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Analysis of performance criteria for ultrafiltration membrane integrity test using magnetic nanoparticles

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HIGHLIGHTS

- A prediction model was developed to calculate the membrane defect size.
- The measured data goes with similar trend with model D.
- Nanoparticle test has 39.33% probability to have a theoretic resolution of 3 μm.

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ABSTRACT

An alternative ultrafiltration membrane integrity test utilizing magnetic nanoparticle as a surrogate has been investigated in previous studies, but the absence of a feasible estimation model for the degree of membrane damage causes that this simple membrane integrity test would be not applied easily. This study proposed a calculating model to predict membrane defect size, and investigated the theoretic resolution of the integrity test method. The results obtained with the evolved prediction model D, which is based on Darcy's law and Bernoulli equation, were satisfactory in predicting the membrane defect size. In this study, this integrity test method had about 39.33% probability to have a theoretic resolution of 3 μm or less under common experimental conditions.

1. Introduction

Ultrafiltration has been used to produce drinking water to meet the more stringent regulations on water quality because of its capacity of removing particulate and pathogens [1]. To make sure that a membrane process acts as an effective barrier against pathogens and other particulate matter, accurate and efficient integrity tests of the membrane system should be applied to guarantee the quality of filtered products and detect the presence of oversized pores or defects that can compromise the retention capability of the filter [2]. The tests for broken fibers or defects should be sensitive to breaches as small as 3 μm which is based on the lower size range of *Cryptosporidium* oocysts, then the tests could make sure that any integrity breach large enough to pass oocysts will contribute to a response from the direct integrity test being used [1,3]. Generally, conventional membrane integrity monitoring techniques are divided into direct methods and indirect methods [2]. Direct methods refer to tests directly applied to membrane or membrane module, i.e. pressure decay test, diffusive air flow test, bubble point test, vacuum decay test, nanoscale probe test [4], marker-based test [5] and binary gas integrity test [3]. Indirect methods involve monitoring some aspect of filtrate water quality as a surrogate measurement of membrane integrity, i.e. particle counting [6,7], particle monitoring [8,9] and turbidity monitoring.

An ultrafiltration membrane integrity test based on the use of Fe_3O_4 magnetic nanoparticles and the measurement of magnetic susceptibility has been investigated in previous studies [10,11]. This membrane integrity test was demonstrated, with the advantages of simplicity, online operation, high detection specificity and sensitivity, quick detection and very low influence on membrane fouling, to be suitable for large-scale drinking water plants [12–14]. This marker-based direct integrity test can be viewed as a "mini challenge study," in which magnetic nanoparticle suspension is periodically applied to the feed water in order to verify the integrity of a membrane filtration system. This test relies primarily on the measurement of downstream magnetic susceptibility. Since magnetic susceptibility performs as a good indicator for magnetic

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nanoparticle concentration, the estimation of the size of the damaged part can be conducted by using a relationship between the amount of permeated particles and the size of the damaged part [15].

A measurable nanoparticle flow concentration, which is in excess of detectable limit or higher than a flow concentration empirically established for a membrane without defect, should signal the presence of a defect. The sensitivity of this test is determined by the minimum detectable nanoparticle concentration or magnetic susceptibility. There could be device-to-device variabilities in membrane area, membrane pore diameter, membrane pore density, pore tortuosity, dynamic viscosity of water and dynamic viscosity of nanoparticle suspension. Other factors such as feed solution concentration, operation mode, fiber breach location, number of fibers per module, fiber diameter, trans-membrane pressure and membrane fouling can also impact the measured nanoparticle concentration. The variability in nanoparticle concentration acting as background noise can diminish the sensitivity of the integrity test based on nanoparticles [12].

The nanoparticle flow concentration could be calculated using the relationship between the flow rate through membrane and the flow rate through the defect. In a membrane filtration process, flow rate through membrane or through a defect can be estimated using different equations based on different flow rate equations, such as Darcy's law, the Hagen–Poiseuille equation and the Bernoulli equation. However, it is unknown that which mechanism or equation is more suitable for the estimate of nanoparticle flow concentration through a broken membrane. As a result, a prediction model of the size of membrane damage should be developed to evaluate the size of the defect.

In addition, as a kind of integrity detection method, the resolution of this method should be determined, and the control limits should be established at the threshold test responding for various degrees of integrity loss [16].

The focus of this paper is the use of Fe_3O_4 magnetic nanoparticles to develop a prediction model of the size of membrane damage using experimental conditions (concentration of permeate nanoparticle concentration and membrane system parameters). And the resolution of this test method has also been investigated.

2. Theoretical background

2.1. Flow rate through integral membrane

In a membrane filtration process, flow rate could be estimated using different equations based on different flow rate equations, such as Darcy's law, the Hagen–Poiseuille equation, the Bernoulli equation.

Firstly, flow rate could be estimated using equation based on Darcy's law:

$$Q_{water} = \frac{TMP}{\mu_{water} R} \cdot A \tag{1}$$

where Q_{water} is the permeation flow rate ($m^3.s^{-1}$), *TMP is* trans-membrane pressure (Pa), μ_{water} is dynamic viscosity of the liquid ($Pa \cdot s^{-1}$), A is membrane area (m^2) and R is membrane resistance (m^{-1}).

Secondly, using the capillary pore diffusion model and the Hagen–Poiseuille equation, the flow rate could be calculated by the Hagen–Poiseuille equation:

$$Q_{water} = \frac{\varepsilon d_0^2 \cdot TMP}{32\tau \mu_{water} \delta} \cdot A \tag{2}$$

where ε is membrane porosity (non-dimensional), d_0 *is* the original mean pore diameter (m), τ is membrane tortuosity (non-dimensional) and δ is the membrane skin layer thickness (m).

In addition, the membrane pure water flow rate could be calculated using another expression mode [17]:

$$Q_{water} = \frac{\pi d_0^4 \cdot TMP}{128\tau \mu_{water} \delta} \cdot N_0 \cdot A \tag{3}$$

where N_0 is the original membrane pore density (non-dimensional).

For an integral membrane, the permeate is depleted of magnetic nanoparticle, and no magnetic susceptibility would be detected in the permeate [2].

2.2. Liquid flow through a pore defect

If a defect is present, the addition of a small flow rate originating from the defect may change the total flow. The liquid flow rate through a pore defect could be calculated using the Hagen–Poiseuille equation for non-compressible fluids:

$$Q_{NP} = \frac{\pi d_{breach}^4 \cdot TMP}{128 \mu_{NP} \delta} \tag{4}$$

where Q_{NP} is the flow rate of nanoparticle suspension passing defect ($m^3.s^{-1}$), d_{breach} is the equivalent diameter of the breach (m), μ_{NP} is the dynamic viscosity of the nanoparticle suspension ($Pa \cdot s^{-1}$). Here, the fluid should be viscous and incompressible, the flow should be laminar through a defect pore of constant circular cross-section that is substantially longer than defect diameter. In the flow process there would be no acceleration of fluid. If adding magnetic nanoparticle suspension into the feed water, the leakage through the membrane defect will contaminate the permeate stream, resulting in an elevated concentration of magnetic nanoparticle and measurable magnetic susceptibility.

If a larger defect (defect size diameter is more than half of membrane thickness) is present, the liquid flow would be a turbulent thin-walled orifice flow and the flow rate could be calculated using the Bernoulli equation for non-compressible fluids:

$$Q_{NP} = C_q \frac{\pi d_{breach}^2}{4} \sqrt{\frac{2 \cdot TMP}{\rho}} \tag{5}$$

where C_q is orifice flow rate coefficient (non-dimensional) and ρ is water density ($kg.m^{-3}$).

2.3. Impact of a defect on the concentration of nanoparticle in the permeate

By combining Q_{NP} and Q_{water}, the concentration of nanoparticle in the permeate could be induced as follows:

$$C_p = \frac{Q_{NP} \cdot C_f}{Q_{water} + Q_{NP}} \tag{6}$$

where C_f and C_p are the concentration of feed and permeate.

By combining Eqs. (1) and (4), the concentration of nanoparticle in the permeate could be induced as shown in Eq. (7).

$$\text{Model A}: \quad C_p = \frac{C_f}{1 + \dfrac{128 \mu_{NP} A \delta}{\pi d_{breach}^4 \mu_{water} R}} \tag{7}$$

Here, Model A is deduced from Darcy's law and Hagen–Poiseuille equation for tiny defect smaller than half of membrane thickness.