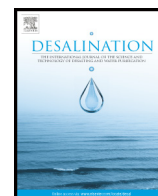




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Influence of colloidal fouling on pressure retarded osmosis

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HIGHLIGHTS

- Colloidal fouling behavior in pressure retarded osmosis (PRO) was systematically investigated.
- Colloidal fouling is dominantly influenced by the cake layer formation and the deposition of foulant within the support layer.
- Severe flux decline was observed with smaller colloidal particles.
- The feed solution pH has potential to mitigate the colloidal fouling.

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ABSTRACT

In this study, colloidal fouling behavior in pressure retarded osmosis (PRO) was systematically investigated in terms of the effects of draw solution concentration, applied hydraulic pressure at the draw side, feed solution pH, and particle size. Commercially-available cellulose triacetate (CTA) membranes were fouled with feed solution containing silica colloidal particles. Two different silica particles with mean diameter of 27 and 152 nm were used as model foulants. Our findings demonstrated that the colloidal fouling in PRO was dominantly affected by the cake layer buildup at the membrane surface. Fouling was further exacerbated by diffused salts from the draw side because retained salts within the cake layer elevated the salt concentration on the membrane surface, and consequently reduced the driving force of PRO. Substantial flux decline with the smaller particles was attributed to the high cake layer resistance due to the formation of the void-less cake layer. In addition, our approaches to mitigate the colloidal fouling revealed that the hydraulic cleaning by increasing the cross-flow rates was not effective to eliminate the compact cake layer. However, adjusting the feed solution pH showed the high potential to relieve the colloidal fouling resulting from the more stabilization of particles at low solution pH.

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1. Introduction

Heavy dependence on fossil fuels which deteriorates the climate change places an increasing demand for alternative energy resources. To secure the sustainable energy supply and simultaneously reduce the carbon emissions, researches to explore the substitutes of current power source have been actively conducted. As a result, now over 20% of the global energy supply is shared by the renewable energies such as solar, biomass, geothermal, wind, and wave energies [1]. Recently, a technology using salinity gradient energy (SGE) is considered as a prospective alternative of traditional energy-production technologies. As harnessing the free energy released from mixing of two solutions

having different salinities, SGE has benefits of less environmental footprint and less periodicity to weather variations. Extractable energy between a cubic meter of fresh river water and seawater was approximately 0.61 kWh in a thermodynamic point of view [2]. Furthermore, considering the freshwater and seawater, the potential of SGE is assessed to be a total of ~2.6 TW, equivalent to the global energy demands [3].

Pressure retarded osmosis (PRO) as one of the SGE processes, has recently drawn strong attention, whose impetus is the chemical potential difference between a diluted feed solution and a concentrated draw solution. Such salinity gradient induces the water permeation from the feed to the draw side, and then the hydraulic pressure lower than osmotic pressure difference is applied at the draw side to retard the permeate water flux. The pressurized volumetric water flow runs a hydro-turbine to convert the mechanical energy to the electric energy. Due to the remarkable advances in membrane technologies, PRO has

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been regarded as a prospective viable energy option, followed by the invigorated research on PRO from lab-scale to pilot-scale demonstration. The world's first PRO pilot plant was opened in Norway in 2009, and since then a reverse osmosis (RO)-PRO hybrid plant was constructed in Japan in 2011, and a RO-membrane distillation (MD)-PRO hybrid pilot plant is under construction in Korea [4–6]. However, several challenges still remain to make the PRO technology economically viable for commercialization.

Membrane fouling can be a critical factor to restrict the PRO performance, particularly when it comes to a commercial-scale plant with a long-term operation. Fouling in pressure-driven processes detrimentally affects the permeate water quality, and operation and maintenance costs [7,8]. Although the fouling layer in PRO is estimated to be less compacted than that in RO due to the relatively low hydraulic pressure, the influence of membrane orientation should be carefully considered. Foulants present in the feed solution are expected to be easily deposited within the support layer due to the favorable membrane orientation in PRO (i.e., active layer facing the draw solution and support layer facing the feed solution, hereafter AL-DS mode), which results in substantial flux reduction.

Several researches have been investigated to clarify the PRO fouling mechanisms [9–14]. For example, organic fouling behavior in PRO was examined under various experimental conditions such as hydraulic pressure, draw solutions, foulant type, and pH [10,11]. Furthermore, the flux decline in PRO due to the natural organic matter (NOM) dissolved in the feed water was studied using cellulose triacetate (CTA) and thin film composite (TFC) membranes [12] and hand-cast TFC membranes [9]. With regard to inorganic based fouling, the gypsum (i.e., calcium sulfate dehydrate) scaling was examined at various draw solution types and concentrations, the hydraulic pressure, and membrane orientation [13]. A recent study compared the impact of organic, inorganic (inorganic ionic species), with combined fouling on PRO performance [14]. Although numerous studies have been performed to elucidate the PRO fouling mechanism, most of them were limited to the organic fouling and inorganic scaling. As the fouling behavior can be varied with respect to the membrane materials and foulant type, membrane fouling caused by the particulate matter or microorganisms needs to be carefully scrutinized.

Colloidal fouling in PRO due to the ubiquitous existence of colloidal particles ranged between 1 and 1000 nm in wastewater effluent, causes severe membrane [15]. In the membrane processes, the deposition of particles on the membrane surface or within the membrane structure is regarded as the major contributor of colloidal fouling, and the extent of fouling is largely dependent on solution chemistry which is solution pH, ionic strength and existence of specific ions like divalent ions (Ca^{2+} , Mg^{2+}) and tri- or multi-valent ions [16]. Although no previous works have so far been reported to clarify the mechanisms of colloidal fouling in PRO, significant efforts have been made to investigate the colloidal fouling mechanisms in pressure-driven membrane processes such as RO, nanofiltration (NF), ultrafiltration (UF) and microfiltration (MF) and osmosis-driven processes like forward osmosis (FO) [16–19]. Fouling in high pressured membranes such as RO and NF is caused by the cake layer formation on the membrane surface while pore plugging and pore adsorption critically affect the membrane fouling in low pressure membranes i.e., UF and MF [16–18]. Colloidal fouling phenomena have been further studied by forward osmosis (FO), which is an osmotically-driven process, demonstrating that flux decline mainly resulted from the accelerated cake-enhanced osmotic pressure (CEOP) [19]. Membrane properties and operating conditions in PRO are not identical to the previously studied membrane processes and therefore systematic investigation of colloidal fouling in PRO is highly desirable.

As such, this study aims to investigate a comprehensive understanding of PRO fouling behaviors specifically caused by colloidal particles and to provide the future insight into fouling mitigation. Accordingly, the influence of colloidal particles on PRO performances was evaluated

in terms of draw solution concentrations, applied hydraulic pressures, and the feed solution pH. In particular, the colloidal fouling mechanisms in PRO are elucidated with regard to the cake layer resistance of two different particle sizes. Fouling propensity and reversibility are also conducted using the typical membrane cleaning protocols after the fouling experiments.

2. Materials and methods

2.1. Membrane

A commercially available flat-sheet membrane from Hydration Technology Innovations (Albany, OR, USA) was used in this study. The membrane consists of the cellulose triacetate (CTA) active layer and the woven embedded polymer support layer. The membrane samples were stored at 4 °C and soaked in deionized (DI) water for over 24 h before use. A new membrane sample was utilized for each fouling experiment and placed in a PRO membrane cell with an AL-DS mode.

2.2. Colloidal foulants

Commercially available silica (SiO_2) particles, Snowtex 30 (ST-30) and Snowtex ZL (ST-ZL) provided by Nissan Chemical Industries (Tokyo, Japan), were used as model foulants. The concentration of the silica particles was 30 wt% for ST-30 and 40 wt% for ST-ZL. The density of the ST-30 and ST-ZL is 2.1 g/cm³ and 2.3 g/cm³, respectively [19]. According to the manufacturer, the diameter of the ST-30 and ST-ZL is ranged from 10 to 15 nm and from 70 to 100 nm, respectively. However, the mean diameter based on our measurement via Zetasizer nano series (Malvern Instruments, UK) was found to be 27 ± 1.20 nm and 152 ± 2.81 nm for the ST-30 and ST-ZL, respectively. Prior to use, the silica suspensions were sonicated for 10 min to ensure uniform dispersion of the colloidal particles. The electrophoretic mobility of the silica particle suspensions was evaluated by ELS-Z (Photal Otsuka Electronics, Japan) as a function of solution pH in the presence of 10 mM NaCl as a background electrolyte. For all fouling experiments, the silica concentration in the feed solution was fixed at 1 g/L.

2.3. Lab-scale PRO system

A lab-scale PRO system used in this study consists of two reservoirs for feed and draw solutions, a custom-made stainless membrane module, a high-pressure pump to pressurize and circulate the draw solution, a variable speed gear pump (Cole-Parmer, Vernon Hills, IL) to circulate the feed solution, a digital scale to measure the weight change of the feed solution, two digital-type pressure sensors at the membrane module outlet, two analogue-type pressure sensors at the membrane module inlet, two flow meters, and a temperature control system (Fig. 1). It is worthwhile to note that the hydraulic pressure applied at the draw side was automatically controlled via a proportional integral derivative control, and thus kept identical during the operation. The dimensions of the feed and draw solution channels were 75 mm long, 26 mm wide, and 2 mm high with an effective membrane area of 19.5 cm².

2.4. Fouling experiments

Fouling experiments were conducted for 10 h with different concentrations of draw solution (1 M NaCl and 2 M NaCl), applied hydraulic pressures (5 bar and 10 bar), silica particle combination (mono-disperse and poly-disperse), and feed solution pH (pH 3, pH 7 and pH 9).

The cross-flow velocities of both feed and draw channels were fixed at 9.6 cm/s under a counter-current flow operation. Total ionic strength of feed solution was kept identical at 10 mM NaCl and temperature was maintained at 20.0 ± 0.5 °C. To withstand the applied hydraulic pressure and consequently prevent the membrane damage, eight layers

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