Journal of Membrane Science 498 (2016) 14–19

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Correlation of membrane fouling with topography of patterned membranes for water treatment

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ARTICLE INFO

Article history:
Received 7 July 2015
Received in revised form
23 September 2015
Accepted 24 September 2015
Available online 28 September 2015

Keywords:
Patterned membrane
Antifouling
Prism pattern
Crossflow filtration
CFD modeling

ABSTRACT

Particle depositions on patterned membrane surface were experimentally measured and compared with those of non-patterned membranes. Prism patterns introduced to membrane surface significantly reduced particle deposition. A larger pattern was less effective against particle deposition than a smaller pattern under low Reynolds number, but was very successful in mitigating particle deposition under high Reynolds number at faster crossflow velocity. The particle deposition and anti-fouling mechanisms were analyzed using computational fluid dynamics simulation. A vortex was formed in the valley region between prism patterns, proposing that particles entering the valley region because of permeation drag had a chance to return back to bulk crossflow stream during flowing along with the vortex. The distance between the vortex and bulk stream was shorter under high Reynolds number than under small Reynolds number, suggesting that the return of particles in the valley region into the bulk stream was quite enhanced by increasing crossflow velocity. To further mitigate particle deposition on the valley region, new patterns were developed by introducing intervals to prism patterns and showed much improvement in antifouling ability by enhancing the vortex and reducing the portion of permeation stream in the valley region.

1. Introduction

Membrane fouling promoted by the unwanted accumulation of substances on membrane surfaces as well as pores is the most important problem in membrane processes since they subsequently increase operational cost [1–3]. There are two main approaches to mitigate the membrane fouling. By making a suitable choice of the membrane material [4,5] or modifying the membrane surface [6,7], the adsorption of foulants on the membrane can be minimized. In an effort to modify the membrane surface, introducing diverse patterns to the membrane surface has been paid attention to with advancements in lithography technology. An extensive amount of researches about the antifouling ability of surface patterns has been performed, and the potential of patterned surfaces for antifouling has been confirmed [8,9].

Efimenko et al. reported that the attachment of marine bacteria was effectively hindered by a hierarchically wrinkled surface [10]. This observation suggested that the surface topography play a significant role in attachment of the bacteria. In the series of researches reported by Culfaz et al., they prepared a micro-structured hollow-fiber membrane for ultrafiltration using a patterned spinneret and observed their performance such as water flux or antifouling mechanism in filtration process [11–15]. As a result, higher water productivity of the patterned hollow fiber membrane was observed due to the enhanced membrane surface. Also, under specific conditions, the patterned hollow fiber membrane showed higher antifouling ability than a flat hollow fiber membrane. Through their researches, they verified that the potential of the patterned membrane could be a breakthrough in solving the membrane fouling problem. However, the types of patterns on the membrane surface were limited because they embedded the patterns by converting the spinneret. Won et al. reported the preparation of patterned membranes by a modified immersion precipitation method [16,17]. They prepared diverse patterned membranes such as pyramid-, prism-, and embossing-patterned membranes and showed that the anti-fouling ability of patterned membranes was improved in comparison to non-patterned membrane. More recently, Maruf et al. prepared a sub-micron patterned membrane via direct surface patterning with nanoimprint lithography [18,19]. Their membrane, which had feature dimensions of about 100 nm, experienced less silica and bovine

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http://dx.doi.org/10.1016/j.memsci.2015.09.058
0376-7388/

serum albumin (BSA) deposition when feed was injected at a specific angle to the pattern direction. They attributed this decrease in foulant deposition to localized turbulence induced by the feature.

Although the antifouling effects of patterns on the membrane surface have been demonstrated in previous works, the mechanism of antifouling ability in a patterned membrane has yet to be explored in detail [20]. The previous studies have paid little attention to (i) parameters affecting the antifouling ability of patterned membranes, and (ii) development of diverse patterns to enhance the antifouling ability in the membrane process.

In this work, therefore, membranes with various prism patterns were fabricated with the aid of a soft-lithographic method. Then, the particle depositions on these patterned membranes were compared with regard to several important parameters including Reynolds number, pattern size and interval between patterns. The effect of the parameters on the particle deposition was analyzed using computational fluid dynamics (CFD) modeling.

2. Experimental

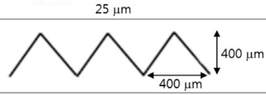
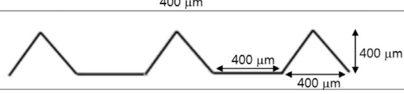
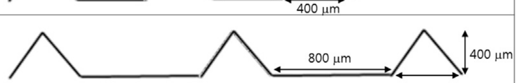
2.1. Types of patterns

Schematic diagrams of diverse prism patterns used in our experiments were described in Table 1. Patterns A and B were designed to identify the relationship between pattern size and antifouling ability of patterned membranes. Patterns C and D had the same pattern dimension as pattern B, except for the intervals between prism patterns. The interval length of the patterns C and D was 400 and 800 μm, respectively. A non-patterned membrane was used as a control membrane in all experiments.

2.2. Preparation of patterned membranes

Polyvinylidene fluoride (PVDF), Dimethyl-formamide (DMF) and acetone were purchased from Sigma Aldrich Korea. Deionized (DI) water used in the experiments was purified by a Milli-Q system (Millipore, USA). All of the chemicals were used without further purification. The preparation steps of a patterned membrane were described in detail in our previous work [17]. In order to prepare a PVDF-casting solution, commercial PVDF pellets were dried under vacuum at 100 °C for 12 h to remove any moisture before the preparation of polymer solution. A polymer solution was prepared by dissolving 0.4 g of PVDF pellets in 2.27 g of DMF at 60 °C for 12 h and then kept at a roller overnight to ensure complete mixing. Before casting a polymer solution, three sets of water bath sonication were performed in series for 10 min each time to degas it. A PVDF polymer solution was cast onto a prepared replica-mold with a casting knife and then immersed into a water bath for 2 h. Immediately after the casting, fabric used as supporting layer was placed on top of the solution. The nascent membrane, together with the fabric and replica mold, was dipped into the precipitation bath and coagulated for 6–10 h. The patterned membrane with the fabric was then released from the replica mold and stored in a bath filled with DI water. The mean pore sizes and thickness of prepared patterned membranes measured by SEM observation were about 0.8–1.2 μm and 190–200 μm, respectively, which were almost identical, irrespective of the pattern types.

Table 1
Cross-sections and dimensions of prism patterns on membrane surface.

Pattern ID	Geometry of pattern
Pattern A	
Pattern B	
Pattern C	
Pattern D	

2.3. Operation of crossflow filtration with latex particles

Fig. 1 shows the schematic diagram of the experimental setup used in the filtration tests. The membranes were fixed in two identical membrane modules. For the particle deposition tests, a latex particle suspension in DI water was prepared at a concentration of 4–7 wt%. Latex particles were purchased from Sigma Aldrich Korea, the diameters of which was 2 μm. They had red fluorescence and, thus, after crossflow operation, could be observed by confocal laser scanning microscopy (CLSM) [21].

Feed solution containing latex particles in a tank was transferred to two separate membrane modules by two separate pumps. The patterned membrane in a module was directly compared to a non-patterned membrane in another module. The pressure applied to the membrane module was maintained at 20 kPa, and filtrate was collected in small jars on electronic balances. Crossflow velocity was fixed at 0.1 and 0.4 m/s, which corresponds to $Re = 600$ and 1600, respectively. Water flux was estimated by calculating the flow rate of filtrate per unit membrane area from the data of the balances. For the calculation, the projection area of the patterned membrane surface was adopted as the membrane surface area, which was the same as that of non-patterned membranes. The filtrate was returned to the feed tank after its mass was measured on the electronic balances for calculating water flux. After 2 h operation, each membrane with deposited latex particles was released from the membrane modules and moved into a small bottle. After adding 20 mL of deionized water in the bottle, the deposited latex particles on the membrane surface were detached from the membrane surface by sonication, and turbidity was measured three times to calculate the concentration of latex particles from a concentration–turbidity calibration curve prepared in advance. The crossflow filtration test was repeated with another virgin membrane under the same conditions to observe the membrane surface with CLSM. Membrane specimens with 1.5 cm × 1.5 cm were taken from the membrane after crossflow filtration and stained with Concanavalin A (green fluorescent, Molecular Probes Inc., Eugene, OR, USA). We observed the membrane surface using a confocal laser scanning microscope (C1 plus, Nikon, Japan).

2.4. CFD modeling

Flow behavior near the patterned membrane was analyzed by using commercial software (Multiphysics 3.5, COMSOL Inc., USA) [20]. Patterned geometry was generated for each type of prism pattern described in Table 1. Navier–Stokes and fluid continuity equations were solved by the finite element method (FEM) in 2D, neglecting possible variances in flow in the z-axis (channel width direction). The numbers of computational meshes were 6000–6500 for all cases of patterned geometry. The fluid was assumed to be Newtonian with the physical properties of water at 20 °C. Non-