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Three-dimensional hydraulic modeling of particle deposition on the patterned isopore membrane in crossflow microfiltration

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ABSTRACT

Patterned membranes have been proposed as a promising solution to membrane fouling in membrane processes for water treatment. CFD modeling studies were carried out to elucidate the anti-fouling effect of patterned membranes However, patterned membranes prepared usually by a phase inversion method are more likely to have broad pore size distributions and most modeling studies are based on two-dimensional space despite the three-dimensional pattern geometry. In this study, a patterned isopore membrane with reverse-pyramid patterns on its surface and a narrow pore size distribution was prepared from UV-curable polymer by the soft lithographic method. Factors affecting particle depositions on patterned isopore membranes were investigated during the crossflow microfiltration of different micro-sized particles and their mixture. The extent of particle deposition was largely dependent on crossflow velocity, pore water flux, and particle size. Particularly, the ratio of crossflow velocity to pore water flux mostly governed the extent of particle depositions (i.e., membrane fouling) on the patterned membrane surface and 3-D modeling based on computational fluid dynamics was also conducted to predict the formation of two distinct stream lines (bulk and vortex) and elucidate the mechanisms of anti-fouling characteristics of patterned membranes.

1. Introduction

Membrane processes have been widely applied to water and wastewater treatments because of their relatively simple operation and high selectivity. However, membrane fouling greatly reduces the economic feasibility of membrane processes and thereby restricts their extensive spread. Membrane fouling is induced by attachment or sorption of foulants such as natural organic matter, microorganisms, and numerous particles on the membrane surface, which subsequently leads to water flux decline, reduction of membrane lifespan and so on [1,2].

A number of scientific and engineering approaches have been employed to mitigate membrane fouling. Among them, patterned membranes are an alternative way to reduce membrane fouling. Patterned membranes with various configurations and shapes have been prepared and tested. Patterned hollow-fiber membranes were fabricated from patterned nozzles and showed increased water flux compared with conventional hollow-fiber membranes [3–7]. Prism, pyramid, and embossed patterns were made on a flat sheet membrane surface, and all of them improved anti-fouling properties by inducing local turbulence or increasing wall shear stress at the upper region of pattern [8,9]. Modeling based on computational fluid dynamics (CFD) for hydraulic flow around the patterned membrane surface revealed that local shear stress distribution and vortex formation were the major reasons for anti-fouling effects of patterned membranes [10]. However, most CFD modeling studies were two-dimensional, despite the fact that patterns have three-dimensional geometry. Moreover, because the models did not often take into account water permeation through membrane pores, the results obtained did not precisely reflect hydraulic flow around the patterned membrane surface during filtration. Furthermore, the patterned membranes used for such tests were usually by a phase inversion method, and thus their pore size distributions were broad due to many factors such as solvent, non-solvent, additives, and so on. Broad pore size distributions would decrease the selectivity of target solutes and thus reduce water quality of the permeate [11,12]. Although there had been attempts using track-etched isopore membranes with relatively uniform pore sizes, their applications were still limited due to their defects such as doublet and triplet pores and insignificant

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anti-fouling property [13–16]. Recently, studies to reduce pore defects have been carried out by applying self-assembly, anodization, and microelectochemical system (MEMS) to isopore membrane fabrication processes [17,18]. The increased selectivity by ordered pore structure enabled enhanced separation of specific microorganism [19,20].

In this study, a patterned isopore membrane with reverse-pyramid patterns and a narrow pore size distribution as well was prepared by using UV-curable polymer and soft lithographic method. And then, factors affecting particle depositions on that membrane were investigated during the crossflow microfiltration of different micro-sized particles and their mixture. Three dimensional CFD modeling was also carried out to analyze and elucidate particle depositions on the patterned membrane surface at various operating conditions for crossflow microfiltration.

2. Materials and methods

2.1. Fabrication of patterned isopore membrane

Patterned isopore membranes were prepared by polymerization of uniformly coated polyurethane acrylate (PUA; 311RM, Minuta, Republic of Korea) oligomer solution [21] on a pyramid-patterned polydimethylsiloxane (PDMS; Sylgard 184, Dow, USA) replica mold after replication of a reverse-pyramid patterned master mold (Fig. 1). The master mold was fabricated by the photolithographic method using nano-patterning organic devices at Seoul National University. The PDMS prepolymer was mixed with curing agent at a weight ratio of 10:1 and cast on a master mold with reverse-pyramid patterns. After trapped air bubbles were removed at room temperature for 1 h, the mixed PDMS solution was cured for 3 h at 60 °C [22]. The cured pyramid-patterned PDMS replica mold was detached from the master mold. Then, UV-curable PUA oligomer solution was applied to the PDMS replica mold surface (Fig. 1a). The excess PUA oligomer solution was removed during the spin-coating step. In this step the final surface pore size of patterned isopore membrane was controlled by the spinning rate (Fig. 1b). In this study, the spinning rate was fixed at 1850 rpm, and total spin coating time was 30 s. After the spin-coating, PUA oligomer solution was UV-cured at 365 nm for 2 h (Fig. 1c). Polymerized PUA film was detached from the PDMS replica mold (Fig. 1d) and attached to the fabric support layer to increase mechanical strength. The PUA reverse-pyramid patterned isopore membranes were observed with a scanning electron microscope (SEM; JSM-6701F, Jeol, USA), and the pore size distribution was measured with NIS-Elements BR 3.2 (Nikon, Japan).

2.2. Particle depositions during crossflow microfiltration

The effects of the patterned isopore membranes on particle depositions were investigated during the crossflow microfiltration under various conditions such as cross-flow velocity and pore water flux. In the cross-flow microfiltration system (Fig. 2), the PUA membrane was installed on a lab-scale membrane module measuring 20 mm in channel length, 20 mm in channel width and 2 mm in channel height. A peristaltic pump was connected to the permeate line of the membrane module to control the pore water flux. The cross-flow velocity was controlled by a circulation pump and a valve next to the module and monitored with a flowmeter. 2 μm (2 wt%) and 5 μm (10 wt%) polystyrene latex bead suspensions (Sigma-Aldrich, USA) were used to prepare feed suspensions for the crossflow microfiltration and 160 μL of 2 μm or 5 μm suspension were diluted into 800 mL of deionized (DI) water. The mixed suspension of 2 μm and 5 μm latex beads was prepared by adding 35 μL of 2 μm latex bead suspension to 60 μL of 5 μm suspension and then the mixed suspension was diluted with 800 mL of DI water.

Operating conditions for microfiltration runs were as follows. Crossflow velocity (V_c) was selected as either 0.25 or 0.42 m/s, and operation pressure corresponding to each V_c was 8.3 or 21.1 kPa, respectively. Pore water flux (J_p), defined by water permeation rate through a membrane pore located at the bottom of each reversed-pyramid pattern, had the same unit as that of V_c (m/s). J_p was adjusted such that the ratio of V_c to J_p was 100 or 1000, i.e., V_c was 100 or 1000 times higher than J_p. When $J_p = V_c/100$ or $J_p = V_c/1000$, they were designated as "high J_p" or "low J_p", respectively. For instance, at $V_c = 0.25$ m/s with low J_p, low J_p is equal to $V_c/1000 = 2.5 \times 10^{-4}$ m/s. Crossflow microfiltration for 2 μm particles was conducted with a combination of $V_c = 0.25$ or 0.42 m/s and at high or low J_p. Those for 5 μm and mixed particles were performed with a combination of 0.25 or 0.42 m/s and only low J_p. After each filtration run, the PUA membrane covered with deposited particles was detached from the membrane module and analyzed by SEM.

2.3. Numerical method

Three-dimensional hydraulic modeling was conducted to analyze fluid stream lines in the vicinity of the isopores on the patterned membrane surface. The Navier–Stokes and fluid continuity equations for the incompressible Newtonian fluid were discretized and solved by the finite element method (FEM). A 3-D rectangular channel was constructed and used as a simulation domain where reverse-pyramid patterns were engraved on the bottom side. The overall domain size was 75 μm (width) × 150 μm (length) × 2000 μm (height), and the size of each pattern was 25 μm (width) × 25 μm (length) × 16 μm (height), which corresponded to the pattern size of the membrane used for the filtration experiment. The number of elements was 57,864, and fine meshes were generated near the membrane surface to solve complicated flow behavior with accuracy (Fig. S1 in Supporting information). The density and viscosity of the fluid (water) were assumed to be 1 g/mL and 1 g/(m s), respectively.

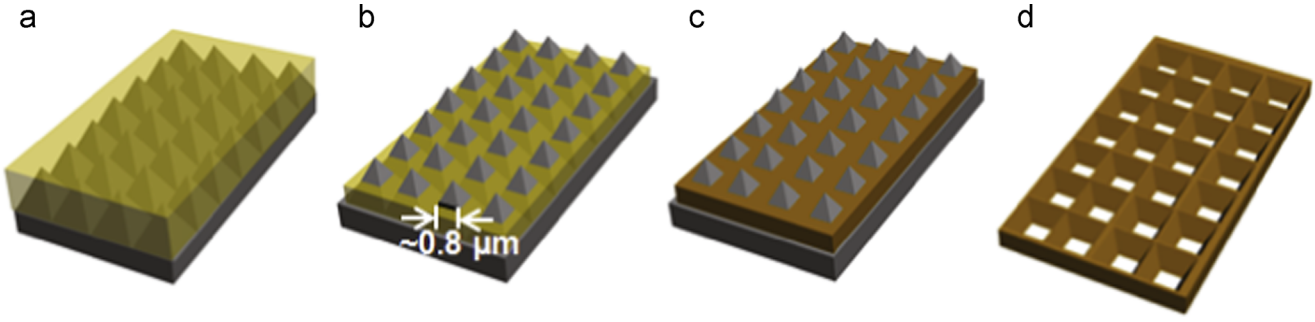

Fig. 1. Schematic diagram of fabrication steps for the patterned isopore membrane. UV-curable PUA precursor solution was (a) dispensed on a pyramid-patterned PDMS replica mold and (b) spin-coated at 1850 rpm. (c) The PUA oligomer solution was polymerized by UV-curing at 365 nm for 2 h. (d) Finally, reverse-pyramid patterned PUA membrane was detached from the PDMS mold.