



Correlation between particle deposition and the size ratio of particles to patterns in nano- and micro-patterned membrane filtration systems



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ABSTRACT

Recently, membrane surface patterning has attracted much attention as an innovative alternative to control membrane fouling that occurs with membrane filtration used in water and wastewater treatment. However, limited attention has been focused on patterned membranes with nano-scale features due to their difficult fabrication. As a result, there is a lack of research on membrane fouling by particle deposition occurring with a wide range of pattern sizes. In this study, we prepared patterned membranes with nano-scale hexagonally packed arrays using nanoimprint lithography as well as micro-scale patterned membranes. Filtration tests were conducted using the membranes in cross-flow ultrafiltration to demonstrate the effect of the size ratio of particles to membrane patterns on fouling by particle deposition on the membrane surface. We found that particle deposition was most efficiently mitigated by the patterned membranes when the size ratio was approximately 3. On the other hand, when the size ratio was much smaller than 3, particle deposition was significant and was nearly as much as that of non-patterned membranes. In addition, when the size ratio was larger than 3, particle deposition increased with the increase in the size ratio. We explained the correlation between particle deposition and the size ratio of particles to membrane patterns in terms of shear stress near the surface of the membrane patterns using a computational fluid dynamics simulation technique. We anticipate that this study will provide a deeper understanding of the particle deposition phenomena in nano- and micro-patterned membrane filtration.

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1. Introduction

Membrane processes for water and wastewater treatment have been extensively utilized over the past decades due to their benefits, including minimal required space of the plant, reliable quality of treated water, and effective disinfection [1–3]. However, the membrane processes still have a critical challenge of membrane fouling caused by colloids, organic compounds, and microorganisms. Membrane fouling is the primary cause of the increased operational and maintenance cost by deteriorating membrane performance and decreasing membrane life [4,5]. To alleviate the fouling problem, various studies have been conducted using biological [6,7], chemical [8], and physical [9] approaches. Among the studies, the control of surface topography has recently been introduced in membrane fabrication to reduce membrane fouling

by promoting shear stress and turbulence near the membrane surface [10]. Previous studies showed an increase in the critical flux due to superior anti-fouling properties of patterned membranes [11,12]. Culfaz et al. observed that micro-structured ultrafiltration (UF) hollow fiber membranes exhibited improvement in water flux, and their fouling reversibility was better than that of round fibers [13,14].

One interesting result reported by the previous studies is that foulants in a feed solution showed different deposition behavior according to their sizes under the condition of fixed pattern size [12,14]. In another case, the size and spacing of patterns were adjusted in accordance to the target organisms to prevent bacterial attachment and bio-fouling. The results indicated that the attachment of the bacterial cell was contingent on the feature dimensions [15–17].

To the best of our knowledge, however, no previous research on patterned membranes has investigated the effect of the correlation between sizes of foulants and membrane patterns on fouling

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mitigation over a wide range of membrane pattern sizes. Membranes with nano-scale patterns, up to a few hundred nanometers, have scarcely been investigated due to the difficulty in the fabrication of well-ordered nano-scale patterns on membrane surfaces. Gohari et al. prepared membranes with aligned nano-scale patterns by adding nanoparticles during a phase inversion process and demonstrated the importance of the orientation between surface patterns and feed flow direction on the fouling behavior [18]. However, the patterns used in their study were not sufficiently regular to distinctly clarify the effect of nano-scale patterns on fouling behavior. In addition, the study examined neither the comparison of the efficiency in fouling inhibition between patterned and flat membranes nor the influence of the interaction between foulant and pattern sizes on fouling behavior.

In this study, we prepared nano-patterned UF membranes using the nanoimprint lithography (NIL) method as well as micro-patterned membranes to elucidate the impact of the size ratio of particles to membrane patterns on particle deposition. Anodized aluminum oxide (AAO), which is well known as a regular nanoporous material, was used as a mold to obtain well-ordered patterned features on the nano-scale. In addition, the relationship between membrane fouling and the ratio of particle to pattern size was investigated in a cross-flow filtration system by varying the particle and pattern sizes.

2. Materials and methods

2.1. Materials

An AAO mold with nano-scale patterns was prepared using high purity aluminum sheets (Al 99.999%, thickness ~0.5 mm, Goodfellow Inc., UK). Additionally, monoglycidyl ether-terminated poly(dimethylsiloxane) (MET-PDMS, Aldrich, USA) was used to make the surface of the mold hydrophobic. Polyurethane acrylate (PUA) molds with micro-scale patterns were prepared using silicon wafers ((100), p-type, Silicon Technology Co., Ltd., Japan), polydimethylsiloxane (PDMS, Sylgard 184 kit, Dow Corning, USA), polyethylene terephthalate (PET) film, polyurethane acrylate oligomer (PUA, Minuta Technology Co., Ltd., Republic of Korea), and sodium dodecyl sulfate (SDS, Sigma–Aldrich, USA). Polyethersulfone (PES, BASF, Germany) was used as a membrane material. N-methyl-2-pyrrolidinone (NMP, Sigma–Aldrich, USA) was used as a solvent to prepare membranes. Glycerin (Duksan Pure Chemical, Republic of Korea) was utilized to prevent pore collapsing during the NIL process. The acids and organic solvents, including perchloric acid (HClO_4), oxalic acid ($\text{H}_2\text{C}_2\text{O}_4$), chromic acid (H_2CrO_4), hydrochloric acid (HCl), phosphoric acid (H_3PO_4), and ethanol, were purchased from Sigma–Aldrich, USA and were used without further purification. Aqueous suspensions of polystyrene latex microspheres (0.1, 0.5, 2, 5, and 6 μm in diameter, Alfa Aesar, USA) were utilized for colloidal filtration tests. Polycarbonate track-etched membranes with a pore diameter of 0.8 and 3 μm (Nuclepore[®], Whatman, GE Healthcare, USA) were purchased to separate mixed particles used in poly-dispersed particle filtration tests prior to determining each concentration.

2.2. Preparation of patterned molds for membrane fabrication

2.2.1. Nano-patterned mold

Nano-patterned AAO mold was prepared via a two-step anodization process [19] (Fig. 1). An aluminum sheet was prepared by electro-polishing in a mixed solution of ethanol and perchloric acid at 20 V and 4 °C to reduce surface roughness (Fig. 1a). Afterward, the electro-polished aluminum sheet was anodized in 0.3 M oxalic acid solution at 40 V and 15 °C for 12 h (Fig. 1b). After

the first anodization step, the nano-porous alumina layer was etched away with a mixture of chromic acid and hydrochloric acid at 65 °C (Fig. 1c). Subsequently, the second anodization was carried out under the same conditions as the previous anodization step except for an anodizing time of 100 s (Fig. 1d). To widen the AAO pore diameter, the porous AAO was immersed in 0.1 M phosphoric acid at 30 °C for 30 min (Fig. 1e). The porous AAO with a wider pore size was treated with MET-PDMS at 80 °C for 4 h to make its surface hydrophobic (Fig. 1f). The hydrophobic surface allows membranes to easily detach from the mold during membrane fabrication.

2.2.2. Micro-patterned mold

Concave dome-shaped micro-patterned PUA mold was prepared using the following procedure (Fig. 2). First, a hexagonally packed monolayer of particles on a silicon wafer was prepared via the self-assembling process [20]. A 2.5 wt% aqueous suspension of polystyrene latex particles with a diameter of 2 μm was diluted with a 4:1 (v/v) ethanol/polystyrene solution and was sonicated for 20 min to homogeneously disperse the particles. The diluted suspension (50 μL) was dropped onto deionized (DI) water to form a monolayer of the particles on the water surface. Then, a few drops of SDS were added to increase the rigidity of the particle monolayer. A piece of silicon wafer (3 cm \times 3 cm) treated with UV/ O_3 as a hydrophilic surface treatment was immersed in the water. By drawing the silicon wafer as shown in Fig. 2a (scooping method), the hexagonally packed polystyrene particle monolayer was formed on the wafer surface and was dried at room temperature (Fig. 2b).

Next, concave dome-shaped micro-patterns were introduced using a pattern-transfer technique. The PDMS prepolymer mixture (10:1 (w/w) base/curing agent) was poured onto the dried particle monolayer array and was cured at 60 °C for 4 h (Fig. 2c). The solidified PDMS with concave patterns was removed, and the PUA mixture was cast onto it (Fig. 2d). The PET film was gently placed on the PUA layer as a support. The PUA polymer replica with convex patterns was cured under UV light ($\lambda = 360 \pm 20 \text{ nm}$). Similarly, the micro-patterned PUA mold was obtained by repeating this replication using PUA, except using another wafer as the support of PUA instead of PET film (Fig. 2e). Finally, the concave dome-shaped micro-patterned PUA mold was obtained on the wafer support (Fig. 2f).

2.2.3. Non-patterned mold

A non-patterned PUA mold with a flat surface was prepared to fabricate non-patterned membranes as a control. A piece of silicon wafer was modified using UV/ O_3 in advance and was treated with MET-PDMS to make a hydrophobic surface.

2.3. Preparation of patterned membranes using molds

A schematic procedure of the preparation of nano- or micro-patterned PES membranes using the molds is depicted in Fig. 3. A PES membrane was prepared by the conventional non-solvent induced phase separation (NIPS) method. A polymer solution consisting of 15 wt% of PES and 85 wt% of NMP was cast onto fabric with a casting knife (Fig. 3a). By dipping the cast PES on the fabric in DI water, phase separation was induced (Fig. 3b). The solidified PES membrane was dehydrated by dipping the wet membrane into a 50 wt% glycerol aqueous solution for 24 h (Fig. 3c) and drying it at room temperature for 24 h to prevent the collapse of the pore structure during imprinting of the patterned mold onto membranes with heat [21]. Then, the patterning process was conducted using a hot-press machine (V-SYSTEM, Republic of Korea). A patterned (or non-patterned) mold prepared previously by the methods described in Section 2.2 was placed on the dried PES

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