

# Effect of biopolymer clusters on the fouling property of sludge from a membrane bioreactor (MBR) and its control by ozonation

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## ABSTRACT

Organic substances in the liquid phase of the sludge in a membrane bioreactor (MBR) have a profound impact on membrane fouling. In this study, a single-fibre microfiltration apparatus was developed to investigate the fouling propensity of MBR sludge and the effectiveness of ozonation in membrane fouling mitigation. The results show that biopolymer clusters (BPC) in the MBR suspension had a significant influence on the fouling potential of the sludge. An increase in BPC concentration by 20% and 60% from around 3.5 mg/l in the mixed sludge liquor drastically increased the fouling rate by 120% and 300%, respectively. Ozonation of the BPC solution greatly reduced the detrimental role of BPC in membrane fouling. An ozone dose of 0.03 mg/mg TOC of BPC could reduce the mean BPC size from 38 to 27  $\mu\text{m}$ , which was further reduced to 12  $\mu\text{m}$  at 0.3 mg  $\text{O}_3$ /mg TOC of BPC. In addition to BPC destruction, ozonation apparently also modified the surface properties of BPC, resulting in an increase in the filterable fraction and a decrease in the liquid viscosity. Based on the experimental findings, an approach for MBR membrane fouling control is proposed that applies ozonation to the supernatant containing BPC in a side-stream application.

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## 1. Introduction

Membrane bioreactors (MBRs) are increasingly being used as an advanced technology for biological wastewater treatment and reuse. With the use of a membrane for sludge filtration, the MBR ensures complete solid–liquid separation [1,2]. In MBRs, the sludge age and concentration can be effectively manipulated, affording this type of bioreactors several advantages over the conventional activated sludge (CAS) process [3]. At the same time, however, because of the retention by the membrane, some of the soluble microbial products (SMP) and other colloidal substances are unable to escape from the system with the effluent [4,5]. The organic interception by membrane filtration results in the formation and accumulation of organic foulants in the MBR sludge suspension, which in turn worsens the membrane-fouling problem.

The effect on membrane fouling of liquid-phase organic substances in the MBR sludge mixture has long been recognised [6–10]. Recent research reveals the presence of a group of large-sized organic solutes, termed biopolymer clusters (BPC), in MBR systems [11–13]. BPC are neither biomass flocs nor SMP or extracellular polymeric substances (EPS). They can be larger than 10  $\mu\text{m}$  in size, and are formed by the affinity clustering of SMP and loose EPS on

the membrane surface [14]. It has been suggested that BPC may facilitate sludge deposition and the fouling layer formation on the membrane surface, and the detrimental role of BPC in membrane fouling has been demonstrated qualitatively during the operation of MBR systems [12,14]. However, more systematic studies remain to be conducted to determine the correlation between the membrane fouling rate and the BPC content of the sludge mixture. In addition, the effect of changes in BPC properties on the fouling potential of the MBR sludge also merits investigation.

The reduction or modification of BPC in MBR sludge mixture is expected to be beneficial for the control of membrane fouling. Removal of BPC and their precursors, such as SMP and loose EPS, is an option, and indeed the use of adsorbents or coagulants in the MBR mixed liquor has been found effective in decelerating membrane fouling [15–19]. However, continuous addition of these chemicals may either be harmful to the membrane due to physical abrasion, as is the case for granular activated carbon, or affect the MBR treatment performance, as is the case with some coagulant metal ions (e.g., Fe(II) and Fe(III) that are reportedly toxic to the nitrifying bacteria [20]). More recently, the ozonation of bulk sludge has been tested as a means of membrane fouling control during continuous MBR operation [21–24]. The results show that at appropriate doses the membrane fouling rate can be effectively reduced, meanwhile, ozonation coupled with MBR appears to be an effective method for sludge reduction and toxic organic wastewater treatment [23,24]. However, a possible overdose of ozone and its impact on the biomass activity is a concern with

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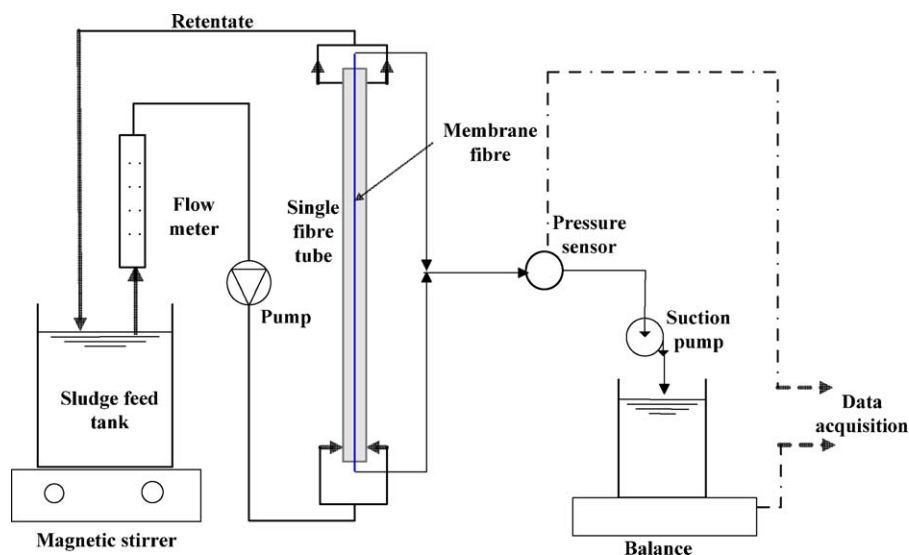


Fig. 1. Schematic diagram of the single-fibre MF testing apparatus.

direct sludge ozonation. Moreover, the underlying mechanisms of sludge ozonation for membrane fouling mitigation are not well understood.

There is thus a need to determine the effect of BPC in MBR sludge mixture on membrane fouling, and to investigate the effectiveness of the ozonation of BPC in reducing the fouling propensity of sludge. In this study, a lab-scale MBR was operated to supply both biomass sludge and BPC dispersion. A newly designed single-fibre microfiltration (MF) system was fabricated for the membrane filtration-fouling tests on different sludge – BPC mixture samples under well-controlled hydrodynamic conditions. Ozonation was applied to the BPC solution only, rather than the entire sludge mixture, before mixing into the sludge suspension. The objectives of the experimental study were (1) to determine the fouling propensity of MBR sludge with different BPC contents, and (2) to investigate the effectiveness of the ozonation of BPC in minimising membrane fouling during sludge filtration. The mechanism of sludge ozonation to mitigate fouling was also identified, and on this basis a more reliable ozonation approach for MBR fouling control is proposed.

## 2. Materials and methods

### 2.1. Filtration setup and operation

A single-fibre filtration apparatus was fabricated for the sludge filtration and fouling tests (Fig. 1). The apparatus was made of a plexiglass tube 1.5 cm in internal diameter and 50 cm in height. A polyethylene (PE) hollow-fibre MF membrane (pore size = 0.4  $\mu\text{m}$ , diameter = 0.14 cm, working length = 40 cm, surface area = 16 cm<sup>2</sup>, Mitsubishi Rayon, Japan) was installed along the centreline of the filtration tube. The sludge suspension in a feed tank was pumped through the MF test tube by a helical pump (SELTZ-L40 II, Hydor, USA). A constant cross-flow rate of 2 l/min (0.19 m/s) was applied by the recirculation of the sludge suspension for continuous membrane surface cleaning. The permeate was drawn out through the MF membrane by a suction pump (MasterFLEX, Cole-Parmer, USA) at a constant flux of 37.5 l/m<sup>2</sup> h. An electronic balance (Arrw 60, OHAUS, USA) was used to record the permeate production during the filtration-fouling tests. Unless sampled for analysis, the collected permeate was returned manually to the feed tank at regular intervals to maintain the same sludge concentration. A pressure sensor (PTX Ex-0129, Druck, USA) was installed before the suction pump to record the trans-membrane pressure (TMP) during sludge filtration. Both the permeate production and TMP data were transferred to a PC for continuous data recording (Fig. 1). The membrane fouling rate was measured by the increase in TMP with the amount of permeate produced (filtrate depth,  $L$ ), or  $\Delta\text{TMP}/\Delta L$ . After each filtration-fouling test, the membrane fibre was taken off the filtration tube and washed with 100 ml of DI water at 40 °C to recover all of the sludge and foulants deposited on the membrane surface. The sludge and foulant dispersion were then settled for 2 h at 4 °C and the supernatant was analysed for total organic carbon (TOC) and chemical composition, including proteins (PN), polysaccharides (PS) and humic-like substances (HS). The sludge in the dis-

persion was collected on a filter, dried for 2 h at 105 °C and then weighed to obtain the suspended solids (SS) content.

### 2.2. MBR activated sludge and biopolymer clusters

The sample activated sludge (AS) and biopolymer substances for the filtration tests were collected from a submerged MBR (SMBR). The laboratory SMBR had a working volume of 5 l and contained a submerged 0.4  $\mu\text{m}$  polyethylene MF module (surface area = 0.2 m<sup>2</sup>, Mitsubishi Rayon, Japan). The SMBR system had been in stable operation for more than four years before the present experiment [12,25]. The influent (feeding wastewater) to the SMBR was a mixture of a glucose-based synthetic wastewater prepared according to the basic recipe given in the Environmental Engineering Process Laboratory Manual of the AEEPS [26] and domestic sewage collected from the Stanley Sewage Treatment Works in Hong Kong. The sewage fraction supplied around 10% of the total organic load in the influent. The wastewater influent had a chemical oxygen demand (COD) of around 500 mg/l and a COD:N:P ratio of 100:9:3. NaHCO<sub>3</sub> was added to the influent at 50 mg/l or higher to maintain the pH of the MBR suspension between 6.5 and 7.5. The biomass concentration, food-to-microorganism (F/M) ratio, solid retention time (SRT) and hydraulic retention time (HRT) of the SMBR system were 10 g/l, 0.2 g COD/g SS d, 25 d and 8 h, respectively.

The AS mixture collected from the SMBR was settled for 1 h, and the settled sludge was then diluted with a 0.05% NaCl solution to a mixed liquor suspended solids (MLSS) concentration of 3 g/l. Large organic substances, or biopolymer clusters, were obtained from the cake sludge (CS) deposited on the surface of the membrane in the SMBR. When the membrane was seriously fouled, the CS layer was scraped off the membrane using a spatula. The CS was then re-suspended and dispersed by stirring it in a 0.05% NaCl solution. The CS suspension was then separated by sedimentation at 4 °C for 12 h and the supernatant was collected. The organic substances in the CS supernatant were regarded as biopolymer clusters [12,14]. The CS supernatant, or BPC solution, was analysed for TOC and PN, PS and HS content.

The BPC solution was added into the AS suspension (3 g/l) at different doses. Each sludge suspension was then tested for its fouling propensity using the single-fibre MF filtration apparatus. In this way, the effect of the BPC content in the sludge mixture on the fouling potential of the sludge during membrane filtration was determined.

### 2.3. Ozonation of the BPC solution

Ozonation was also applied to the BPC solution with an intention to modify the BPC properties before their addition to the sludge. Ozonation was performed quantitatively by adding ozone-containing water into the BPC solution. Ozone was generated in the gaseous phase by an ozone generator (5000 BF, Enaly, China) that was supplied with pure oxygen. To dissolve the ozone in water and prepare an ozone solution, 500 ml ultra-pure water (Milli-Q-Advantage, Water Purification, Millipore, USA) was bubbled with the ozone gas at 4 °C for 10 min or longer. The ozone concentration achieved in the ozone solution was about 8 mg/l. A pre-determined amount of the ozone solution was then added to 30 ml of the BPC solution. The mixed solution was placed in the dark and stirred for 5 min at 60 rpm to ensure complete ozonation. Similar to the previous sludge filtration tests, the ozonated BPC solution was added into the AS suspension at different doses, and the sludge mixtures were then tested for their fouling potential using the single-fibre filtration apparatus.

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