



Surface modification of a microfiltration membrane for enhanced anti-biofouling capability in wastewater treatment process

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ABSTRACT

Membrane bioreactors (MBR) are now widely implemented in wastewater treatment plants due to their compact design and high effluent water quality. However, membrane biofouling has significant impact on MBR performance and energy cost. Quorum sensing inhibiting (QSI) compounds have the ability to disrupt bacterial communication pathways that lead to biofilm formation and biofouling. In this study, an anti-biofouling membrane was developed by incorporating vanillin, a known QSI molecule onto the surface of a commercial chlorinated polyethylene (CPE) microfiltration membrane by Williamson ether synthesis reaction. The CPE membrane was reacted with the vanillin in a solvent mixture containing potassium carbonate and N, N-dimethylformamide for vanillin incorporation. The solvent mixture without vanillin was also used to create a control membrane to understand the QSI anti-biofouling capabilities. Raman and FTIR spectroscopy indicated that vanillin was retained on the modified membrane surface under cross-flow condition and sodium hypochlorite cleaning. The modified membrane had significantly less biofouling compared to the unmodified and control membranes tested using activated sludge collected from wastewater treatment plants. The control membrane experienced up to a 52% reduction in live cells, dead cells, and polysaccharides in the fouling layer compared to unmodified membrane. The incorporation of vanillin further decreased the amount of live cells, dead cells, and polysaccharides covering the surface by 82%, 77%, and 51%, respectively, compared to the unmodified membrane. This research showed that vanillin-modified membranes have potential to improve the membrane anti-biofouling capabilities and MBR performance.

1. Introduction

Membrane bioreactors (MBRs) are now widely used in municipal and industrial wastewater treatment plants. MBR technology integrates two processes, activated sludge and low-pressure membrane filtration, into a single system that degrades organic matter and clarifies the effluent [1]. A microfiltration or ultrafiltration membrane is typically used in the low-pressure membrane filtration. MBR removes the need of a secondary sedimentation basin, which greatly decreases the space needed in a densely populated urban area. MBR technology offers a stable effluent water quality, shorter sludge retention times, and a potential 30–50% smaller energy footprint than the conventional process [2,3].

However, MBR systems are subjected to membrane fouling from particulate deposition, bacteria accumulation, and biofilm formation on the membrane surface. Microbial community composition and cell

density in activated sludge also influences membrane fouling [4]. Fouling reduces permeate flux and increases trans-membrane pressure, which leads to an elevated energy demand and operational costs for the unit [5]. Currently, MBR fouling is mitigated using physical and chemical cleaning methods to remove particulate matter and bacterial biofilm on the membrane surface [5]. Yet, even with repeated cleanings, membrane biofouling is still inevitable due to the dense mixture of bacteria and suspended particles in activated sludge. Therefore, it is imperative to find alternative methods to mitigate MBR biofouling.

Past research found that bacteria in wastewater MBRs rely on cell-to-cell signaling systems known as quorum sensing (QS) to coordinate community behavior leading to biofilm formation [6–10]. The QS pathway is a concentration dependent system where production of sensing molecules depends on the amount of bacteria present in the local environment. Blocking these bacterial communication pathways may offer a promising approach to control membrane biofouling

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without exerting external forces that often lead to bacterial resistance. Several natural or synthetic compounds have been found to impede QS functions and reduce bacterial biofilm formation [11,12].

Vanillin (VA), a natural favoring agent, was shown to inhibit a broad range of gram-negative bacterial QS functions relevant to MBR biofouling [13]. VA is known to modify the signaling molecule in the cell culture to prevent it from binding to the appropriate transcription regulator so that biofilm formation is not activated [14,15]. Past studies showed that VA dosed in the bulk fluid reduced biofilm production and MBR biofouling [11,13]. However, continuously dosing vanillin in the bulk fluid is not economical because a constant high dose is required to effectively block bacterial communication pathways. Modification of membrane surface with VA could be a promising approach to combat biofilm formation at membrane surfaces instead of in the bulk fluid. We recently reported that adsorption of VA onto the surface of polyamide reverse osmosis (RO) membranes [16] can enhance the membrane's anti-biofouling property [17,18]. The additional VA layer also did not impact membrane performance in respect to pure water permeate flux and salt rejection. In a high pressure RO system subjected to high density of marine bacteria and nutrients, biofilm production was suppressed on VA modified membranes and the water flux was improved compared to the unmodified membrane [16]. However, VA coated by physical adsorption was not stable on the membrane surface. VA was washed off after subjecting to cross-flow for longer than 48 h.

The goal of this study was to develop an in-situ anti-biofouling membrane by chemically linking VA to the surface of a microfiltration membrane to improve VA's retention. A Williamson ether synthesis reaction was used to modify the membrane surface to maintain VA's QS inhibition (QSI) activity. Raman and Fourier transform infrared (FTIR) spectroscopy were used to verify VA attachment and retention on the modified membrane surface. The modified membrane performance was evaluated in a membrane biofouling monitoring system and compared with control and unmodified membranes. Confocal scanning laser microscopy (CSLM) was used to examine biofilm formation on the fouled membrane samples. Biofilm coverage on membrane was quantified using COMSTAT 2 software for comparisons between samples tested.

2. Experimental

2.1. Membrane modification

A commercially available and commonly used chlorinated polyethylene (CPE) microfiltration membrane (Kubota Corporation, Osaka, Japan) was adopted for surface modification in this study due to its proven efficiency in MBR operation. The following chemicals from Sigma-Aldrich (St. Louis, USA) were used for membrane surface modification without further purification: Vanillin (VA, 4-hydroxy-3-methoxybenzaldehyde), potassium carbonate (PC), and N, N-dimethylformamide (DMF).

A Williamson ether synthesis reaction [19] was adapted to bond VA to the CPE membrane surface through the following anticipated steps (Fig. 1): 1) PC deprotonates VA to produce an alkoxide ion; 2) DMF slows down the reaction rate of the highly reactive alkoxide ion; 3) Alkoxide ion replaces the haloalkane bond on CPE to form an ether bond between VA and CPE [20]. In preparation for this reaction, the CPE membrane was first cleaned with 80% ethanol and then rinsed with DI water. VA-PC-DMF testing solution was prepared by adding VA (0.22 mM final concentration) and PC (1.1 mM final concentration) to DMF. The CPE membrane was soaked in the VA-PC-DMF solution for various time periods to determine the best duration to complete the reaction.

To further understand changes in the membrane surface chemical structure during VA incorporation and the new antifouling properties, a second modification solution was prepared by mixing PC with DMF without VA. This PC-DMF testing solution was used for CPE membrane modification following the same reaction procedure as described above.

The PC-DMF modified membrane was used as VA negative control for evaluating the membrane performance. All membrane samples were rinsed by DI water thoroughly after modification reaction to avoid chemical carry over and dried before further analysis.

2.2. Spectroscopy

Unmodified and modified membrane samples were analyzed using Raman and FTIR spectroscopy. For Raman *in vivo* microscopy (Renishaw, Wotton-under-Edge, UK) analysis, samples were placed directly under the diode laser at an excitation of 532 nm (visible light range) under 50,000 magnifications. Spectrums were collected using either 3 mW or 15 mW of laser power and 1 s exposure time from about ten random locations along the membrane surface. The baseline was corrected for all spectrums using WIRE 3.4 software (Renishaw, Wotton-under-Edge, UK). For FTIR (R-4700 type A, JASCO Corp., Tokyo, Japan) analysis, each membrane sample was examined at room temperature between 400–4000 cm^{-1} . The signals were collected for 32 scans with a scan interval of 0.241 cm^{-1} and optical resolution of 1 cm^{-1} .

The target ether bond and other relevant peaks were determined by comparing the FTIR and Raman spectra of reference samples (i.e. unmodified CPE membrane, VA, DMF, and PC in its natural state) with VA-PC-DMF and PC-DMF modified CPE membrane samples. Spectral peaks were also identified and confirmed by comparing with literature reports [21–29].

2.3. Stability of VA modified membrane

VA modified membrane stability was assessed under cross-flow conditions without high-pressure and recirculation of the return line (Fig. S1A). VA-PC-DMF-CPE modified membrane was framed in a membrane cassette holder (Centramate PE TFF, Pall Corp., Portsmouth, UK). The membrane surface (dimensions: $9.4 \times 25.1 \times 19.3$ cm and the cross-sectional area of the water path defined by the spacer and o-ring was 0.01 m^2) was exposed to DI water with cross-flow of 360 $\text{L}/\text{m}^2/\text{h}$ for 24 h driven by a peristaltic pump (Masterflex; Cole-Parmer, Vernon Hills, USA). After drying, the modified membranes were analyzed using Raman spectroscopy. This test was repeated twice to confirm the reproducibility of the results. Raman spectra on membrane surfaces were compared before and after the flushing experiments to identify any loss of peaks.

VA-PC-DMF-CPE and PC-DMF-CPE modified membranes were also subjected to cleaning by sodium hypochlorite solution [30,31] (free chlorine: ~ 8 ppm and pH ~ 9.6) for 30 and 60 min. Afterwards, membranes were rinsed using DI water and dried for Raman spectroscopy analysis. A handheld photometer (OMEGA engineering, Stamford, USA) was used before and after cleaning tests to determine total chlorine and free chlorine concentrations. pH values were measured using Fisher Scientific™ accumet™ AB15 Basic and BioBasic™ pH/mV/°C Meters (Fisher Scientific, Pittsburgh, USA).

2.4. Membrane anti-biofouling potential

Biofilm formation on the surface of modified and unmodified membranes was evaluated in the membrane cassette system in recirculation mode (Fig. S1B). For comparisons, unmodified and modified (both PC-DMF-CPE and VA-PC-DMF-CPE) membranes were inserted in parallel membrane cassette holders (dimensions: $9.4 \times 25.1 \times 19.3$ cm and the cross-sectional area of the water path defined by the spacer and o-ring was 0.01 m^2) and were subjected to the same cross-flow driven by the same peristaltic pump. To test the membrane anti-fouling potential, aerobic activated sludge was collected from the Orange County Sanitation District wastewater treatment facility (Fountain Valley, USA) in September 2016. The concentration of mixed liquor suspended solids (MLSS) and volatile suspended solids (VSS) were 7.6 g/L and 6.1 g/L,

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