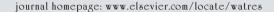


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# Comparison of fouling characteristics of two different poly-vinylidene fluoride microfiltration membranes in a pilot-scale drinking water treatment system using pre-coagulation/sedimentation, sand filtration, and chlorination

### So-Ryong Chae<sup>a,\*</sup>, Hiroshi Yamamura<sup>b</sup>, Keiichi Ikeda<sup>c</sup>, Yoshimasa Watanabe<sup>b</sup>

<sup>a</sup>Department of Civil and Environmental Engineering, School of Engineering, 121 Hudson Hall, Duke University, Durham, NC 27708, USA <sup>b</sup>Division of Built Environment, Graduate School of Engineering, Hokkaido University, Nishi 8, Kita 13, Kita-ku, Sapporo 060-8628, Japan <sup>c</sup>Water Treatment Technology Center, TORAY Institutes Inc., 3-3 Sonoyama 3-chome, Otsu, Shiga 520-0842, Japan

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#### ABSTRACT

Two pilot-scale hybrid water treatment systems using two different poly-vinylidene fluoride (PVDF) microfiltration (MF) membranes (i.e. symmetric and composite) were operated at a constant permeate flux of  $104.2\,l\,m^{-2}\,h^{-1}$  (=  $2.5\,md^{-1}$ ) with a pre-coagulation/sedimentation, sand filtration (SF), and chlorination to produce potable water from surface water. Turbidity was removed completely. And humic substances, Al, and Fe were removed very well by the pilot-scale membrane system. To control microbial growth and mitigate membrane fouling, a NaOCl solution was injected into the effluent from SF before reaching the two membranes (pre-chlorination). However, it adversly affected membrane fouling due to the oxidization and adsorption of inorganic substances such as Al, Fe, and Mn. In the next run, the NaOCl was introduced during backwash (post-chlorination). As compared with the result of pre-chlorination, this change increased the operating period of the symmetric and the composite membranes from about 10 and 50 days to about 60 and 200 days, respectively.

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### 1. Introduction

As a means of complying with current and anticipated water quality regulations, membrane technologies have been widely adopted in the world. In particular, low-pressure membrane techniques such as microfiltration (MF) and ultrafiltration (UF) have attracted a considerable amount of attention for removal of particulates by size exclusion, and usually produce a filtrate

free of turbidity and bacteria from river, lake, and underground waters (Bian et al., 1999; Hagen, 1998; Kimura et al., 2004; Klijn et al., 2000; Ma et al., 1998; Yuasa, 1998). An advantage of the MF membrane material is that it can be adapted to most requirements by easy modification of its chemical composition, which makes low-cost production possible. Before the membrane is fully ready for field application, however, some of its limitations must be addressed.

Nomenclature		J	pure water permeability after each chemical
K <sub>w</sub> K <sub>wo</sub> J <sub>T</sub> J <sub>0</sub>	permeability coefficient ( $lm^{-2}min^{-1}$ ) at 1 atm and 20 °C permeability coefficient ( $lm^{-2}min^{-1}$ ) of the new membrane at 1 atm and 20 °C permeate flux ( $lm^{-2}min^{-1}$ ) at T °C pure water permeability of a new membrane at 20 °C ( $lm^{-2}min^{-1}$ )	$\begin{array}{c} P_{\rm f} \\ P_{\rm p} \\ \Delta P_{\rm T} \\ \mu_{\rm T} \\ \mu_{\rm 20} \end{array}$	cleaning at 20 °C (l m <sup>-2</sup> min <sup>-1</sup> ) feed stream pressures (Pa) permeate stream pressures (Pa) trans-membrane pressure, TMP (kPa) at T °C dynamic viscosity of permeate (Pa*s) T °C dynamic viscosity of permeate (Pa*s) 20 °C

First of all, there is often a rapid decline in flux due to membrane fouling, which is caused by colloidal and soluble natural organic matter (NOM), as well as inorganic matter. To remove these substances and to mitigate membrane fouling, a hybrid membrane system combined with chemical/physical processes such as coagulation/sedimentation, ozonation, chlorination, and activated carbon adsorption has been widely adopted (Chang et al., 2005; Jang et al., 2005; Lee et al., 2004; Tomaszewska and Mozia, 2002; Watanabe et al., 1998). For an example, use of the coagulation/sedimentation process extended the membrane operation period by reducing cake deposit on the membrane and removing dissolved organic matter including humic substances (Jang et al., 2005). Moreover, hybrid membrane systems combined with chemical or physical processes enhanced the removal of micropollutants and virus (Fane, 1996).

A suitable membrane must have high permeability, be strongly hydrophilic, and have excellent chemical resistance to the feed streams. In order to obtain high permeability, membranes should have high surface porosity and good pore structure. Polyvinylidene fluoride (PVDF) is a polymer receiving increased scientific attention and gaining industrial importance because of its outstanding properties: good electrical insulation and chemical resistance, well-controlled porosity, good thermal properties, durability and biocompability (Salamone, 1999).

Regarding these merits, PVDF membranes have recently been adopted due to their tolerance of oxidants during drinking water treatment (Kurihara et al., 2004; Lee et al., 2004). The effectiveness of membrane filtration depends on the raw water quality and the operating conditions. Therefore, it is still necessary to study the performance characteristics and fouling of the PVDF membrane under various operating conditions.

A number of module designs such as plate (flat sheet), spiral-wound, tubular, capillary, and hollow fiber modules are available. In order to apply membranes on a technical scale, large membrane areas are normally required. Among these, the hollow fiber module has the highest packing density (Mulder, 1996).

Membranes may be classified by morphology or structure. When solid synthetic membranes are considered, two types of membrane can be distinguished, i.e. symmetric or asymmetric membranes. In symmetric membranes (porous or nonporous), the resistance of mass transfer is determined by the total membrane thickness. However, the resistance of mass transfer in asymmetric membranes is determined largely or completely by the thin top layer (Mulder, 1996).

Childress et al. (2005) reported that asymmetric membranes generally provide similar rejection with lower intrinsic hydraulic resistance than symmetric membranes. However, there were no available reports comparing the process performance and membrane fouling characteristic of symmetric and asymmetric MF/UF membranes used in drinking water treatment. Focused on those points, this study was aimed at investigating both process performance and fouling characteristics of the new PVDF MF membranes having different structures (i.e. symmetric and composite), after coagulation/sedimentation and sand filtration (SF) with chlorination.

#### 2. Material and methods

#### 2.1. Raw water

In this study, Chitose River surface water was used as raw water, which was obtained from the Kami-Ebetsu Water Purification Plant in Ebetsu, Japan. Although the water quality of Chitose River changes seasonally, it typically contains relatively high turbidity, organic matter, humic substances, and inorganic matter (Jang et al., 2005; Kimura et al., 2004; Lee et al., 2004).

# 2.2. Experimental set-up for a pilot-scale hybrid membrane filtration system

A pilot-scale hybrid membrane filtration system was composed of pre-coagulation/sedimentation, SF, and membrane filtration using two PVDF MF membranes with different structures. One (HFS module, TORAY Industries, Inc., Japan) is a composite membrane having a skin layer (pore size =  $0.02\,\mu m$ ) on a support layer (pore size =  $0.1\,m$ icron) and the other (HFM module, TORAY Industries, Inc., Japan) is a symmetric membrane ( $0.1\,\mu m$ ). Cross-sectional images of the two membranes were observed by FE-SEM (S-800; Hitachi, Tokyo, Japan). According to the manufacturer, the HFM module is suitable for very low turbidity water which hardly contains turbidity and NOM. And the HFS module is suitable for surface water such as river water or lake water which contains turbidity and NOM, and so on (Minegishi et al., 2007).

For coagulation and sedimentation, the jet mixed separator (JMS) with 84-min HRT was used (Watanabe et al., 1998) for the whole experiment. Polyaluminium chloride (PACI) was used as a coagulant, and dosages of PACI were controlled to maintain turbidity below 0.3° in the effluent from the JMS.

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