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Combination of lauroyl arginate ethyl and nisin for biofouling control in reverse osmosis processes

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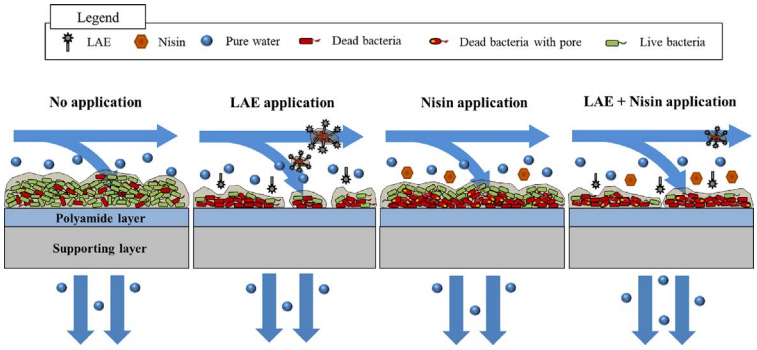
GRAPHICAL ABSTRACT



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ABSTRACT

Lauroyl arginate ethyl (LAE) is a non-oxidizing biocide effective in controlling biofouling in reverse osmosis (RO) processes. However, the operating concentrations of LAE should be lowered to reduce the possible adsorption of LAE onto RO membranes and to reduce operational costs. This study investigated combinations of LAE with another non-oxidizing biocide, nisin, to overcome such practical limitation of LAE in a bench-scale RO unit operated with full recirculation with the model bacteria *Staphylococcus aureus* ATCC6538 and *Pseudomonas aeruginosa* PA14. Several combinations of LAE and nisin (e.g. 1 mg/L LAE + 2 mg/L nisin) were more effective in killing bacteria than LAE or nisin treatment alone, possibly due to permeability synergy. Furthermore, in a bench-scale RO unit operated with full recirculation and model bacteria, flux declines were more effectively moderated by dosing LAE and nisin together compared with the dosing of LAE or nisin alone. LAE dosing alone showed rapid flux decline in the early operational period, while nisin dosing alone demonstrated the lowest flux at the end of the operation. In conclusion, combinations of LAE and nisin appear more effective to mitigate biofouling developed on membranes than LAE or nisin alone.

1. Introduction

Desalination of seawater or brackish water using RO processes has been regarded as a promising water treatment technology for coping with drinking water shortage [1,2]. However, similar to other membrane-based technologies, RO processes are prone to suffer from the membrane fouling phenomenon. Compared with fouling associated with organic, inorganic, particulate, and colloidal matters, biofouling is a major operational problem in the RO processes [3]. Biofouling is a result of the unwanted growth of microorganisms in the form of biofilm on the membrane surface [4], and negatively affects permeate flux, feed pressure, energy efficiency, and salt/boron rejection [5]. Feed water

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pretreatment, hydraulic flushing, chemical treatment and conditioning, and chemical cleaning of the membrane are typically used to mitigate problems associated with biofouling [6]. In addition to the operational practices, recent studies demonstrated that membranes can be fabricated to be more resistant to biofouling [7–10].

Chlorine-based chemicals (e.g. chlorine, chloramine, and chlorine dioxide) are frequently used to reduce biofouling in the RO processes [11]. However, they should be inactivated by the reducing agents (e.g. bisulfite) before the RO membrane vessel to avoid damage to the skin layer of the RO membrane [12]. The inactivation of chlorine-based chemicals results in the regrowth of microorganisms in the membrane module and eventually causes biofouling [13]. To overcome this unwanted condition, non-oxidizing biocides having compatibility with RO membranes can be directly dosed in the membrane vessel [14,15]. Dibromo-nitrilopropionamide (DBNPA), tributyl tetradecyl phosphonium chloride, and isothiazolone are typical non-oxidizing biocides applicable in RO processes for the purpose of dosing in the membrane vessel [14,16]. Nevertheless, these biocides can only be used in RO processes that produce non-potable water, due to their potential toxicity to humans [17].

Our recent study [18] demonstrated that a food preservative named lauroyl arginate ethyl (LAE) had compatibility with the polyamide RO membrane and strong antimicrobial and antibiofilm properties. Moreover, the study showed that RO performance such as water flux and salt rejection did not deteriorate in response to LAE dosing. Above all, LAE can be used to produce potable water, because LAE can be completely metabolized into carbon dioxide and water by mammalian bodies [19,20]. However, cationic arginine moiety of LAE can potentially interact with the anionic skin layer of the RO membrane and form hemimicelles [18], which can negatively affect the RO performance over a long period of operation [18]. In addition, the relatively high cost of LAE compared to conventional non-oxidizing biocides that are used in RO processes can limit its applications.

In order to overcome the two problems outlined above, the dosing concentrations of LAE should be lowered. However, low concentrations can reduce the antimicrobial and antibiofilm effects of LAE. LAE thus needs to be mixed with another inexpensive food preservative to achieve an equivalent biocidal effect. Nisin, pimaricin, dimethyldicarbonate, and propionic acid are low-cost food preservatives that are potentially applicable for the purpose. Nisin, a natural antimicrobial peptide that is derived from *Lactococcus lactis* [21], was selected for this study given its antibacterial property, water solubility, and stability in water. In addition, the nisin is different from LAE in antimicrobial mechanism, which can exert a synergistic biocidal effect between the two biocides [22]. Nevertheless, whether or not the combination is effective in reducing biofouling in RO processes has not been reported.

The primary aim of this study was to evaluate the effects of the combination of the two biocides (LAE + nisin) in RO processes associated with biofouling. Initially, the compatibility of the two biocides with an RO membrane was evaluated and the optimal combinations of the two biocides for synergistic effects were determined. A biocidal mechanism of a combination of the two biocides was then proposed based on electron microscopic images of bacteria treated with the biocides. Finally, the flux decline and biofilm formation on the RO membrane were analyzed by a combination of the two biocides.

2. Materials and methods

2.1. Bacteria, biocides, and RO membrane

Staphylococcus aureus ATCC6538 and *Pseudomonas aeruginosa* PA14 were used as model gram-positive and gram-negative bacteria for antimicrobial and antibiofilm tests. Both species were cultured in a shaking incubator at 250 rpm and 37 °C using a tryptic soy broth (TSB) (BD). LAE (CDI, Hwasung, South Korea) was dissolved in deionized (DI) water, while nisin (Sigma-Aldrich) was dissolved in 0.02 N HCl (pH = 2). Both solutions were filtrated using a 0.22 μm syringe filter and adjusted to pH 7 using 1 M NaOH or 1 M HNO_3. Filmtec SW30HR-380 RO membrane (Dow Chemical) was used to evaluate compatibility with the two biocides and biofouling tests.

2.2. Compatibility of biocide with RO membrane

Coupons of the RO membrane (1.5 cm diameter) were prepared and washed to remove residuals using DI water. To test the possible deterioration by the biocides, the coupons were immersed in 10 mL of 1000, 10,000, and 100,000 mg/L LAE or nisin solution, and incubated in a shaker at 100 rpm and room temperature for 1 h. Residuals on the coupons were removed five times using 500 mL DI water in a shaker at 140 rpm. The coupons were dried in a desiccator at room temperature for several days before evaluation of the physical and chemical damage using atomic force microscopy (AFM) and attenuated total reflection-Fourier transform infrared (ATR-FTIR) analysis.

2.3. AFM and ATR-FTIR analyses

AFM analysis was used for a quantitative analysis of the morphological damages on the membrane surface by LAE or nisin. The membrane surface was scanned at 5 random positions using the PUCO Station AFM (Surface Imaging Systems) at operation conditions of 10 μm × 10 μm scan area, 4.0–6.0 N/m approaching force, and 0.7 line/s scanning speed. The root mean square (RMS) and average surface roughness (S_a) were estimated using the scanned images obtained by the SPIP software (Surface Image Systems). FT-IR 4100 ATR-FTIR (Jasco) was used to evaluate the potential oxidative damages of the chemical bonds of the membrane. A minimum of 100 scans per sample were analyzed at a resolution of 1.0 cm^{-1} wave number in the range from 1300 cm^{-1} to 1900 cm^{-1}.

2.4. Antimicrobial and antibiofilm tests

For the antimicrobial test, the minimum inhibitory concentration (MIC) was measured using a broth micro-dilution method [14,23]. First, 100 μL TSB was added to each well of a 96-well plate (Sigma-Aldrich). Biocides were added to the wells in a concentration range from 62.5 to 0.5 mg/L using two-fold serial dilutions. 100 μL of bacterial culture (~10^6 colony forming unit/mL) was then added to the wells. The 96-well plate was incubated at 37 °C for 24 h. MIC was determined as the lowest biocide concentration for inhibition of bacterial growth which was measured by optical density (OD) at 595 nm using a microplate reader. An antibiofilm test was performed based on a static biofilm formation assay [14]. Similar to the antimicrobial test, TSB, biocide, and bacterial culture were added to the wells of a 96-well plate. The 96-well plate was incubated for 24 h at 37 °C. The planktonic cells in the wells were discarded using an eight-channel pipette. Afterward, the wells were washed twice using phosphate buffered saline (PBS) (pH = 7.2) for the removal of residual planktonic cells and TSB. The biofilm in the wells was stained with 0.1% crystal violet solution for 30 min. The unbound dye was washed twice with DI water. The bound dye in the biofilm was eluted using 99.9% ethyl alcohol. The biofilm was quantified by measuring OD at 545 nm using a microplate reader.

2.5. Evaluation of synergistic effects between LAE and nisin

The synergistic effects between LAE and nisin were evaluated for various ratios of the two biocides based on a previous study [24]. Initially, 100 μL TSB were added to each well of a 96-well plate. LAE (100 μL) was distributed to each well by two-fold serial dilutions from the left to right of the 96-well plate to adjust the LAE concentrations from 62.5 to 0 mg/L, while nisin (100 μL) was distributed in each well by two-fold serial dilutions from the top to bottom of the 96-well plate to adjust the nisin concentrations from 62.5 to 0 mg/L. The bacterial