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Effect of membrane surface morphology on membrane fouling with sodium alginate

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ABSTRACT

The object of this study is to clarify the effect of membrane surface morphology on membrane fouling with sodium alginate (SA) which is a kind of natural organic matter (NOM) and is considered to be the main foulant in membrane water treatment. Cellulose acetate butyrate polymer which is superior in heat-resistance and mechanical strength was used as membrane material. Four kinds of membranes having different surface and cross-sectional structures were prepared via thermally induced phase separation and non-solvent induced phase separation. In the case of the filtration of 50 mg L^{-1} SA solution from the outer surface to inner surface, the membrane having the dense outer surface showed higher permeability and higher recovery of permeability by backwashing. An addition of calcium chloride into the sodium alginate solution resulted in significant flux decline due to thick cake layer formed by cross-linking of sodium alginate. In this case, the membranes with the dense outer surface also showed higher permeability. Furthermore, it was found that membrane fouling with sodium alginate took place mainly on the outer surface rather than inside the membrane by measuring infrared absorption spectrum of the membrane after filtration of sodium alginate solution with and without calcium chloride.

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1. Introduction

Membrane technology has spread to the water processing applications such as drinking water purification and waste water treatment as well as industrial applications such as semiconductor industry and pharmaceutical and food industries. Membrane technology using microfiltration (MF) and ultrafiltration (UF) membrane can remove particle material from feed water, including microorganisms such as protozoa (Giardia and Cryotosporidium) and has potential to meet stringent water quality regulations [1]. Membrane technology also has some benefits such as ease of operation, reduction of chemicals and small footprint.

Although membrane technology has many advantages superior to existing technology, its spread is limited by a problem of so-called membrane fouling. Membrane fouling, which increases filtration resistance, results in increase of energy consumption, chemical consumption due to frequent chemical washing, and membrane replace cost [2]. Operating costs including membrane replacement cost strongly influence the overall cost-effectiveness of membrane systems [3]. Many studies have been carried out to understand and control membrane fouling. Membrane fouling is classified into two categories [4–6]. One is a reversible membrane fouling to be removed by physical cleaning such as hydraulic backwashing. Another is an irreversible membrane fouling not to be removed by physical cleaning and to be removed by chemical cleaning. Natural organic matter (NOM) existing in surface or grand waters, which is a complex heterogeneous mixture composed of humic substances, carbohydrates, proteins and other compound [7,8], is considered to be the main foulants to cause irreversible membrane fouling [9–20].

Some researchers reported that hydrophobic components in NOM such as humic substances were contributed to membrane fouling [9–12]. Yuan and Zydney examined membrane fouling with humic acid for MF [9] and UF [10] membranes and found that the large flux decline was caused by the formation of a humic acid deposition located on the upper surface of the MF membrane, while humic acid adsorption on the UF membrane caused relatively little flux decline. Katsoufidou et al. [11] carried out the filtration experiments and modeling for UF membrane fouling with humic acid and found that a relatively rapid irreversible fouling took place due to adsorption, and pore blocking and cake formation caused flux decline for a long time. On the other hand, some researchers reported that hydrophilic components in NOM such as polysaccharide-like and/or protein-like substances were con-

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tributed to membrane fouling [13–19]. Cho et al. [13] examined UF membrane fouling with NOM isolated from the natural water and found that the foulants were the larger-sized neutral and/or basic NOM components, and not the humic substances. Carroll et al. [14] and Fan et al. [15] also fractionated NOM from the natural water on the basis of hydrophobicity and charge, and found that neutral hydrophilic components had higher fouling potential compared with hydrophobic acids. Jermann et al. [20] examined UF membrane fouling with individual and mixed humic acid and alginate (polysaccharide). They found that the fouling layer formed for the mixed solution was less reversible than that by alginate alone due to a bridge formation by humic acid between alginate and the membrane. Furthermore, in many previous studies, it was reported that the membrane fouling with NOM could be influenced by the operating conditions such as permeate flux and cross-flow velocity, and the pretreatments such as addition of coagulant [5,21–23].

Amy and Cho suggested that NOM rejection and membrane fouling were effected by membrane characteristic as well as NOM characteristic [21]. However, few studies have been carried out about the influence of the characteristics of the membrane on membrane fouling regardless of its importance in membrane fouling. Lee et al. [24] elucidated that UF membrane with smother surface had less fouling potential, while MF membrane with rough surface resulted in severe flux decline due to deposition of fouling materials in the valleys of the membrane surface. Fu et al. [25] prepared cellulose acetate butyrate (CAB) hollow fiber membranes with various surface structures and examined the membrane fouling with humic acid. They elucidated that the CAB membrane with lower humic acid rejection and lower surface porosity resulted in significantly flux decline.

The object of this study is to elucidate the effect of membrane surface morphology on membrane fouling for the better understanding and controlling of the membrane fouling. We prepared the hollow fiber membranes of the different surface and cross-sectional structures made of CAB polymer. The effect of the membrane morphology on membrane fouling was investigated with sodium alginate as hydrophilic NOM instead of humic acid as hydrophobic NOM used in the previous study [25]. We also examined the effect of addition of calcium chloride on the membrane fouling with sodium alginate. Furthermore, the distribution of sodium alginate in the CAB membrane after filtration was examined by using infrared absorption spectrum measurement.

2. Experimental

2.1. Materials

Cellulose acetate butyrate (CAB, Mw = 65,000, Daicel Chemical industries, Japan) was used as a membrane material. Triethylene glycol (TEG, Wako Pure Chemical Industries, Japan) was used as a diluent for membrane preparation via thermally induced phase separation. Dimethyl sulfoxide (DMSO, Wako Pure Chemical Industries, Japan) was used as a solvent for membrane preparation via non-solvent induced phase separation. The filtration solution of sodium alginate (SA) was prepared by mixing sodium alginate (Aldrich Chemical Co., Milwaukee, WI) with sodium bicarbonate as buffer (Wako Pure Chemical Industries, Japan). In some experiments, calcium chloride (Wako Pure Chemical Industries, Japan) was also added into the sodium alginate solution. All chemicals were used without further purification.

2.2. Preparation of hollow fiber membranes

Hollow fiber membranes were prepared by using a batch-type extruder via thermally induced phase separation (TIPS) and non-solvent induced phase separation (NIPS) under the preparation conditions shown in Table 1 according to the previous preparation method [25]. The CAB membranes via TIPS were prepared by the following preparation method. CAB polymer and TEG were put into the vessel equipped with a stirrer and were heated to 443 K. Then the mixture was blended for 1 h and was hold for 2 h without blending to remove air bubble from polymer solution. The homogeneous polymer solution was fed to a spinneret with a gear pump by using pressure of N_2 gas. The spinneret consists of inner tube with a diameter of 0.83 mm and outer tube with a diameter of 1.58 mm. The hollow fiber was extruded from the spinneret with TEG as an inner coagulant and was introduced into water bath with temperature of 323 K where phase separation proceeded and porous structure was solidified. The hollow fiber was wound at a take-up speed of 0.21–0.26 m s^{-1}. Finally, TEG remained in the membrane was extracted by water.

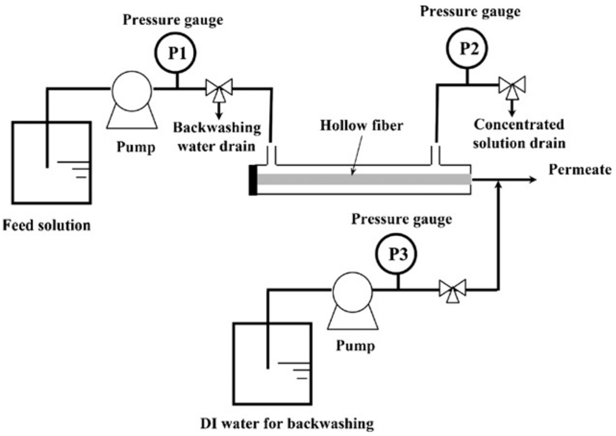

Fig. 1. Schematic diagram of the filtration-backwashing experiments using the single hollow fiber module.

The preparation method of CAB membranes via NIPS was as follows. CAB polymer was dissolved in DMSO at room temperature for 12 h to obtain homogenous polymer solution. After defoaming, the homogenous polymer solution was extruded with the inner coagulant of water and DMSO mixture from a spinneret at a spinning rate of 0.27 m s^{-1}. The spinneret consists of inner tube with a diameter of 0.7 mm and outer tube with a diameter of 1.0 mm. The extruded hollow fiber was passed through air gap of 30 mm and was introduced into coagulation bath at 298 K to induce phase separation and solidify membrane. The hollow fiber was wound at a take-up speed of 0.23 m s^{-1}. The hollow fiber membrane was immersed in water to remove DMSO in the membrane.

2.3. Filtration experiment

The schematic diagram of the filtration-backwashing experiments by using the single hollow fiber module is shown in Fig. 1. Feed solution was fed to the module packed with a single hollow fiber of length of about 110 mm by the pump. Cross-flow filtration was carried out from outer surface to inner surface of the hollow fiber at transmembrane pressure of 50 kPa. The cross-flow rate was 0.04 m s^{-1}. For each filtration experiment, deionized (DI) water was permeated as a feed at first and the pure water permeability J_0 was measured. Then, feed solution, which consists of 50 mg L^{-1} sodium alginate and 0.5 mmol L^{-1} sodium bicarbonate of as a buffer solution (pH = 8), was filtrated and the permeability of the sodium alginate solution J was measured. After filtration of one hour, backwashing by using DI water was carried out at pressure of 100 kPa for a minute. Filtration-backwashing cycle was repeated four times. During filtration of sodium alginate, concentration of sodium algi-

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