (1998). Based on the hydrophonic sensor technique, the acoustic integrity monitoring (AIM) technique measures the noise signal (i.e., pressure fluctuation) in a given frequency range (e.g., 280–650 Hz). A distinctive noise signal is created due to a compromised fiber. The main advantage of a hydrophonic sensor is that the membrane integrity is continuously monitored during filtration (on-line sensor) independently of feed water quality. It has been reported that AIM technology is able to ensure more than 6 log removal of viruses. However, experimental results from the full-scale plants showed that the acoustic detection depends significantly on background noise and flow rate. The higher the background noise is, the lower the sensor response is. In the same way, the higher the flow rate is, the higher the noise generated by a compromised fiber is. Laîne et al. (1998) designed an AIM prototype to check 28 modules. This prototype includes 28 acoustic sensors, two collectors and one processor. The total cost for a full system was about $7000, i.e., $250 per module showing that AIM technology is economically competitive.

3.4. *Liquid porosimetry technique*

The liquid porosimetry technique (CorrTest™) proposed by DiLeo and Phillips (DiLeo and Phillips, 1994, 1995; Phillips and DiLeo, 1996), has been used to monitor UF membrane integrity and to characterize UF membranes. This technique uses a pair of mutually immiscible fluids to validate the pore size distribution and particularly the maximum pore size. One of the fluids is employed as a membrane wetting agent and the other is used as an intrusion fluid. A transmembrane pressure is used so that the intrusion fluid can selectively penetrate the pores accessible to a given sized particle such as virus. Another transmembrane pressure is used to make the intrude fluid penetrate nearly all the membrane pores. The ratio of these two permeabilities obtained with each operating pressure is thus the percentage of total flow through the membrane pores accessible to a given size particle. That is to say, the different size particle retention characteristics can be expressed with the permeability ratio of these two liquids. Thus it is possible to obtain the pore size distribution or the maximum pore size when using the liquid porosimetry to characterize UF membranes. For this method, low intrusion pressure is needed due to the low interfacial tension associated with many pairs of immiscible liquids. Gekas and Zhang (1989) have used liquid porosimetry to characterize the entire pore size distribution of UF membranes at pressures less than 8 bars, which is much less than the required pressure when using gas–liquid porosimetry. In other words, to detect a defect with a given size, the required pressure of this method is lower than that of gas–liquid methods (e.g., pressure-based tests). The lower pressure results in less cost requirements and is more feasible during practical operation. As a result, this method is feasible for economic and operational point of views.

For this method, the CorrTest value (CTV) of the membrane, the ratio of the intrusion fluid flow rate through all of the membrane pores to the flow rate through those pores penetrated at the test pressure, is calculated at each intrusion pressure (P) using the following equation:

$$\mathrm{CTV} = \log\left[\frac{L_{\mathrm{total}}}{L_{\mathrm{int}}(P)}\right] \tag{6}$$

where L_{total} is the total membrane permeability of the intrusion fluid and it is essentially pressure independent. $L_{\mathrm{int}}(P)$ is the membrane permeability measured at each intrusion pressure. Assuming Poiseuille flow through cylindrical pores, the CTV can be rewritten as:

$$\mathrm{CTV} = \log\left(\frac{\int_0^{\infty} n(r_p) r_p^4 \, dr_p}{\int_{R_{flow}*}^{\infty} n(r_p) r_p^4 \, dr_p}\right) \tag{7}$$

where r_p is the penetrated pore radius, n is the number of membrane pores with r_p radius and $R_{flow}*$ is the minimum radius of penetrated pores at the test pressure. As seen in Eq. (7), CTV is dependent only upon the membrane pore size distribution and $R_{flow}*$. $R_{flow}*$ is related to the intrusion pressure through Washburn's equation:

$$R_{\mathrm{flow}}* = \frac{2\gamma \cos\theta}{P} \tag{8}$$

where γ is the interfacial tension, θ is the contact angle and P is the intrusion pressure. By definition, CTV is independent of membrane thickness and porosity, membrane surface area and the fluid properties. This is an important distinction of the CTV compared to the membrane hydraulic permeability which is strongly dependent on the membrane porosity and thickness as well as the pore size distribution. Gadam et al. (1997) found that the liquid porosimetry technique is able to accurately characterize the membrane pore size distribution, independently of module configuration. This technique is able to identify membranes of different molecular weight cut-off and is sensitive enough to capture slight changes of sieving coefficient for the same membrane cut-off with slight variations. Therefore, it is sufficiently sensitive to detect membrane sieving changes resulting from membrane breaches. Additionally, the liquid porosimetry technique is non-destructive and relatively simple to perform, which is convenient for practical operation. However it should be noted that this method is performed off-line.

3.5. *Novel membrane-based sensor*

Phattaranawik et al. (2007, 2008) proposed a novel membrane-based sensor device for upstream membrane integrity monitoring shown in Fig. 2.

The sensor is based on monitoring relative transmembrane pressure, which is created by two unimpaired membranes set in series inside the sensor device that detects deposition from the sample stream onto the first membrane of the sensor. Based on the principle of a transition from "sustainable flux" to "non-sustainable flux" conditions (Field et al., 1995; Howell, 1995) when a contaminated sample passes (Fig. 3), the sensor pressure signals are able to detect either intact or damaged membranes in the upstream membrane filtration process.

The sensitivity of this method, based on response time of the membrane sensor for particle detection, depends on

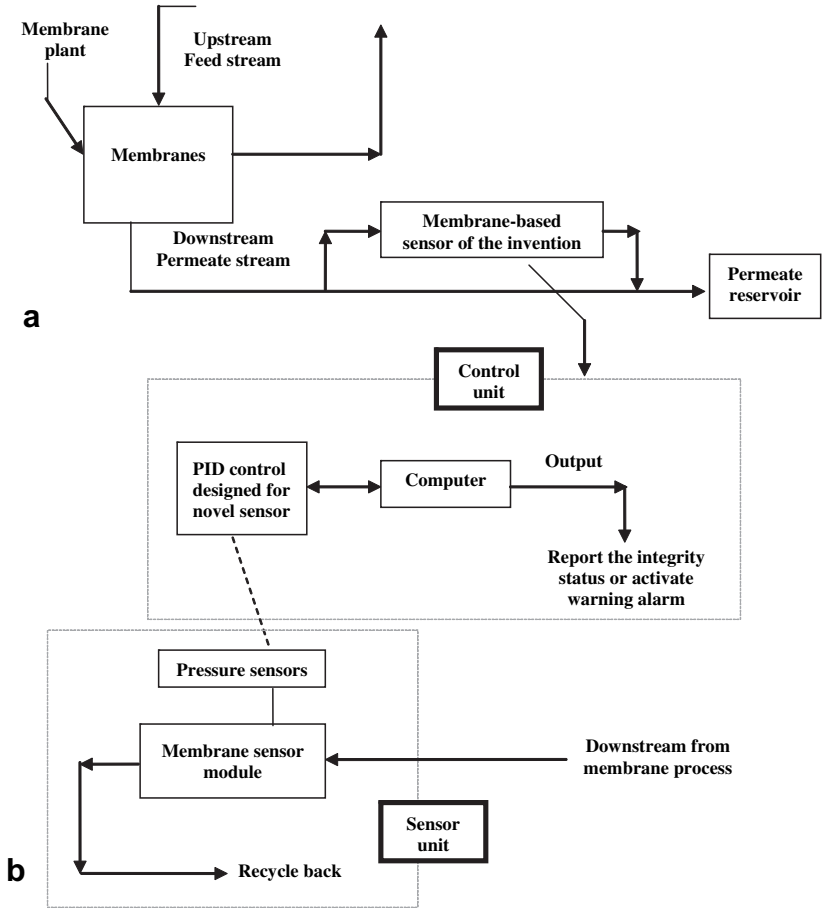

Fig. 2 – (a) Installation and (b) schematic diagram of the membrane-based sensor and control unit (Phattaranawik et al., 2008).

increasing supply pressures and particle concentrations. Results obtained by Phattaranawik et al. (2008) also showed that both sensitivity and stability of this sensor strongly depends on the membrane sandwich configurations (membrane characteristics) in the sensor and operation mode (pressurized or vacuum). In addition, the membrane sensor has shown to be able to detect bentonite particles with a 0.3 mg L^{-1} concentration in approximately 35 min in the vacuum mode. This membrane-based sensor is reliable, sensitive and low cost. It has potential applications in decentralized systems or in multi-channel monitoring of local conditions in a large plant. A disadvantage of this test is that it is necessary to use first an integrity test to validate the integrity of the sensor itself.

3.6. *Particle counting and particle monitoring*

The particle concentration in a membrane system with impaired fibers can be estimated by mass-balance equations (Panglisch et al., 1998). In the case of a negligible particle concentration in the permeate of an intact membrane, the particle concentration in the permeate of impaired membrane can be calculated from flux ratio of defect to total membranes, as demonstrated at Eq. (9).

$$c_P = \frac{n_d v_d}{n_i v_i + n_d v_d} c_F \quad (9)$$

where c_P is the particle concentration in the permeate of the membrane system, c_F is the particle concentration in the feed, v_i is the volume flow through the intact fiber, v_d is the volume

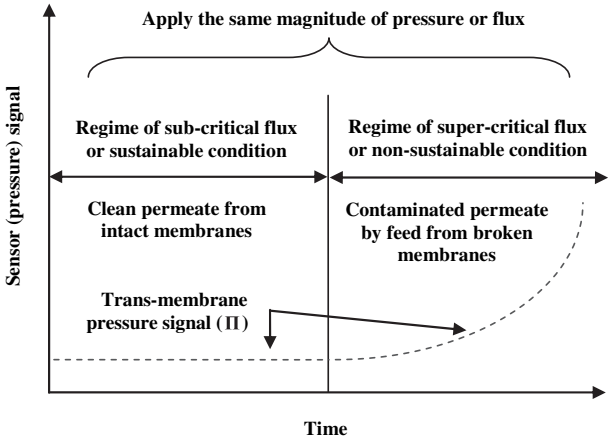

Fig. 3 – Concept of sustainable conditions for relative transmembrane pressure technique (Phattaranawik et al., 2008).

flow through the impaired fiber, n_i is the number of the intact fibers and n_d is the number of impaired fibers. It can be seen from Eq. (9) that the particle concentration in the permeate strongly depends on the particle concentration in the feed. In general, it is impossible to estimate the total number of particles in a water sample, because there are always particles which are smaller than the particle threshold of a particle counter. Below this threshold, the particles cannot be counted by the particle counter. It should be noted that, if it refers to particle counting, the counted particles are larger than the threshold of the particle counter.

A particle counter can count and monitor particles with different sizes in the feed and filtrate using a laser-based light scattering technique. As mentioned above, particle counting is related to a given size or size distribution of particles. The threshold of particle counters will directly affect the sensitivity of this method for membrane integrity monitoring. The sensitivity of particle counters increases with the decreasing threshold. For this reason, particle counters should have a threshold as small as possible. However, the cost of counters will spurt if the threshold of the counter is smaller. Adham et al. (1995) speculated that the 0.5–1 μm range is too sensitive because contaminants in this size range are likely to take place under field conditions and the data of this range could mislead to an operational standpoint. Thus, they only counted particles larger than 1 μm. It should be noted that particle counters are non-specific; i.e., they detect all particles in the appropriate size range. This results in a lower sensitivity of the membrane integrity monitoring. Many factors for on-line particle counting configuration also affect the quality and the accuracy of results. Adham et al. (1995) affirmed that some measures have to be taken when using a more sensitive particle counter. Flow control devices are recommended since they are able to maintain the flow rate through the particle sensor within very strict limits. The ideal flow controller has to be operated without introducing severe mechanical forces (pumps, gears, etc.), pulsations in sample flow rate through the sensor or without conditions that allow the particle size distribution to change by means of flocculation, settling or contamination. The distance of the particle sensor from the sample source or the flow controller should be as short as possible. Tubes made of an inert material should be used to prevent the adhesion of particles. The sensors have to be cleaned and the electronic background noise has to be checked in periodical intervals. In addition, the sensor should be calibrated at least every year.

Many studies (Adham et al., 1995; Panglisch et al., 1998; Glucina et al., 1997) were implemented to investigate how sensitive the particle counter has to be and how many membrane modules can be monitored by only one counter. The results about the membrane quantity which can be monitored by one counter are different. Adham et al. (1995) estimated that for a feed particle concentration of 200,000 particles mL^{-1}, a maximum membrane area of 18 m^2 can be monitored by one particle counter in case of dead-end microfiltration with the Memtec system, while Glucina et al. (1997) estimated the maximum membrane area for a dead-end filtration with Aquasource membranes at a particle feed concentration of 200,000 particles mL^{-1} to be 385 m^2. It has been demonstrated that the sensitivity of particle counting for membrane integrity monitoring strongly depends on the particle concentration in feed solution and it increases with the rising feed concentration. This conclusion is consistent with the result obtained by Panglisch et al. (1998). That is to say, the number of membrane elements, which can be controlled by just one particle counter, increases with the increasing feed particle concentration. If the feed solution is relatively clean, differences before and after filtration would be beyond the limits of current particle counters and cannot be detected by the particle counter. Under these circumstances, particle counters are not able to detect water quality changes at the levels required to ensure pathogen removal such as *Cryptosporidium* and *Giardia*. As the two most frequently used indirect membrane integrity tests, particle counting is more expensive than turbidity monitoring but it has higher detection sensitivity than turbidity monitoring (Adham et al., 1995; Jacangelo et al., 1991). However, it should be noted that current particle counting presents a lack of sensitivity (less than 4 LRV) sufficient for membrane integrity monitoring, which has also been reported by Landsness (2001). Particle counting may not be suitable in some cases, especially in the case of membrane operation in dead-end mode on low turbidity water (high diluted effect). In addition, this method is able to count air bubbles like particles as a result of air entrapment, especially for the membrane systems that use air for backwashing. This may make development of a stable baseline value more difficult for particle counters. Particle counting is also susceptible to counting particles shedding from downstream plumbing (Farahbakhsh et al., 2004). Another problem for particle counters is that it maybe produce unstable output during startup and shutdown.

Particle monitoring is similar to particle counting in principle. It provides an index of the water quality. Compared to particle counters which give a direct measurement of particle numbers by size, particle monitoring provides qualitative data based on a relative scale. This method is cheaper than particle counting. It offers less sensitivity for monitoring water quality changes due to membrane failures but is more sensitive than turbidity (Adham et al., 1995). Particle monitoring is seldom used, thus the water industry has limited experience with these devices.

3.7. *Turbidity monitoring*

This test is based on the difference of turbidity between the feed water and the filtrate. An intact membrane would be expected to show a 90% reduction in turbidity from feed to filtrate. Turbidity monitoring is less expensive than particle counting but offers lower sensitivity, which is reportedly not adequate to respond to changes in membrane integrity (Hirata and Hashimoto, 1998). For instance, even filtrate of low turbidity –<0.1 NTU– can have significant bacterial contamination (Rajagopalan et al., 2001), limiting the operation of this technique as a separate method for membrane integrity monitoring. Compared to the conventional turbidimeters, laser turbidimeters can improve detection sensitivity in excess of two orders of magnitude over conventional turbidimeters and are able to measure very low turbidity in the range of 0–1 NTU. Since most MF and UF systems produce filtrate water consistently in the range of 0.01–0.05 NTU, the

laser turbidimeter may be well suited to monitor membrane integrity. However, the results obtained by Colvin et al. (2001) indicated that the laser turbidimeter is somewhat less sensitive to integrity breaches than particle counting. Other experiences using laser turbidimeter have been also reported (Banerjee et al., 2001; Van Hoof et al., 2001; Naismith, 2005).

Turbidity monitoring is also subject to some of the same problems as particle counting. For example, if the feed water is relatively clean, the differences in measurements between feed and filtrate water would be beyond the capacity of current turbiditimeters. In addition, it is also subject to air entrapment error.

3.8. Surrogate challenge tests

As an alternative to on-line indirect methods, surrogate challenge tests are proposed to improve sensitivity of membrane integrity monitoring and to guarantee more efficiently drinking water quality. This method overcomes the low detection sensitivity of conventional indirect methods and the drawbacks of conventional direct integrity testing systems, such as off-line operation and lack of a direct relationship between the measured data and the removal efficiency (log removal). Certain surrogate challenge tests are used to represent the pathogen retention characteristics by membranes. Walsh et al. (2005) employed dissolved organic carbon (DOC) and color measurements to evaluate UF membrane integrity and showed promising results in their ability to identify a significant increase in the concentration of dissolved contaminants within the permeate stream. This study demonstrated the need for robust on-line indirect test methodologies for monitoring membrane integrity in the drinking water industry. Sakaji (2001) reported that maintaining membrane integrity monitoring with challenge tests can validate high removal levels of 5 or 6 LRV.

For drinking water treatment, the ultimate integrity test is the natural bacterial challenge test. Unfortunately, this test may result in additional problems, such as membrane biofouling or penetration of bacteria into permeate in case of compromised integrity. Therefore, the key point for surrogate challenge tests is to search for a suitable surrogate and corresponding measurement method. Surrogate challenge tests should be sensitive enough to detect changes in membrane integrity so that regulatory requirements for drinking water can be achieved. The precision and the minimum detection level of the measurement method directly affect the sensitivity of surrogate challenge tests. As for the choice of surrogate, multiple criteria have to be considered, such as well defined size, easily detectable, non-destructive, reasonable price and representative of pathogenic retention properties in drinking water treatment. Monodisperse surrogates are preferred because they can be more accurate to predict membrane retention based on the size exclusion (Causserand et al., 2002).

Here, several surrogate challenge tests for monitoring low-pressure membrane integrity are introduced, including the spiked integrity monitoring system-SIM™, microbial challenge tests, nanoscale probe challenge tests and magnetic particle challenge tests.

3.8.1. Spiked integrity monitoring system – SIM™

The spiked integrity monitoring system or SIM®-system was developed by NORIT Membrane Technology in close co-operation with Water Supply Company North Holland and IWW Rhenish–Westphalian Institute for Water Research, which combines the accuracy of a challenge test with the speed of a pressure test, while keeping the system under test during operation (Van Hoof et al., 2001). For this test, high concentrations of powdered activated carbon (PAC) are spiked in the membrane feed, followed by the monitoring of particulate level in permeate with particle or turbidity monitors. Then, a log removal value can be calculated. It was reported that the SIM™ system provides increased sensitivity by increasing feed particle concentration (Kruithof et al., 2001). The SIM™ is an on-line membrane integrity test with high sensitivity. However, it is not suitable from a regulatory point of view, since the test results cannot be linked directly to pathogen removal. The PAC particle size does not remain constant during the test, and the particle size distribution may vary considerably, depending on the manufacturer, the dispersion mode of the PAC and the water quality. In addition, the problems derived from the measurement apparatus, which exist in particle counting test or turbidity monitoring test, also affect the sensitivity of this method. Finally, it should be noted that the PAC particles may occur in the permeate but it cannot be removed with a backwash due to the module capacity.

3.8.2. Microbial challenge tests

Microbial challenge tests, e.g., bacteriophage and oocyst, spike a given concentration of microorganisms in the feed and measure the particulate levels in the permeate in the case of no chloration. Bacteriophages, a class of viruses that infect bacteria, are often used as microbial surrogates because they avoid the health risks of native pathogen. It can well represent viral pathogen removal by the membrane system because they are close in size, shape, and surface properties. Microbial challenge tests can provide a good sensitivity for fiber break detection and provide some indication of membrane disinfection efficiency. This method is also a basic research tool to allow the log removal calculation for a specific organism. However, microbial monitoring is not practical for routine test in large-scale membrane plants because of several reasons (Gitis et al., 2002). Firstly, the measurement of bacteriophage in the water is based on the plaque forming unit (PFU) count. Because the PFU count is very complicated and needs a long time (12–48 h), it cannot reflect the membrane integrity in real time. In addition, plaque techniques count a virus cluster as a single unit rather than reflecting the actual number of viruses. Secondly, bacteriophage inactivation and transport phenomena are always coupled and it is difficult to distinguish the decrease of bacteriophage which is due to membrane retention or biological inactivation. Thirdly, microbial monitoring may not correctly characterize the membrane integrity since bacterial re-growth may take place in the permeate piping system. So bacteria in the permeate may be over-estimated although there is no membrane failure. Trimboli et al. (2001) described the implementation of a bacillus spore challenge test to measure the integrity of a large MF membrane system. This test is sensitive but expensive. In addition, some researches which use

fluorescent-dye-labelled MS2 bacteriophages to evaluate the UF membrane integrity have also been reported (Gitis et al., 2006a,b). Generally, microbial challenge tests are performed simultaneously with pressure-based integrity tests in drinking water treatments to validate and improve membrane integrity monitoring techniques (Brehant et al., 2008).

3.8.3. *Nanoscale probe challenge tests*

To efficiently detect nanoscale breaches in membranes, Gitis et al. (2006a,b) proposed applying new monodispersed nanoscale probes – citrate-stabilized (12 nm) or thiol-stabilized (15 nm) gold nanoparticles – to evaluate the UF membrane integrity. Gold nanoparticles are preferred to other metal nanoparticles due to the extremely low background level in water system (usually between 5 and 50 ppt), non-pathogenicity and safe use, high monodispersity, and inexpensive price. The gold nanoparticles challenge test assumes that the membrane retains particles by a size exclusion mechanism regardless of surface chemistry, vitality, pathogenesis, and other characteristics that differentiate organic from inorganic particles. Experimental results showed that breaches in membrane integrity were detected as early as the first appearance of small holes with an average diameter of 20 nm when applying these two nanoscale probes. These results indicate that gold nanoparticle challenge tests are sufficiently sensitive to detect breaches which permit viruses to pass through the membrane. Here, gold nanoparticles were detected using anodic stripping voltammetry (ASV). ASV system showed high sensitivity (low minimum detection level) to the order of a single part per billion, indicating the feasibility to develop the experimental protocol for simple and sensitive on-line detection. But ASV needs a prolonged run time or the addition of some other metal to resolve this problem. Another highly sensitive nanoparticle analysis method based on the laser-induced breakdown detection (LIBD) was found to be sensitive enough to monitor the particle passage through low-pressure membranes (Lipp et al., 2008). This may also provide a suitable on-line monitoring method for UF/MF membrane integrity. However, it is also reported (Lipp et al., 2008; Lohwacharin and Takizawa, 2009) that more significant membrane fouling due to intermediate pore blocking took place with small size of nanoparticles. Nanoparticle challenge tests provide a basis for developing a new on-line membrane integrity monitoring method with high sensitivity to detect virus-size breaches in membranes.

3.8.4. *Magnetic particle challenge tests*

Currently on-line membrane integrity tests are generally analyzed by non-specific turbidimeters or particle counters. An alternative method of probing membrane integrity by the use of magnetically susceptible particles was developed to improve both specificity and sensitivity (Moulin, 2008; Rajagopalan et al., 2006; Deluhery and Rajagopalan, 2008). The principle of magnetic particle challenge tests is spiking a certain concentration of magnetic particles in the feed and detecting the material magnetic properties in the filtrate. The particle size used should be greater than the membrane pore size to ensure that there is no particle passing through an intact membrane. In the case of an impaired membrane, particles are detected in the filtrate by an appropriate magnetic sensor, indicating the loss of membrane integrity. Magnetic sensors include superconducting quantum interference device (SQUID) magnetometer, giant magneto-resistive (GMR) sensors, measurement of magnetic permeability and measurement of magnetic susceptibility. Non-magnetic sensors can also be used but it is non-specific. Superparamagnetic particles are preferred for the test due to their high magnetic susceptibility and low density. High magnetic susceptibility makes it more easily detected by magnetic sensors and decreases the external magnetic field strength needed for high capture efficiency if particle capture is needed. Lower particle densities reduce the possibility of particle sedimentation at low flow rates. Here, the particle size and detection sensor have a decisive effect on the sensitivity of this membrane integrity test. For example, nanoscale particles can detect nanometric breaches of the membrane. Recently, some researchers (Rajagopalan et al., 2006; Deluhery and Rajagopalan, 2008) proposed to use a magnetic field to concentrate the particles for detection, which allows the use of smaller and less powerful sensors and thus realizes cost savings or enhanced detection limits of this integrity test. They implemented the membrane integrity test with magnetic particles of 1 μm diameter and the particles in the filtrate were successfully separated and concentrated in the influence of a magnetic field. Moulin (2008) is the first to propose an integrity test based on the magnetic characteristics without a concentration step due to a high sensitivity of the analytical apparatus (LRV >6).

Lab-scale results about this method have been reported. On-line magnetic challenge tests for monitoring membrane integrity, with the advantages of detection specificity, high detection sensitivity and on-line operation, is a plausible one for large-scale applications.

4. Operation of membrane integrity tests

Thus far, the most widely used membrane configuration in drinking water treatment is hollow fiber membrane filtration. The main membrane system suppliers have employed hollow fiber membranes. In a membrane drinking water treatment plant, the detection of membrane system failures requires that an effective membrane integrity test system at an acceptable frequency is used. As the most widely used methods, the PDT and DAF test are considered to be reliable for monitoring membrane system integrity. The PDT and DAF test are generally proposed to be conducted according to the same sequences: (1) detecting compromised rack(s) in the plant; (2) detecting compromised module(s) in the rack; and (3) detecting compromised fiber(s) in the module.

Current PDT or DAF test for hollow fiber membranes are typically set to alarm against parameters based on an absolute size removal (4 μm and greater) and a particle log removal value (4–5 LRV). Some companies set alarms against other measured variables, e.g., feed turbidity, high pH in treated water. These criteria reflect the international position but do not satisfy the regulatory requirements for an absolute removal of more than 1 μm. Higher starting pressure for the test would enable smaller defects (e.g., 1 μm) to be detected thereby satisfying the current requirements. However, higher

Table 2 – Advantages and disadvantages of different membrane integrity tests.

Membrane integrity test	Advantages	Disadvantages	Typical module test time	References
Pressure-driven tests				
PDT	(1) Independent of feed water quality (2) Low maintenance (3) Non-destructive (4) Reliable and high sensitive to detect membrane breaches to meet the regulatory requirements, 4.5–5 LRV (5) The most widely used and accepted by both utilities and primacy agencies (6) High degree of automation	(1) Off-line operation (2) No measurement of filtrate water quality (3) Not sufficiently sensitive to detect nanoscale breaches (4) Need more sensitive pressure sensor when the number of fibers tested is important (5) Potential to yield false–positive results if the membrane is not fully wetted	10 min (5 min for Memsure™ PDT)	Adham et al., 1995 Johnson, 1997, 1998 Landsness, 2001 U.S. EPA, 2001, 2003 Jackson, 2001
DAF test	(1), (2) and (3) Are the same as PDT (4) More sensitive than the PDT, even more than 6 LRV (5) Widespread use (6) Ease to conduct and accuracy of test (when measuring water displacement)	(1), (2), (3) Are the same as PDT (4) Not included as standard equipment for MF and UF systems (5) Sensitive to temperature (6) Requires some additional pipe work when measuring water displacement	15 min (5 min for Memsure™ DAF)	Adham et al., 1995 Johnson, 1997, 1998 Trimboli et al., 2001 Cheryan, 1998 Jacangelo et al., 1997 Jackson, 2001
Bubble point test	(1) Is the same as PDT (2) Ease of conducting (3) Useful tool for pinpointing compromised fiber(s) and leaks identified by other test methods (4) High sensitivity of 6 LRV for Memcor® CMF process	(1) and (2) Are the same as PDT, and the module to be tested should be removed from the rack (3) Manual application (4) Practically unattainable for UF membranes	Depends on the bubble point	Randles, 1997 Giglia and Krishnan, 2008
Vacuum decay test	(1) and (3) Are the same as PDT (2) Useful as a screening procedure (3) Ability to test spiral-wound membranes or other systems that cannot be pressurized on the filtrate side of the membranes	(1) and (2) Are the same as PDT (3) Rarely performed as a mean of absolute verification of a leak in drinking water treatments (4) Should be repeated to confirm its reproducibility (5) Difficult in removing entrained air after the test has been completed	Several minutes	FILMTEC Membranes, troubleshooting: membrane element evaluation

Other direct tests				
Binary gas integrity test	(1) Non-destructive (2) Higher detection sensitivity than the single gas test, greater than 6 LRV (3) Independent of feed water quality	(1) Off-line operation (2) No measurement of filtrate water quality (3) Just in pilot-scale study (4) the storage of gas and the gas toxicity (e.g., C_2F_6)	Longer than single gas tests	Giglia and Krishnan, 2008
Acoustic sensor test	(1) Simple and non-destructive (2) Economic competitive (3) On-line sensor (4) Independent of feed water quality (5) High sensitivity of 6 LRV of viruses	(1) Depends on the background noise and flow rate (2) No measurement of filtrate water quality	Direct	Adham et al., 1995 Laîne et al., 1998 Jackson, 2001
Liquid–liquid porosimetry	(1) Non-destructive and relatively simple to perform (2) Independent of feed water quality (3) More feasible than gas–liquid porosimetry to characterize UF membrane	(1) Off-line operation (2) No measurement of filtrate water quality (3) No report on practical operations	Longer than pressure-based tests	DiLeo and Phillips, 1994, 1995 Phillips and DiLeo, 1996 Gekas and Zhang, 1989
Indirect tests				
Particle counting	(1) Continuous and on-line operation (2) Convenient for routine qualitative monitoring (3) Independent of membrane configurations (4) Widespread use and familiarity in the water industry (5) More sensitive than particle and turbidity monitoring	(1) Low detection sensitivity (<4 LRV) (2) Strongly depends on feed concentration and operating conditions (3) Susceptible to entrapment air as particles and to count particles shedding from downstream plumbing (4) Relatively high cost (5) Produces unstable output during startup and shutdown (6) Periodic cleaning and calibration of the sensor (7) Flow control devices are recommended before the sensor	Direct	Jacangelo et al., 1991 Adham et al., 1995 Panglisch et al., 1998 Landsness, 2001 Glucina et al., 1997 Farahbakhsh et al., 2004 Jackson, 2001
Particle monitoring	(1), (2), (3) Are the same as particle counting (4) Significantly lower cost than particle counters (5) More sensitive than turbidity monitoring (6) No calibration required	(1), (2), (3) Are the same as particle counting (4) Seldom used in the water industry	Direct	Adham et al., 1995 Jackson, 2001

(*continued on next page*)

Table 2 (*continued*)

Membrane integrity test	Advantages	Disadvantages	Typical module test time	References
Turbidity monitoring	(1), (2), (3), (4) Are the same as particle monitoring (5) Widely used in drinking water plants	(1), (2), (3) Are the same as particle counting	Direct	Banerjee et al., 2001 Van Hoof et al., 2001 Colvin et al., 2001 Naismith, 2005 Jackson, 2001
Membrane-based sensor	(1) Can be operated on-line and continuously (2) Sensitive and low cost	(1) Needs an integrity test firstly for the integrity test (2) No further information about membrane pilot operation	35 min	Phattaranawik et al., 2007, 2008
Surrogate challenge tests				
SIM™	(1) On-line operation (2) High detection sensitivity	(1) High feed concentration (2) Cannot be linked directly to the virus removal (3) The PAC size may vary considerably during the test (4) Subject to the same problems that particle counters or turbidity monitoring	30 min	Van Hoof et al., 2001 Kruithof et al., 2001 Jackson, 2001
Microbial challenge tests	(1) Sensitive (2) Generally performed simultaneously with other direct integrity tests to validate and improve membrane integrity monitoring techniques	(1) Natural bacterial challenge test is destructive (2) Long time for microbial analysis and cannot reflect membrane integrity in time (3) Not continuous and off-line operation	24–48 h	Brehant et al., 2008 Trimboli et al., 2001 Gitis et al., 2002 Jackson, 2001
Nanoscale probe challenge tests	(1) On-line operation (2) High detection sensitivity (3) Ability to detect virus-size breaches	(1) Significant membrane fouling due to the pore blocking with the smaller size of nanoparticles (2) Cannot be linked directly to the pathogen removal (3) Just in pilot-scale study	Direct	Gitis et al., 2006a,b Lipp et al., 2008 Lohwacharin and Takizawa, 2009
Magnetic particle challenge tests	(1), (2) Are the same as the nanoscale probe challenge test (3) Ability to detect virus-size breaches when using magnetic nanoparticles as surrogates (4) Simple and detection specificity (5) Low cost (6) Independent of feed water quality	(1) Just in pilot-scale study, in progress on industrial scale (2) Need more information on the effect on nanomaterials on health	Direct	Moulin 2008 Rajagopalan et al., 2006 Deluhery and Rajagopalan, 2008

start pressures may cause some membranes to experience higher defect rate than that are currently assumed. For different commercially available membranes, an annual fiber failure rate has been found between 1 and 10 per million fibers (Gijsbertsen et al., 2006).

The frequency of the membrane integrity test is important for maintaining specific LRV requirement. There are no common guidelines for the frequency of membrane integrity tests. Membrane integrity test frequencies need to be related to a risk assessment as the first priority and operational feasibility as the second priority. Given an expected number of fiber breaks per year and a LRV target, the frequency of membrane integrity tests can be calculated based on a simple mode or a statistical model (Bennett, 2005). Membrane integrity tests can be implemented once a day, once every 72 h or longer, or after cleaning. It has been summarized that, in most cases, a weekly test is required to ensure LRV more than 4.5 (Pearce, 2007). For Bristol Water, a PDT frequency of once every week to once every other week is recommended to provide a margin of safety for the target of 4 LRV (Bennett, 2005). Generally, a daily test frequency would be beneficial when ascertaining system integrity, which is consistent with the continuous sampling and analysis practice. In general, the data from membrane integrity tests during operation should be kept between 2 and 5 years (Jackson, 2001).

In general, a perfect method for low-pressure membrane integrity monitoring should be low cost, simple, on-line and continuous. In addition, it should be reliable and highly sensitive to detect membrane breaches, even for nanoscale breaches. Here, a summarization of current different membrane integrity tests is shown in Table 2.

5. Conclusions

Membrane integrity tests are very important for membrane-based drinking water treatment because they can ensure the quality of treated water, especially in terms of pathogen removal. The criteria of assessing a method for monitoring membrane integrity include sensitivity, continuity, reliability and cost effectiveness. Generally, the sensitivity of membrane integrity monitoring methods is expressed in term of log removal value (LRV) of pathogens for drinking water treatments. Of current different membrane integrity tests, PDT and DAF test are the most frequently used and considered to be simple and reliable. These two tests are able to detect membrane integrity at high levels of 5–6 LRV, even more than 6 LRV for DAF test. However, they are conducted off-line. In contrast, indirect integrity tests, such as particle counting and turbidity monitoring, are performed simply and on-line but have lower sensitivity. They are not able to detect water quality changes at the levels required to ensure pathogen removal. To realize reliable and efficient on-line membrane integrity monitoring, some other methods are proposed, including acoustic sensor method, liquid porosimetry, surrogate challenge tests, etc. All these methods have their own advantages and disadvantages. Membrane integrity tests are specific for the type of membrane used and are dependent of membrane manufacturer and membrane system supplier. In general, membrane suppliers have their own membrane integrity test procedures based on 4–5 LRV. During practical operation, the frequency of membrane integrity tests is very important to achieve the required LRV.

So, to better satisfy the regulatory requirements of the drinking water industry, it remains urgent to develop an alternative on-line monitoring technique for quick, accurate, simple, continuous and relatively inexpensive evaluation of the low-pressure membrane (UF and MF) integrity. Based on the above information, surrogate challenge tests with nanometric material are interesting and promising because it can ensure the disinfection efficiency of the UF membrane and make it possible to realize the required objective with the development of more accurate and advanced measurement apparatus. For this purpose, an alternative on-line UF membrane integrity test by using magnetic nanoparticles as surrogates reveals that this challenge test is suitable for large-scale drinking water applications. But in this case, more information on the effect of nanometric material on health is needed.

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