Desalination 349 (2014) 51–59

Contents lists available at ScienceDirect

Desalination

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

Effects of phosphate limitation in feed water on biofouling in forward osmosis (FO) process

Chang-Min Kim [a], Sung-Jo Kim [a], Lan Hee Kim [a], Myung Seop Shin [a], Hye-Weon Yu [b], In S. Kim [a,*]

[a] *School of Environmental Science and Engineering, Gwangju Institute of Science and Technology (GIST), 261 Cheomdan-gwagiro, Buk-gu, Gwangju 500-712, Republic of Korea*
[b] *Department of Soil, Water and Environmental Science, College of Agriculture and Life Sciences, University of Arizona, Tucson, AZ, United States*

HIGHLIGHTS

- Effects of phosphate limitation on biofouling in FO process were confirmed.
- Characteristics of biofouling in FO process were investigated.
- Role of accelerated BEOP and Gibbs–Donnan equilibrium in phosphate limitation was explored.
- Applicability of phosphate limitation as pretreatment in FO process was shown.

ARTICLE INFO

Article history:
Received 9 April 2014
Received in revised form 9 June 2014
Accepted 10 June 2014
Available online 9 July 2014

Keywords:
Forward osmosis (FO)
Biofouling
Phosphate limitation
Accelerated biofilm-enhanced osmotic pressure (Accelerated BEOP)
Wastewater treatment

ABSTRACT

Forward osmosis (FO) has been applied to various water treatment processes because of its low energy consumption and lower fouling propensity. Fouling, however, especially biofouling, is still problematic in FO. To resolve the biofouling issue, phosphate limitation was investigated to reduce biofouling instead of commonly used chemical cleaning. To establish the effects of phosphate limitation on biofouling formation, various phosphate concentrations and nutritional conditions were tested to seek a basic correlation between phosphate limitation and biofilm formation, and effects of phosphate limitation on performance of FO were checked. As a result, it was discovered that the phosphate limitation in feed water greatly reduced microbial growth and biofilm formation, thereby FO performance was maintained. This phenomenon was explained in accelerated biofilm-enhanced osmotic pressure (Accelerated BEOP) and Gibbs–Donnan equilibrium mechanism, and it shows that phosphate limitation can effectively control biofouling by alleviating the accelerated BEOP and balancing the Gibbs–Donnan equilibrium. Therefore, it may be feasible to use the phosphate limitation in FO pretreatment to control biofouling.

1. Introduction

The depletion of fresh water sources due to climate change has led to the development of water reuse and purification systems as alternative water sources. In particular, membrane-based seawater desalination and wastewater treatment are attracting increasing attention from researchers because of their high cost-effectiveness and excellent water quality in comparison to that produced by conventional water treatment technologies. Among these alternative membrane processes, in recent decades forward osmosis (FO) technology has displayed great potential in a wide variety of applications. This versatility has been attributed to its primary advantage, i.e., FO employs only a differential chemical potential between feed and draw solutions, without requiring pressurization. Hence, FO may consume much less energy and causes less irreversible fouling than pressurized processes such as reverse osmosis (RO) and nanofiltration (NF) [1–4].

The use of FO in various water treatment processes has also been explored in recent years. Hybrid processes that combine FO with other membrane-based or water treatment processes are now seeing applications in the areas of seawater desalination and wastewater reuse [5–8]. With respect to wastewater treatment, osmotic membrane bioreactors (OMBRs), which incorporate the respective advantages of both biological treatment and membrane filtration, have been attracting increasing attention due to their low fouling propensity, which is considered a major advantage of FO [9–13].

Although FO has a significantly lower fouling tendency and higher recovery rate compared to other pressurized membrane processes, fouling problems remain the most important and inevitable issues in the practical applications of FO. In particular, among many types of fouling, biofouling is a main concern in wastewater treatment because biofouling is not easily removed due to continuous provision of microorganisms and substrates from wastewater. Therefore, it is necessary to

---

* Corresponding author. Tel.: +82 62 715 2436; fax: +82 62 715 2434.
E-mail address: iskim@gist.ac.kr (I.S. Kim).

http://dx.doi.org/10.1016/j.desal.2014.06.013
0011-9164/© 2014 Elsevier B.V. All rights reserved.

reduce biofouling potential by developing effective biofouling control or reduction technology to effectively apply the FO process to wastewater treatment and reuse.

Conventional physical or chemical cleaning methods have shown great efficiency in short-term biofouling reduction; however, these methods incur many problems over the long term, including high energy requirements, shortening of the membrane lifetime, and the toxicity of chemical agents used to the environment and the human body [14,15]. Moreover, once a biofilm is formed on a membrane surface, the physical and chemical cleaning efficiency gradually decreases so that biofouling becomes increasingly difficult to eliminate by chemical cleaning, due to both the biofilm organisms protecting themselves by producing more microbial products and their significant and rapidly increasing tolerance to biocides [16–18]. Hence, understanding biofouling characteristics in the FO process and developing novel, environmental-friendly, and highly efficient approaches are important for the continued success of the FO process.

However, few works have reported on biofouling in the FO process, and no published works were found pertaining to biofouling reduction technology in FO. Thus, the study of biofouling mechanisms and reduction technology in FO has become the first priority. As more environment-friendly and effective biofouling reduction technologies develop, limiting nutritional compositions (carbon (C), nitrogen (N), and phosphate (P)) in the feed solution has been introduced in attempts to prevent or restrict microbial growth, instead of killing microorganisms using chemical biocide [19–21]. This approach can reduce the problems of conventional cleaning methods mentioned above and remove side effects such as the production of better bioavailable organic carbon through reactions between biocide and recalcitrant organic matter [22]. This limitation of nutrients in the feed solution has great potential for biofouling reduction. In particular, among available nutrients, phosphorous or phosphate is a potentially significant limiting factor as the molar ratio of carbon, nitrogen, and phosphorous is ~100:20:1.7 in a microbial biomass [23]; indicating a much lower concentration of phosphate than carbon is required for cell growth [24–26]. However, even low concentrations of phosphorous can more significantly affect the microbial status than other major nutrients (carbon and nitrogen). In this study, therefore, phosphate was identified as a vital nutritional factor for establishing the relationship between phosphate and biofouling in the FO process.

The objectives of the study included the establishment of biofouling characteristics in the FO process, and determining whether the limitation of phosphate in the feed water reduces the biofouling potential. To determine the influence of phosphate concentration and its limitation on microbial growth, batch tests were conducted, and the impact on the reduction of biofouling in a lab-scale FO system was examined for two kinds of solution: a synthetic solution, and a secondary wastewater effluent. Finally, FO biofouling characteristics were found based on a mechanistic understanding. In addition, the applicability and efficiency of phosphate limitations for biofouling reduction in an FO system were confirmed.

2. Materials and methods

Our experiments can be divided into two parts and differentiated by feed water solution or experimental hydraulic conditions. In order to prove the effects of phosphate limitation, a batch test and an FO test were conducted with synthetic wastewater as feed water. Furthermore, secondary wastewater effluent was used as feed water in a lab-scale FO system to evaluate the applicability of phosphate limitation to real wastewater treatment.

2.1. Experimental conditions

2.1.1. FO membrane

The FO membrane used in this study was provided by Hydration Technologies, Inc. (Albany, USA). It has an asymmetric structure comprised of cellulose triacetate cast onto a non-woven backing consisting of polyester fibers individually coated with polyethylene. To remove glycerin on the membrane, it was soaked in deionized water for more than 30 min. The detailed characteristics of this membrane can be found in a published report [27].

2.1.2. Model bacteria preparation

Pseudomonas aeruginosa PAO1 was employed as the model bacterium because it is widely considered to be a dominant bacteria on biofouled membranes [28–30]. Our *P. aeruginosa* PAO1 strain was kindly received from Seoul National University, with permission from the Center for Biofilm Engineering (Montana State University, USA).

P. aeruginosa PAO1 was transferred and cultured in a nutrient broth (Becton Dickinson, Difco, USA) for 15 h at 36 ± 1 °C. Cultivated bacteria were harvested by centrifugation for 10 min at 8000 rpm at 4 °C. The precipitated pellet was washed three times with 0.9% saline to remove nutrients. The concentration or optical density of the cultivated cells re-suspended was then calculated using a UV-visible spectrophotometer (Ultraspec 3100 pro, GE Healthcare Bio-Sciences Corp, Little Chalfont, UK) at 600 nm (OD_{600}), and the re-suspended solution was inoculated into the feed solution to achieve a cell concentration of 10^7 cells/mL in each experiment.

2.1.3. Synthetic wastewater

In order to verify the effects of phosphate limitation on biofilm formation, synthetic wastewater was used. The composition of the synthetic wastewater was previously described by Herzberg et al. [31], and constituents of the synthetic inorganic medium used in this study are also presented in Table A.1. In brief, glucose ($C_6H_{12}O_6$), ammonium chloride (NH_4Cl), and mono-potassium phosphate (KH_2PO_4) were selected as the carbon (C), nitrogen (N), and phosphorus (P) sources, respectively. Specifically, ortho-phosphates were employed as representative phosphorous substances because most microorganisms uptake and utilize ortho-phosphate (H_3PO_4, $H_2PO_4^-$, HPO_4^{2-}) [32]. In addition, the C, N, and P sources were separately restricted or injected depending on the experimental conditions.

2.1.4. Secondary wastewater

The secondary wastewater effluent used in this study was taken from the Gwangju Wastewater Treatment Plant (35° 9' 18.77'' N, 126° 49' 50.75'' E). To confirm the effects of phosphate limitation on biofouling, the secondary wastewater effluent source was prepared in two ways: unaltered secondary wastewater effluent, and phosphate-limited secondary wastewater effluent (removing only phosphate). Removal of phosphate has been achieved and reported in a variety of materials which have cost effectiveness and high removal efficiency [33–40]. In this study, low cost lime water, $Ca(OH)_2$ (aq) was added into the secondary wastewater effluent to remove phosphate as forming hydroxyapatite, which has a chemical formula $Ca_5(OH)(PO_4)_3$ (s) [41]. Hydroxyapatite can be formed in alkaline conditions, enabling a very low concentration of phosphate to be obtained at a pH higher than 10.5.

In this study, the weight of the required calcium hydroxide was calculated from Eqs. (1) and (2). The phosphate concentration in wastewater was measured using a water quality analysis kit (HUMAS, South Korea) after 5 times dilution with deionized water, and was found to be 4.92 mg/L. Thus, 4.92 mL of lime water solution (3.98 g $Ca(OH)_2$/L), which can precipitate 4.92 mg/L of phosphate, was injected into 1 L of wastewater. The phosphate formed precipitates including $CaCO_3$(s) (calcite) and $Ca_5(OH)(PO_4)_3$ (s) (hydroxyapatite).

$$5Ca^{2+} + 4OH^- + 3HPO_4^{3-} = Ca_5(OH)(PO_4)_3 + 3H^+ \tag{1}$$