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Determining flux behavior via a modified flux-step method for surface water treatment: pilot-scale ultrafiltration membrane operation

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HIGHLIGHTS

- Flux behaviors for surface water were determined via a modified flux-step method.
- The proposed flux-step method was evaluated with an ultrafiltration membrane.

• We determined threshold flux and critical flux for irreversibility.

- Long-term operation was achieved above the threshold flux with a cleaning regime.
- Membrane fouling and the TMP increment were minimized in turbid long-term tests.

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ABSTRACT

We present an alternative method for determining permeate flux behavior for membrane filtration of surface water. A modified flux-step method was developed and evaluated for two different submerged ultrafiltration membrane systems. Experimental results for a short-term pilot-scale system indicated that the modified method was able to determine the critical flux for irreversibility, improving on the conventional flux-step method. Using the modified method, the total membrane fouling rate and irreversible fouling rate can be used to simultaneously determine the threshold flux and critical flux for irreversibility. Based on the fluxes obtained in short-term tests, the long-term performance of the membrane system was also evaluated under operational conditions above the threshold flux and frequent variations in the turbidity of the surface water influent, combined with a cleaning regime. At a flux of $50 \text{ L/m}^2 \cdot \text{h}$ for membrane A and $40 \text{ L/m}^2 \cdot \text{h}$ for membrane B, the membrane fouling and fouling history were improved by cleaning through backwashing, draining of the membrane tank, and intermittent chemically enhanced backwash. Thus, by minimizing membrane fouling and transmembrane pressure increments based on the modified flux-step method, the membrane separation system cleaned using these methods achieved efficient surface water treatment under realistic operating conditions at high flux.

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1. Introduction

Filtration processes for surface water treatment are the fastestgrowing membrane technology, with an estimated global market of US \$1200 million in 2007, increasing rapidly at an average annual growth rate of 21% with a predicted value of US \$8300 million in 2016 [1]. The number of membrane filtration plants treating surface water in Korea is estimated to increase 7-fold from 2007 to 2016, consistent with growth in the global market [2]. Membrane technology offers many advantages over conventional processes, such as a small footprint, easier process maintenance, and higher effluent quality with a short operating time. However, membrane fouling remains one of the major challenges of this technology [3–5]. A number of ongoing studies are examining effective control of membrane fouling to optimize permeate flux over long-term operation. Several concepts have been proposed to characterize permeate flux behaviors, including critical flux, sustainable flux, threshold flux, critical flux for irreversibility, and limiting flux [6,7]. Sustainable flux incorporates economic considerations, whereas the other flux concepts are solely related to fouling accumulations on the membrane.

These critical flux concepts describe formation of a cake or gel layer on the membrane surface by particles or colloids [8]. Formation of this layer depends on factors such as characteristics of the materials in the influent, membrane properties, interactions between the membrane surface and particles, and shear and Brownian diffusion at the membrane surface. These factors ultimately determine whether the backtransport velocity of material from the membrane counterbalances the convection of these materials towards the membrane by permeate drag flow [9]. Previous studies have determined that below the critical

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flux there is no transmembrane pressure (TMP) increment or decline in flux with operating time [6,7,10–14]. At or below the threshold flux, the membrane fouling rate is independent of the permeate flux, whereas above it, the fouling rate increases with the permeate flux. The critical flux for irreversibility represents the transition between reversible and irreversible fouling, and the limiting flux is the plateau reached by the permeate flux with increased pressure. The threshold flux and critical flux for irreversibility are more relevant to industrial applications, because both extremely low flux with no fouling and high flux with heavy fouling should be avoided [6].

These flux parameters (other than sustainable flux) are often measured by the flux-step method (termed "conventional method" here) via short-term experiments. The method applies successive fluxstepping tests at a constant interval up to a maximum flux and ramps down over the same duration. Several authors have introduced fluxstep methods based on parameters such as total membrane fouling rate, instantaneous TMP increase, average TMP, and change in permeability [15,16]. These experiments have determined the rate of total membrane fouling and have introduced the term "threshold flux" to differentiate between low and high total fouling rates [7]. Each flux step in the test sequence below the threshold flux shows a constant low TMP. An increasing fouling rate occurs above the threshold flux, because more material (i.e., fouling history) will have been deposited at higher fluxes. The critical flux for irreversibility is determined by irreversible fouling and is defined as the flux at which fouling cannot be removed by physical cleaning methods [6]. A previous study showed that the critical flux for irreversibility could be determined by an improved fluxstep method using applied relaxation techniques [9]. The reversibility of fouling clearly depends on the physical cleaning techniques applied. Therefore, the type of cleaning method should always be considered in defining the critical flux for irreversibility.

Several studies have suggested different forms of flux-stepping to observe membrane fouling, but a general flux-step method is necessary for industrial applications [17–21]. Because physical cleaning is required for membrane separation processes, characterization of permeate flux behaviors for operating membrane processes over the short-term should also incorporate physical cleaning techniques; backwashing and/or draining the membrane tank were employed in this study. The objective of this study was to determine the threshold flux and critical flux for irreversibility via a modified flux-step method incorporating the two physical cleaning techniques described above. Our experimental surface water treatment tests used two different submerged ultrafiltration (UF) membrane systems in combination with physical cleaning. The critical flux for irreversibility was also determined via the proposed method and compared with the conventional method. Based on the values of threshold flux and critical flux for irreversibility obtained in the short-term tests, long-term operation of these membrane systems was also assessed under more realistic operational conditions of high flux combined with membrane cleaning during a period when the influent surface water had frequent variations in turbidity.

2. Materials and methods

2.1. Experimental setup

A pilot-scale setup was installed in an existing water treatment plant (Fig. 1). The pilot plant was fed with raw water influent from the Imjin River in the North Han River Basin, Korea. The influent was pumped directly from a receiving well to the influent raw water tank.

The pH and temperature of the influent were regularly recorded by a DYS DWA-2000A-pH meter (Korea). Turbidity and Cl₂ concentrations were measured using a HACH sc200 universal controller (USA). Total organic carbon (TOC) and dissolved organic carbon (DOC) concentrations were measured using a GE 5310C analyzer (USA) and UV₂₅₄ was determined with a HACH DR/4000 spectrophotometer (USA). Fe and Mn concentrations were measured using a PerkinElmer Optima 8300 ICP-OES (USA) using the standard 3120B digestion method [22]. All samples were analyzed 3 times/week for 3 months. The average concentrations, standard deviations (SDs), and relative standard deviations (RSDs) for the major constituents in the raw water are shown in Table 1.

Each polyvinylidene fluoride (PVDF) hollow-fiber UF membrane unit (A: Cheil Industries, Inc.; B: Zenon Environmental, Inc.) was submerged in a separate 1.2 m^3 water tank. The nominal pore sizes of membranes A and B were $0.03 \mu\text{m}$ and $0.04 \mu\text{m}$, respectively, with overall membrane surface areas of 37.9 m^2 and 40.8 m^2 and membrane length of 1.9 m. Membrane A is newly developed and is not yet commercially available, so a membrane A product with similar characteristics to membrane B was employed in this study. Filtration was terminated at the allowable TMP of 70 kPa suggested by the supplier. The TMP was measured with Sensys SSGC pressure sensors (Korea). A PC running Wonderware InTouch software (USA) controlled the peristaltic pump by automatically setting a constant flux for the desired experimental test.

2.2. Operating conditions

The two membrane systems were operated at several fluxes under conditions of constant cross-flow of air combined with frequent



Fig. 1. Schematic of the pilot-scale plant for surface water treatment using two different submerged UF membranes.

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