

Short communication

Characterization of membrane foulants in a pilot-scale powdered activated carbon–membrane bioreactor for drinking water treatment



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ABSTRACT

The external and internal foulants of a pilot-scale powdered activated carbon–membrane bioreactor (PAC–MBR) used for drinking water treatment were systematically examined by scanning electron microscopy (SEM), three-dimensional excitation emission matrix (EEM) fluorescence spectroscopy, Fourier transform infrared spectroscopy (FTIR) and energy dispersive X-ray (EDX) analysis. The results showed that external fouling, which comprised 31.68% of the total fouling, was caused by the deposition of a large amount of biological PAC on the membrane surface. Bacteria and organic matter comprised only a small fraction of the external foulants. Biologically derived proteins and polysaccharides were the major constituents of the internal foulants. EDX analysis indicated that the external and internal foulants also included inorganic elements such as Mg, Al, Si, Ca, Mn and Fe. During the operation of PAC–MBR, low flux and effective physical cleaning protocols should be adopted; proteins, polysaccharides and inorganic elements in the bioreactor should also be controlled.

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1. Introduction

The use of powdered activated carbon–membrane bioreactor (PAC–MBR) in drinking water treatment has recently received considerable interest [1–6]. PAC–MBR systems show good performance regarding the removal of pathogens, ammonia (NH₃-N), disinfection byproduct (DBP) precursors and biodegradable organic matter [4,7,8], and have potential as an alternative to biological activated carbon filtration process [4]. Unfortunately, as with all membrane filtration processes, membrane fouling is a major hurdle in the extensive use of PAC–MBR. To control membrane fouling, it is critical to understand the characteristics of membrane foulants.

There are two major origins of membrane foulants. One source is the substances present in feed water. For instance, natural organic matter (NOM) in raw water is widely perceived as the major foulants for microfiltration/ultrafiltration (MF/UF) processes in drinking water treatment [9]. The other foulant source is the substances generated during the process. For instance, in MBR systems for wastewater treatment, extracellular polymeric substances

(EPS), the main metabolic products of bacteria, are primarily responsible for membrane fouling [10,11]. In PAC–MBR for drinking water treatment, both types of the aforementioned origins are involved (i.e., the NOM in feed water and the bacteria in the bioreactor), which makes the foulants of this process special and complex.

Previous research on PAC–MBR mainly focused on the removal efficiency of various pollutants and the optimization of operational parameters [1,2,4,8,12]. Limited information is available regarding foulants characteristics. A detailed investigation of the foulants in PAC–MBR will also allow us to make comparisons between PAC–MBR and other well-understood processes (e.g., MBR and MF/UF processes). Therefore, a better understanding of the foulants in PAC–MBR is required.

In this study, a hollow fiber UF membrane that was fouled in a long-term pilot-scale submerged PAC–MBR for surface water treatment (with good performance) was investigated to characterize the internal and external foulants. The foulants were evaluated using scanning electron microscopy (SEM), three-dimensional excitation emission matrix (EEM) fluorescence spectroscopy, Fourier transform infrared spectroscopy (FTIR) and energy dispersive X-ray (EDX) analysis. Based on the obtained data, the fouling features of the PAC–MBR were compared to those of MBR and MF/UF systems reported in previous studies. Countermeasures for fouling control of the PAC–MBR were proposed according to these results.

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2. Materials and methods

2.1. Pilot-scale PAC–MBR configuration and operation

A long-term (six months) submerged hollow fiber membrane filtration system developed at the pilot scale ($2.2 \text{ m}^3/\text{h}$) was continuously operated at a drinking water treatment works (Dongguan, Southern China). A schematic diagram of the PAC–MBR is presented in Supplementary data Fig. S1. A UF membrane module (Litree, China) for full-scale application was used. The UF membrane was made of polyvinylchloride (PVC), with a nominal pore size of $0.01 \text{ }\mu\text{m}$ and a total membrane area of 110 m^2 . The effective volume of the bioreactor tank was 0.75 m^3 .

Settled water was employed as the feeding for the PAC–MBR. Polyaluminum chloride (PACl) at a dosage of $6\text{--}8 \text{ mg PACl/L}$ raw water was adopted as the coagulant in the drinking water works. To simulate more severe ammonia pollution, NH_4Cl (analytical grade) was dosed into the feed water. The average feed water quality of the PAC–MBR is listed in Supplementary data Table S1.

The system was operated continuously from December 29, 2011 to June 25, 2012. Membrane filtration was performed at a constant flux of $20 \text{ L}/(\text{m}^2 \text{ h})$. Periodic backwashing (15 min filtration; 20 s backflush at $60 \text{ L}/(\text{m}^2 \text{ h})$) was implemented. The hydraulic retention time (HRT) was set at 20 min , and the sludge retention time (SRT) was set at 15 days . PAC (see Supplementary data for detailed information) at a dosage of 3 mg/L feed water was directly added to the bioreactor tank once per day. The level of mixed liquor suspended solids (MLSS) was maintained at $2.3\text{--}2.6 \text{ g/L}$ (Supplementary data Fig. S3). Intermittent aeration ($20 \text{ s on}/3\text{--}6 \text{ min off}$) at an intensity of $50 \text{ m}^3/(\text{m}^2 \text{ h})$ (calculated based on the area of the bottom of the bioreactor tank) was used. The system was automatically controlled by a programmable logic controller (PLC). The trans-membrane pressure (TMP) was recorded by the PLC during the operation. The pollutants removal efficiency listed in Supplementary data Table S1 indicated that the performance of the PAC–MBR was excellent.

After a six-month operation, approximately 10 m^2 of the fouled membrane was taken out carefully in consideration of the vulnerability of fouling cake. This fouled membrane was used to characterize the external and internal foulants.

2.2. Extraction of the foulants

The external foulants on the membrane surface were carefully scraped off using a plastic brush and simultaneously flushed with Milli-Q water. Approximately 2 m^2 of the membrane was scraped, and 8 L of mixed liquor was obtained. The collected sample was fully mixed using a magnetic blender for 24 h . The mixed liquor of the external foulants consisted of three fractions (i.e., suspended solids, colloidal matter and dissolved matter). After centrifugation at 3500 rpm for 10 min , the supernatant was sampled and referred to as colloidal and dissolved matter. A portion of this supernatant was further filtered using a $0.45 \text{ }\mu\text{m}$ mixed cellulose ester membrane (Taoyuan, China), and the filtrate was collected and designated as dissolved matter.

The internal foulants were extracted from the membrane without external foulants using 0.01 mol/L NaOH. Fibers with a membrane area of 0.25 m^2 were soaked in the alkaline solution (1 L) for 24 h at $20 \text{ }^\circ\text{C}$. A permeability recovery of 91% was achieved, indicating that most of the internal foulants were extracted. The chemical solution was adjusted to neutral pH using HCl and saved at $4 \text{ }^\circ\text{C}$ for further analysis.

2.3. SEM–EDX analysis

Morphological observation and chemical components analysis of the foulants were conducted by SEM–EDX analysis. The fouled

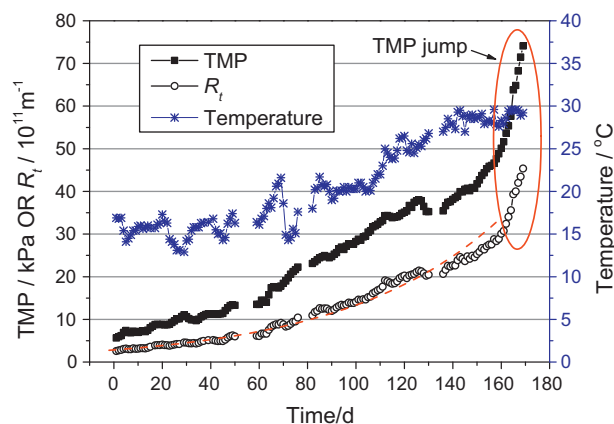


Fig. 1. Evolution of the TMP and total resistance (R_t) during the pilot running.

membrane fibers with and without external foulants were collected and prepared for observation. Three pieces of parallel samples were cut, dried under ambient conditions, gold-coated by sputtering and then observed using a SEM–EDX system (HITACHI S4800 HSD, Japan). The chemical components of the foulants were simultaneously determined by EDX analysis. For each sample, SEM images and energy spectra were obtained in several different positions, and typical results were used for analysis.

2.4. EEM fluorescence spectroscopy analysis

EEM was used to characterize the internal foulants and the dissolved matter in the external foulants. Fluorescence was measured in a 1 cm cuvette using a fluorescence spectrophotometer (F7000, Hitachi, Japan) at ambient temperatures ($21 \pm 1 \text{ }^\circ\text{C}$). An EEM spectrum of Milli-Q water was subtracted from the EEM spectrum of each sample. Because qualitative rather than quantitative results were needed, the inner-filtering and metal-binding effects were not taken into consideration.

2.5. FTIR analysis

FTIR was used to investigate the internal foulants as well as the colloidal and dissolved matter in the external foulants. The supernatant sample of the external foulants (2 L) and the solution sample of the internal foulants (0.8 L) were dried at $60 \text{ }^\circ\text{C}$ to obtain powdered samples of the foulants. Although the powders of the internal foulants contained NaCl, it did not have a substantive influence on the spectra [13]. The powders were mixed with KBr and analyzed using a FTIR spectrometer (PerkinElmer Spectrum One, USA) at a resolution of 4 cm^{-1} .

3. Results and discussion

3.1. TMP evolution and fouling resistance

The increases in TMP and total resistance (R_t) during the long-term membrane filtration are shown in Fig. 1. The R_t data were adjusted to equivalent values at $20 \text{ }^\circ\text{C}$ considering the change in water viscosity. Chemical cleaning was not performed during the extended period of operation, and the membrane fouling rate was moderate (approximately 0.25 kPa/d). The R_t increased exponentially at a constant flux of $20 \text{ L}/\text{m}^2 \text{ h}$, and a sudden increase in the R_t and TMP (TMP jump) was observed on the last few days. This TMP jump could be ascribed to inhomogeneous fouling and cake compression during filtration [14]. In MBR for wastewater treatment, either an approximately exponential or three-stage TMP transient is typically encountered [10,14]. Therefore, the TMP evolution of

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