



# Membrane bioreactor pilot machine test for a newly developed membrane fabricated from chlorinated poly (vinyl chloride) and non-woven poly (ethyleneterephthalate) and its anti-fouling properties



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## ARTICLE INFO

### Article history:

Received 18 May 2015

Received in revised form

27 September 2015

Accepted 8 October 2015

Available online 27 October 2015

### Keywords:

MBR

Pilot-machine test

Fouling

Membrane

Hydrophilicity

## ABSTRACT

We have succeeded in fabricating a flat sheet-type microfiltration membrane for anti-fouling bioreactors. The membrane is made from chlorinated poly (vinyl chloride) and non-woven poly(ethyleneterephthalate), and hydrophilized with hydroxyl-propyl cellulose. The membrane has a novel structure in which small hydrophilic gels are homogeneously attached. This membrane was tested for one year in a bioreactor to treat sewage and estimate the proceeding state of membrane fouling. The membrane shows good anti-fouling properties compared with a similar commercially available membrane fabricated from same materials with a different hydrophilization process. The filtration resistance of the foulants deposited on the membrane was analyzed. The foulants were decomposed into their organic, inorganic, and irreversible forms. Due to membrane fouling, the resistance of the developed membrane is relatively less than that of the purchased one.

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## 1. Introduction

Membrane fouling is a phenomenon that results in the closure of membrane pores, wherein the organic materials that are produced by microbial activity become foulants [1]. The microbial activity digest the components of organic pollutants from the wastewater in the activated sludge in a bioreactor and sometimes produce extracellular pollutants, which are composed of polysaccharides and polypeptides. These two materials are hydrophobic and may interact with other hydrophobic materials, including the ones that form the body of the membrane, and deposit in the pores of membrane. Here, the activated sludge is an important material that can treat the pollutants in wastewater and produce clean water. The need to separate clear water from other materials in a bioreactor led to the development of membranes [2]. A micro-filtration (MF) membrane is placed in a bioreactor and the treated water is sucked up with a pump through membrane, which has small pores to allow the passage of water, but exclude other turbid materials such as *E. coli* in the activated sludge. This separation ability is useful, because we can get safe water that is not contaminated

with microbes and pollutants. One of other characteristics of this method is that the membrane is in direct contact with the activated sludge. It might be possible to accelerate interactions between the materials in water that may cause the membrane pores to close. In most cases, polymeric membranes for the bioreactor are made from a hydrophobic polymer. Foulants in the water interact with hydrophobic membrane surfaces, and the probability that foulants deposit in the membrane pores is not low. Mitigating membrane fouling in the pores is a challenge.

We will discuss the suitable materials that can be used in membrane bioreactors (MBRs). The membrane should have a pore size in the range of 0.01–0.5  $\mu\text{m}$ , because they must filter activated sludge and microbes, including *E. coli*, but allow clean water to easily pass through it, which is sucked up by a pump. The energy consumption for the pumping operation should be minimized. For this purpose, the membrane pore area should be kept as wide as possible, because the porosity, the openness of pores, and the membrane thickness are the factors that affect the filtration resistance of water passage. The pore-size distribution is also expected to be narrow so that the membrane can lead to high-resolution separation of the materials. To meet the above mentioned conditions for MBRs, polyethylene [3], polyvinylidene [4], and polytetrafluoroethylene [5] are often chosen as membrane materials.

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## Nomenclature

$A$	effective area of membrane ( $\text{m}^2$ )
$D$	diameter of the pore ( $\mu\text{m}$ )
$J_w$	water flux ( $\text{m}^3/\text{m}^2/\text{s}$ )
$L_p$	water permeability ( $\text{L}/\text{m}^2/\text{h}/\text{kPa}$ )
$p$	value of the applied pressure (kPa)
$P$	the bubble point (MPa)
$\Delta P$	pressure difference between the feed side and the permeate side through the membrane (Pa)
$Q$	quantity of outflow water through the membrane in a unit time (L/h)
$R$	filtration resistance ( $\text{m}^{-1}$ )
$R_{\text{in}}$	filtration resistance of inorganic foulants ( $\text{m}^{-1}$ )
$R_{\text{irr}}$	filtration resistance of irreversible foulants ( $\text{m}^{-1}$ )
$R_{\text{memb}}$	filtration resistance of membrane ( $\text{m}^{-1}$ )
$R_{\text{org}}$	filtration resistance of organic foulants ( $\text{m}^{-1}$ )
$R_{\text{other}}$	filtration resistance of all the other contributions ( $\text{m}^{-1}$ )
$R_t$	filtration resistance of the total contributions ( $\text{m}^{-1}$ )
$\sigma$	surface tension at the solvent/air interface (N/m)
$\eta$	viscosity of the water (Pa s)
$\theta$	contact angle of the liquid/solid (degree)

Chlorinated poly(vinyl chloride) (CPVC) is widely used in MBRs [6–10], because it exhibits an open-pore structure with high porosity and a narrow pore-size distribution. This membrane structure is formed according to the phase-separation principle [11], but it is hydrophobic and may cause membrane fouling, which results in membrane pore closure. There are only a few publications that discuss the hydrophilization of CPVC membranes. One of the procedures to reduce the hydrophobicity is to blend hydrophilic materials in a membrane. A proposal to blend sucrose fatty acid ester is presented as a patent [12]. This additive will mitigate the interaction between membranes and foulants, resulting in less fouling, but more force is required to keep the materials inside the membrane structure. Thus, in the long term, it may fall off the membrane in a bioreactor and cause pore closure.

A cross-linking method has been proposed to fix hydrophilic materials to a membrane [9]. Polyvinyl pyrrolidone (PVP) is blended when the membrane is fabricated through phase separation from a solution of raw materials, which include solvents, additives, and CPVC. The resulting membrane is subjected to high temperature to accelerate the cross-linking reaction of PVP with initiators for radical reaction, which increases the ability of the PVP to stick to the CPVC membrane for an extended period of time. However, it exhibits relatively larger pores with broad pore-size distribution, because the PVP additive in the solution may disturb the phase-separation condition, resulting in a relatively wider pore-size distribution.

We propose a new method to make a hydrophilized CPVC flat-sheet using hydroxyl-propyl cellulose (HPC). This procedure consists of two separate processes as follows: phase separation and hydrophilization with HPC. The phase-separation condition can be determined independently with the hydrophilization process. Therefore, pore size and its distribution on membranes can be controlled independently and carefully. This HPC for enhancing the hydrophilicity is fixed with a finely dispersed particle gel on a CPVC membrane structure, the characteristics of which have not yet been reported in any scientific or engineering journals. The details of the preparation process and its membrane properties will be presented in a separate paper [13].

In this article, we discuss to a great extent, the mechanism by which HPC treatment can reduce membrane fouling. To

demonstrate the difference between our developed membrane and the commercially available one, which was purchased from another company, a pilot machine test was performed for one year. The pilot machine treated sewage (municipal wastewater) and was installed at the Konan-Chubu Wastewater Treatment Plant, located in Kusatsu city, Shiga prefecture, Japan. A brief summary of the single-year operation is explained later. Much emphasis is placed on analyzing the degree and mechanism by which the membranes are protected against fouling after being used for one year in a bioreactor. Our developed membrane is proven to exhibit good anti-fouling properties, and the hydrophilization with HPC can better prevent membrane pore closure by the foulants.

## 2. Experimental

### 2.1. Sample membranes

We have developed a new flat sheet-type micro-filtration membrane made from CPVC and non-woven PET (poly(ethyleneterephthalate)), hydrophilized with HPC [13]. The non-woven PET was dipped in a CPVC solution diluted with a mixture of tetrahydrofuran and polyalcohols and then subjected to a drying process. The resulting material was covered with a network structure made from CPVC fibrils. Subsequently, it was guided to a HPC solution bath and a hot chamber. The membrane sheet became hydrophilic after it passed through this HPC application process. The small gel particles made from HPC with an average diameter of 45 nm were attached homogeneously on the CPVC fibrils.

The fact that HPC is in gel form attached on the surface is important, because we believe that the long-standing hydrophilicity of this membrane would result from this configuration. The HPC solution generally transforms from a sol to a gel with heat [16]. The gel does not dissolve in water and we utilize this irreversible change with temperature as a fixation method. The membrane is reported to be operated stably at a flux of 1.0 m/d (41.7 LMH) for one month in a bioreactor [13]. Realizing the operation at this flux was one of the challenges and only a few membranes can be stably operated at this operating condition [14,15]. A commercially available flat-sheet membrane for MBRs was used as a comparative case. This membrane was also made from CPVC and non-woven PET, but there was no reference in the literature to tell how to increase its hydrophilicity.

### 2.2. Pore flow porometry

Pore-size distribution on the membrane was measured using a flow CFP-1200 APEX capillary porometer, supplied by Porous Materials, Inc. (USA). This apparatus is used to perform the liquid replacement method. First, the sample membrane was thoroughly wetted with a Galwick liquid having a low surface tension of  $1.57 \times 10^{-2}$  N/m. The wetted membrane was then subjected to pressurized air at room temperature. The pressure was then raised and the wet flow rate of air passing through the membrane was measured as a function of the applied pressure. Here, the flow never occurred if the applied pressure was below the critical point, which is governed by the following equation.

$$P = \frac{4\sigma\cos\theta}{D} \quad (1)$$

where  $P$  is the bubble point,  $\sigma$  is the surface tension at the solvent/air interface,  $\theta$  is the liquid/solid contact angle, and  $D$  is the diameter of the pore. The applied pressure and the diameter of the pore is related to this equation assuming  $\theta = 0^\circ$ . The dry flow rate of the sample membrane without dipping in the low-surface-tension liquid was also measured. Therefore, pore-size distribution can be

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