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# Fouling characterization in entrapped cells-based-membrane bioreactor treating wastewater



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### Chaipon Juntawang<sup>a</sup>, Chaiwat Rongsayamanont<sup>b</sup>, Eakalak Khan<sup>c,\*</sup>

<sup>a</sup> Environmental and Conservation Sciences Program, North Dakota State University, Fargo, ND 58108, USA
<sup>b</sup> Faculty of Environmental Management, Prince of Songkla University, Songkhla 90112, Thailand
<sup>c</sup> Department of Civil and Environmental Engineering, North Dakota State University, Fargo, ND 58108, USA

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#### ABSTRACT

Membrane fouling in an entrapped cells-based-membrane bioreactor (E-MBR) treating synthetic medium strength municipal wastewater was investigated. Laboratory scale models of E-MBR and regular (suspended cell) membrane bioreactor (S-MBR) were operated in parallel. The two systems showed comparable performances on organic matter and nutrient removal. The removal efficiencies of soluble chemical oxygen demand and ammonia of E-MBR were  $95.6 \pm 0.9\%$  and  $92.6 \pm 2.1\%$  while those of S-MBR were  $95.8 \pm 0.8\%$  and  $93.3 \pm 0.8\%$ . Membrane fouling was monitored based on transmembrane pressure through a constant permeate flux at 10.63 L/m<sup>2</sup> h. Due to lower concentrations of bound extracellular polymeric substances (bEPS) and soluble microbial products (SMP), E-MBR experienced less fouling and provided longer operation time before required chemical cleaning compared to S-MBR (16 days for E-MBR and 9 days for S-MBR). bEPS in E-MBR consisted of higher molecular weight compounds and had a broader molecular weight distribution than those in S-MBR. The Fourier transform infrared analysis of bEPS suggested proteins and carbohydrates as major components. According to the 3dimension fluorescence excitation-emission matrix spectra, SMP in E-MBR were mainly tryptophan, protein-like and humic acid-like substances, while those in S-MBR were mainly humic acid-like substances, hydrophobic acid substances and fulvic acid substances. The particles size of sludge in E-MBR was smaller than that in S-MBR. The delayed membrane fouling in E-MBR is a great advantage as it lowers costs associated with membrane cleaning processes and prolongs the membrane lifespan.

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#### 1. Introduction

With more stringent standard for treated effluents, membrane bioreactors (MBRs) have been a process of choice for wastewater treatment and reuse. MBRs provide several advantages over conventional treatment process (activated sludge) including superior quality effluent, less biomass yields and more compact design [1]. Therefore, MBRs are currently used as a secondary treatment unit for numerous full-scale municipal wastewater treatment and reclamation facilities in the United States and Europe [1]. At biochemical oxygen demand (BOD) and ammonia loading rates of 0.2–0.7 kg/m<sup>3</sup>·d and 0.05–0.17 kg N/m<sup>3</sup>·d, the treatment efficiencies of MBRs were excellent achieving effluent BOD, ammonia and total suspended solids of 0.7–3.0 mg/L, 0.5–2.0 mg N/L, and

1–2.5 mg/L, respectively [2,3]. However, membrane fouling is still a major drawback that hampers widespread and full-scale applications of MBRs. Fouling is a reduction of membrane permeability that is originated by adsorption or accumulation of deposits on the surface and/or in the pores of membrane during operation. Loss of membrane permeability results in higher transmembrane pressure (TMP) leading to a higher operating cost of MBRs for keeping a constant permeate flux with increased applied pressure and frequent chemical cleaning [1,4].

Fouling, commonly found in submerged MBRs operation, is deposition of sludge cake onto membrane surface (cake deposition) and clogging of small deposits within membrane pores (pore blocking). Microbial products including bound extracellular polymeric substances (bEPS) and soluble microbial products (SMP) that are released during utilization, growth and decay of active cells in sludge are currently considered as the predominant cause of cake deposition and pore blocking in MBRs [5]. bEPS consist of proteins, polysaccharides, nucleic acids, lipids, and humic acids, which are located at or outside the cell surface. SMP are soluble organic pools

<sup>\*</sup> Corresponding author at: Civil and Environmental Engineering Department, North Dakota State University, CIE Building, Room 201, Dept. # 2470, PO Box 6050, Fargo, ND 58108-6050, USA.

E-mail address: eakalak.khan@ndsu.edu (E. Khan).

that occur during substrate metabolism, and cell decay and bEPS hydrolysis [6,7]. Cake resistance is often contributed by hydrophobic SMP which have molecular weights in a range of 10–100 kDa [8].

The primary concern of fouling in long term operation is irreversible fouling as chemical cleaning cannot remove all accumulation from membrane pores. Another concern is high chemical use will damage the membrane texture and shorten the membrane lifetime. Thus, finding a strategy to prevent accumulation of bEPS and SMP on membrane surface is worthy of investigation.

A study on suspended-growth MBR (S-MBR), in which bacteria can grow freely, showed higher bEPS and SMP (in terms of proteins and carbohydrates) in the system when compared with attached-growth MBR (AG-MBR), which is a MBR containing biofilm carriers [9–11]. Khan et al. [10,11] compared AG-MBR and moving biofilm MBR (MB-MBR), which is a specific type of AG-MBR containing free-floating biofilm carriers, with S-MBR. They found that SMP concentration in S-MBR was higher than those in AG-MBR and MB-MBR indicating more severe fouling in S-MBR. In addition, Di Trapani et al. [12] and Rodríguez et al. [13] reported less fouling in MB-MBR than S-MBR. These previous studies are in agreement that biofilm-based MBR have less/slower fouling condition compared to S-MBR.

The concept of attached-growth/biofilm is similar to cell entrapment/immobilization principle. Entrapped cells-based-MBR (E-MBR) reduces the concentrations of bEPS and SMP leading to less fouling compared to S-MBR [9,10,14]. Tsen et al. [15] reported that entrapment matrices can provide protection to the cells against unsatisfied environment such as toxic compounds or low pH. Moreover, the porosity within the entrapment cells allows the diffusion of substrates and products across the matrix but prevent the release of cells into the bulk liquid. Polyvinyl alcohol has been successfully used as an entrapment matrix for wastewater treatment because of high durability, applicability in a wide range of pH (4–10), nontoxicity to bacteria, and higher specific growth and specific substrate utilization rates when compared to alginate and carrageenan [16–18]. Gel entrapped cells-based-MBR has been studied but none of them specifically explored on how entrapped cells can reduce/delay fouling in MBR system [19,20].

The objective of this study was to investigate the role of entrapped cells on membrane fouling and fouling mechanism in E-MBR treating medium-strength domestic wastewater through analysis of membrane resistance and characterizations of bEPS and SMP. A fouling condition in E-MBR was observed in parallel with S-MBR. S-MBR was used for comparison in this study because it is widely used while attached-growth MBR has not been applied at full-scale. It is hypothesized that the porous gel matrix can prevent not only cells but also bEPS and SMP releases into bulk liquid resulting in lower membrane fouling because of reduction in cake deposition and pore blocking.

#### 2. Materials and methods

#### 2.1. Synthetic wastewater and chemicals

Synthetic medium-strength domestic wastewater, which has a soluble chemical oxygen demand (SCOD):N:P ratio of 100:5:0.7 [21], was prepared by adding 0.5 mL of concentrated CH<sub>3</sub>COOH (as 500 mg/L SCOD), 100 mg of NH<sub>4</sub>Cl, 16 mg of KH<sub>2</sub>PO<sub>4</sub>, 4 mg of FeCl<sub>3</sub> 6H<sub>2</sub>O, 11 mg of CaCl<sub>2</sub>, 17 mg of MgSO<sub>4</sub>, 8 mg of KCl, 8 mg of NaCl and 350 mg of NaHCO<sub>3</sub> into one liter of de-ionized water. Polyvinyl alcohol (99.0–99.8% fully hydrolyzed, J.T. Baker, USA) was used as a gel entrapment matrix. All chemicals used are the American Chemical Society reagent grade.

#### 2.2. Preparation of entrapped cells

Mixed liquor taken from an aeration tank of the Moorhead wastewater treatment facility (Minnesota, USA) was inoculated to start up S-MBR and E-MBR. Entrapped cells in E-MBR were prepared using a procedure described in Chen and Lin [22]. In brief, the concentrated sludge (80 g wet weight) was thoroughly mixed with 535 ml of a 10% polyvinyl alcohol gel solution (w/v). The mixture was then dropped into a saturated boric acid solution to form spherical beads. The formed gel beads (diameter of  $2.4 \pm 0.15$  mm) were transferred and incubated for 4 h in a saturated orthophosphate solution for hardening resulting in phosphorylated polyvinyl alcohol (PPVA) gel beads. Both boric acid and orthophosphate were not adsorbed onto the polyvinyl alcohol gel but rather incorporated into the gel structure [16,22]. The beads were washed thoroughly with de-ionized water and inoculated into E-MBR.

#### 2.3. Membrane bioreactor setup and operation

For S-MBR and E-MBR setups, two rectangular acrylic tanks with working volume of 10 L were used. Each tank consisted of two compartments divided by an acrylic plate. One compartment was for aeration zone (7 L) and the other was for filtration zone (3 L) (Fig. 1). Both E-MBR and S-MBR had the same configuration and operation conditions except that E-MBR was inoculated with entrapped cells instead of mixed liquor. A hollow fiber membrane module (ZW-1, GE Water and Power, Canada) with a pore diameter of 0.04  $\mu$ m and an effective surface area of 0.047 m<sup>2</sup> was submerged in the filtration zone of MBR.

Oxygen was supplied using a laboratory air system and diffused through small stone diffusers to maintain sufficient mixing and aerobic condition in the reactors. Two peristaltic pumps were used as influent and permeate pumps (Model 7553-60, Barnant, USA and Model 7554-90, Thermo Fisher Scientific, USA). TMP was monitored during filtration via a vacuum gauge (Model 14902.5, Ashcroft, USA). Under a 1-day hydraulic retention time (HRT), a constant permeate flux at 10.63 L/m<sup>2</sup> h (LMH) was maintained through 15 min backflushing and air scouring for every 3 h. Permeate water was pumped from the permeate tank to backflush membrane while air was blown through an air pump (Whisper 100, Tetra, USA) to scour cake deposited on the membranes. An electronic timer (ODT309-M2, Smart Electrician, USA) was used to control the backflush pump and air scouring system. Membrane was cleaned when TMP reached 55 kPa or it was not possible to maintain a constant permeate flux by soaking under 200 ppm sodium hypochlorite for a minimum of 5 h and then moving to a 5 g/L citric acid solution for a minimum of 5 h. The reactors were operated at room temperature  $(22.2 \pm 0.9 \text{ °C})$  after steady state for 90 days without wasting sludge. Conditions for the reactor operation are summarized in Table 1.

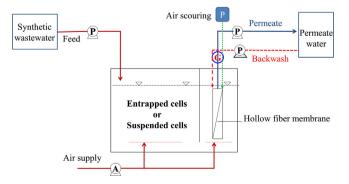


Fig. 1. Schematic diagrams of E-MBR and S-MBR.

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