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Accelerated testing for fouling of microfiltration membranes using model foulants

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HIGHLIGHTS

- Fouling of hollow fiber membranes was evaluated by means of accelerated testing experiments.
- A simple model based on pseudo-cake filtration model was applied to estimate the normalized fouling rates, θ/J^2.
- In the accelerated testing of membranes, the fouling rate was less sensitive to foulant concentration than to flux.

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ABSTRACT

Hollow fiber microfiltration (MF) membranes have been widely employed for water and wastewater treatment. Nevertheless, membrane fouling is still one of the most serious issues in operating hollow fiber membrane systems. In this research, fouling of hollow fiber MF membranes was evaluated by means of accelerated testing experiments. A single fiber filtration unit was used to perform the fouling experiments in an accelerated way. Permeate flux and foulant concentrations were used as fouling control parameters to adjust the rate of fouling. Four model foulants used were kaolin, silica, natural organic matters (NOM), and alginate. A simple theoretical model was applied to investigate the fouling characteristics of the membrane by the model foulants. The analysis showed that there was a nonlinear correlation between the fouling rate and the fouling control parameters. The "normalized" fouling rate, θ/J^2, was found to be useful to quantify the fouling rate and to implement a long-term simulation of TMP changes.

1. Introduction

Microfiltration or ultrafiltration (MF/UF) membrane has been gaining popularity as a feasible option for advanced water and wastewater treatment [1,2]. The use of MF/UF has been studied by researchers since the mid-1990s and cost reduction in these technologies in the mid-2000s led to the installation of MF/UF plants [3]. MF and UF systems typically utilize hollow fiber modules. A major advantage of hollow fiber membrane modules over other configurations of membranes is the high membrane surface area to footprint ratio achieved by low aspect ratio (diameter-to-length ratio) of fibers. Moreover, they provide cost-effective methods of removing particles and pathogenic microorganisms from treated water [4].

However, membrane fouling is still one of the most serious shortcomings in hollow fiber MF/UF systems [3,5]. The problem lies with the fact that membrane fouling is difficult to predict and control [6]. Fouling behavior is influenced by various factors, including membrane surface properties, the nature of the particle or dissolved foulants, and feed water properties [7,8]. Accordingly, it is highly desirable to have an accelerated fouling test method that is short in duration, utilizes a minimum amount of test solution, only requires a small membrane area, and is representative of the large-scale process [9].

Accelerated life testing, which is defined as the process of testing a product by subjecting it to conditions in excess of its normal service parameters, has been widely applied to many industries to predict long-term performance of a product [10,11]. Nevertheless, relatively few works have been done in the field of MF/UF membranes. Previous works on accelerated testing of MF/UF membranes have focused on the chemical degradation and aging [12–14]. Although fouling status of membrane was identified as an important aging factor [14], little information is available on the accelerated testing conditions of membrane fouling.

This study focuses on developing accelerated testing protocols as a tool to predict long-term performance and lifetime of hollow fiber MF membranes. Using model foulants such as kaolin, silica, natural organic matters (NOM), and alginate, fouling propensity of the membrane was quantitatively analyzed. A theoretical model was applied to analyze the fouling performance of hollow fiber MF membranes. The flux and

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foulant concentrations were used as important parameters to accelerate fouling. Using the filtration model and the parameters obtained from the experiments, the fouling rates could be estimated.

2. Theory

2.1. Analysis of data from accelerated fouling test

We applied a simple filtration model to estimate the fouling rates of dead-end microfiltration. Although there are a lot of different fouling mechanisms depending on the characteristics of foulants, pseudo-cake filtration model was adopted in this study for simple analysis of data. Based on this model, the permeate flux (J) on the TMP can be described by Darcy's law [15].

$$\Delta P = \eta(R_\mathrm{m} + R_\mathrm{c})J \tag{1}$$

where ΔP is the transmembrane pressure (TMP); η is the viscosity of water; R_m is the membrane resistance; R_c is the cake resistance; and J is the permeate flux. The cake resistance (R_c) is given by

$$R_\mathrm{c} = \frac{\alpha m_\mathrm{c}}{A_\mathrm{m}} \tag{2}$$

where α is the specific cake resistance; m_c is the mass of cake deposited on the membrane; and A_m is the membrane area. Here, m_c is proportional to the flux of the foulants:

$$m_\mathrm{c} = JA_\mathrm{m}c_\mathrm{f}t \tag{3}$$

where c_f is the effective foulant concentration. Note that c_f is different from c_b, which is the bulk concentration of foulant. This implies that all foulants cannot approach the membrane surface due to the back transport. By combining Eqs. (1), (2), and (3), ΔP is given by

$$\Delta P = \eta R_\mathrm{m}J + \eta\alpha c_\mathrm{f}J^2t = \eta JR_\mathrm{m} + \theta t \tag{4}$$

where θ is the rate of membrane fouling in dead-end filtration tests. Under constant flux conditions, θ can be calculated from the slope of the plot between t and ΔP.

To accelerate the fouling rate (θ), either J or c_f may be increased. However, θ may not be linearly proportional to J (or c_f). Accordingly, it is important to understand the correlations between θ and J (or c_f), which should be experimentally determined.

2.2. Using the simulation based on the mathematical model

Once θ is known, it can be used for long-term simulation of hollow fiber MF systems. Of course, there should be a substantial difference between the results from this simple simulation and the actual data from a pilot or full scale plants. The information from the simulation should be used just for initial projection of the performance of MF membranes.

Considering the situation that periodic backwash is applied, Eq. (4) is modified as

$$\Delta P(t + \Delta t) = \Delta P(t) + \alpha\eta J^2 c_\mathrm{f}\Delta t \tag{5}$$

$$\Delta P(t)|_\text{after backwash} = \Delta P(t)|_\text{before backwash} - J\eta R_\mathrm{c}(t)\beta \tag{6}$$

where β is the constant to describe the effect of backwash. To consider the effect of viscosity, the following correlation was used [16]:

$$\eta = 2.414 \times 10^{-5} \times 10^{\left(\frac{247.8}{T-140}\right)} \tag{7}$$

The flow chart for the simulation is shown in Fig. 1. Here, β may be determined from a set of experiments. Based on the algorithm in Fig. 1, a simulation model was developed using Matlab (Fig. 2).

3. Experimental section

3.1. Experimental setup

A schematic diagram of a laboratory-scale, submerged hollow fiber membrane system for accelerated fouling test is shown in Fig. 3. The system consisted of 12 filtration tanks, allowing the simultaneous testing of MF fibers at the same time. Each tank had a working volume of 1 L and MF fiber was immersed vertically in the reactor. A magnetic stirrer was positioned just below the membrane and the stirring speed was controlled by a magnetic stirrer plate.

The MF fibers were made of polyvinylidene fluoride (PVDF) with the nominal pore size of 0.2 μm. They had an internal diameter of 0.7 mm and an external diameter of 1.3 mm. The length of the fiber was 18 cm. Since the fiber was relatively short, the pressure drop along the fiber was neglected.

Permeate from the membrane module was pulled by a peristaltic pump (EW-07551-00, Cole-Parmer, USA). A permeate volume was frequently measured by collecting permeate volume using a mass cylinder. The transmembrane pressure was continuously measured using a pressure transducer (ISE40A-01-R, SMC, Japan) and a data logger (usb-6008, NI. U.S.A.), which were connected to a computer. The temperature of solution was kept constant at 20 °C. Total recycle mode, where both the retentate from the MF loop and permeate were recycled into the tank, was adopted to keep the reactor volume constant during the operation time.

3.2. Model foulants and test conditions

Model foulants used in this study were kaolin (Sigma Aldrich), silica (Sigma Aldrich, Ludox colloidal silica AM-30), NOM (IHSS, Swannee river natural organic matter), and alginate (Sigma Aldrich, alginic acid sodium salt from brown algae). The concentrations of the foulants were 2, 5, 10, 20 mg/L, respectively. The flux step method was applied to investigate the effect of applied flux on fouling rate by adjusting flux between 50 L/m^2 h and 200 L/m^2 h. Prior to each filtration test, all membranes were stabilized using deionized water during 500 min.

4. Results and discussion

4.1. Effect of flux and foulant concentrations on transmembrane pressure

To examine the basic properties of the MF membranes, a set of filtration tests were out using colloidal silica under the following operating conditions: flux, 50, 100, 150, and 200 L/m^2 h; concentrations of the silica, 2, 5, 10, and 20 mg/L. The results are shown in Fig. 4. As the flux increases, the fouling rate, which is the slope of the TMP curve, increases. Nevertheless, the fouling rates at same flux were different in some cases. For example, the fouling rate at 2 mg/L of silica was 9.05×10^{-5} bar/min when the flux increases from 100 L/m^2 h to 150 L/m^2 h. On the other hand, it was 3.02×10^{-4} bar/min when the flux decreases from 200 L/m^2 h to 150 L/m^2 h. It is evident that there is a hysteresis in TMP changes due to the irreversibility of membrane fouling. If the fouling layer is removed by relaxing the flux, the first and second fouling rates should be identical. Accordingly, the difference between these fouling rates, which may be defined as the degree of hysteresis, may be used as a quantitative measure to describe the characteristics of fouling.