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Combined influence by humic acid (HA) and powdered activated carbon (PAC) particles on ultrafiltration membrane fouling

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ABSTRACT

This research focused on the combined fouling by humic acid (HA) and powdered activated carbon (PAC) which possibly occurred in the PAC-ultrafiltration (PAC-UF) hybrid process. The membrane fouling and the HA transmission were systematically investigated with HA (Aldrich, 10 mg/L) and PAC (with different sizes and dosages) in the presence and absence of 0.5 mM calcium. Experimental results suggested that PAC and HA exhibited a significant synergistic fouling effect when they formed a fouling cake together. In the absence of calcium, the adsorption of HA on PAC surface increased the repulsive interaction between PAC particles, and thus a fragile cake layer was formed by the PAC–HA mixture. In the presence of calcium, however, a dense cake layer was formed due to particle bridging by HA–Ca–HA associations. The combined fouling effect increased as the PAC dosage increased and the PAC size decreased, both in the presence and absence of calcium, suggesting that the steric effect play an important role in the combined fouling. In the absence of calcium, an unexpected phenomenon that the HA transmission increased in the presence of PAC suggested that in addition to the steric effect, the hindered foulant back diffusion effect was also involved in the combined fouling.

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1. Introduction

Ultrafiltration (UF) coupled with powdered activated carbon (PAC) adsorption under integrated configurations (PAC-UF hybrid process) is a proven process to remove organic micropollutants and reduce the amount of disinfection byproducts [1–3]. Organic compounds which are normally not rejected by UF membrane could be adsorbed by PAC particles that are then separated from the product water by the membrane [4]. However, PAC is not tailor-made to match the requirements of the operating of UF, and possibly undermines the performance of UF [2]. One of the core questions is that the addition of PAC may influence the fouling behavior of the UF membrane.

Over the past two decades, extensive researches have been conducted to investigate the impact of PAC on the membrane fouling in PAC-UF hybrid process, but the results were controversial. Some studies demonstrated that the presence of PAC

allows reducing the amount of organic matter deposited on membrane and thereby mitigating membrane fouling [5–11]. But some other studies have reported the opposite [3,8,11,12] and suggested that PAC adversely affected membrane fouling. Meanwhile, there are also some studies that showed that PAC addition neither promoted nor controlled the membrane reversible fouling [13,14]. It is widely accepted that inorganic particles and organic matter can result in combined membrane fouling, which cannot easily be predicted from the fouling behavior of the individual components alone [15,16]. Therefore, the controversial results imply that PAC potentially leads to combined fouling with organic matter in PAC-UF hybrid process.

Previous studies mainly focused on the combined fouling by inorganic colloidal particles and organic matter in nanofiltration (NF) process [16–18], forward osmosis (FO) process [19,20], and UF process [21,22]. There are limited works that discuss the combined fouling by PAC and organic matter in PAC-UF hybrid process. In some studies, natural organic matter (NOM) is believed to act as a glue that binds the PAC particles to one another and to the membrane surface [23]. Zhao et al. [24] reported that metal ions played important roles in the deposition of PAC on the membrane surface, and that a higher fouling resistance was achieved with a

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larger PAC. Nevertheless, combined fouling by PAC and organic matter is still poorly understood; its underlying mechanisms and influencing factors need to be further studied.

Moreover, the combined fouling by PAC and organic matter cannot easily be inferred from the published literatures. Firstly, the sizes of the inorganic colloidal particles studied in previous researches were 20–450 nm [18,22,25], which were much smaller than the size of PAC that generally used in water treatment (tens of micrometers). Secondly, unlike NF and RO membrane, UF membrane cannot reject salts. Consequently, the combined fouling in PAC-UF hybrid process may differ from what was previously studied.

The main objective of this study was to gain more insight into the combined fouling by organics and PAC particles on UF membrane fouling. Humic acid (HA), a commercially available PAC and a 100 kDa UF membrane were used in this study. The combined fouling effects of HA and PAC with different sizes and dosages in the presence and absence of calcium were systematically investigated, and the underlying mechanisms of the combined fouling were discussed.

2. Materials and methods

2.1. HA and PAC

HA (Aldrich) was selected as the model organic matter for three reasons: (1) humic substances, which are considered as major components of dissolved organic matter in natural water, play a significant role in UF membrane fouling [26]; (2) it has been well characterized and used in numerous fouling studies [21,25,27]; (3) they lead to a significant cake fouling [27]. Stock HA solution (2 g/L) was prepared according to the method provided by Li et al. [28], and was stored in the dark at 4 °C. The concentration of working HA solution was 10 mg/L, and the corresponding dissolved organic carbon was 4.38 mg C/L (to simulate the polluted surface water, e.g., the concentration of organic matter in the Songhua river was ~4.85 mg C/L [29]). The background solutions were consisted of Milli-Q water, NaCl, NaHCO₃ (1 mmol/L), and calcium (0 or 0.5 mmol/L). The pH of the solutions was adjusted to 7.5 ± 0.1 with 0.1 mmol/L HCl, and the total ionic strength was 10 mmol/L. The HA solution was filtered through 0.45 μm cellulose ester membrane (Taoyuan, China) prior to use. The adsorption of the HA by the filter has been proved to be negligible in preliminary tests.

Wood-based PAC (Bench Chemicals, China) was used without further purification. The PAC was sieved through 140, 200, and 800 mesh screens to obtain PAC with the *d*₅₀ of 69.4, 24.2 and 12.7 μm (measured after ultrasonic pretreatment, the particle size distribution of the PAC is illustrated in Fig. S1 of the Supplementary data). Unless otherwise specified, the PAC with the size of 24.2 μm was used. The BET surface area (BET is the Brunauer, Emmett, and Teller method for measuring surface area based on gas adsorption) and the average pore size of the PAC were 1219 m²/g and 2.2 nm, respectively. Stock PAC solution (10 g/L) was prepared and stored in the dark at 4 °C. No calcium was detected in the stock solution. The PAC was dispersed using ultrasound before being used.

2.2. UF experimental setup and fouling tests

All the UF experiments were performed in a membrane test cell (Amicon 8400, Millipore Corp., USA) in dead-end mode at room temperature (21 ± 2 °C). A polyethersulfone (PES) membrane (OM100076, Pall, USA) with a molecular weight cut-off of 100 kDa was employed. The contact angle and the zeta potential of the membrane was 58.2° and –16.9 mV, respectively (measured using

the methods described in a previous study [30]). The membrane was washed with Milli-Q water and the intrinsic membrane resistance was determined before being used. A constant flux of 150 L/(m² h) was maintained using a peristaltic pump during the filtration. The trans-membrane pressure (TMP) was monitored using a pressure transducer and the data were automatically recorded using a computer.

The fouling behavior of HA in the presence of PAC (denoted as “PAC+HA”) and in the absence of PAC (denoted as “PAC-treated HA”), the PAC was removed using a 0.45 μm cellulose ester membrane) was investigated. PAC at a dosage of 50 mg/L was added to the HA solution and the mixture was stirred for 1 h prior to the filtration of PAC+HA and PAC-treated HA. After the pre-adsorption, the further adsorption of HA onto PAC was found very small (< 1.5%) during the filtration of PAC+HA (Fig. S2 of the Supplementary data), therefore the HA concentration in the HA–PAC mixture was approximately constant during the UF experiments.

In the fouling experiments, feed solution with a volume of 350 mL was used. The protocol of fouling experiment is as follows (Fig. S3 of the Supplementary data): (1) solution filtration (300 mL, no stirring), (2) physical cleaning (stirring at a speed of ~500 rpm for 2 min with the residual 50 mL concentrate), (3) filtration with the corresponding background solution (100 mL) to assess the reversibility of the membrane fouling. To determine the HA transmission, the UF permeate was also collected at 20–50, 50–100, 100–150, 150–200, 200–250, 250–300 mL during the filtration.

Membrane fouling index (FI) was utilized for membrane fouling assessment. Total fouling index (TFI) was used to describe the membrane fouling during the filtration cycle, and hydraulically irreversible fouling index (HIFI) was used to quantify the membrane fouling which cannot be recovered by physical cleaning (stirring for 2 min). Under constant flux mode, the FI can be defined as the slope of the line if the TMP/TMP₀ is plotted as a function of the specific volume (*V*_{sp}) [31]. The equation of the calculation of FI is as follows:

$$\text{TMP}/\text{TMP}_0 = 1 + \text{FI} \cdot V_{\text{sp}} \quad (1)$$

where, FI=membrane fouling index, m⁻¹ or m²/L; TMP=trans-membrane pressure, Pa; TMP₀=transmembrane pressure of the virgin membrane, Pa; *V*_{sp}=specific permeate throughput (permeate volume/membrane area), m³/m² or L/m².

A typical plot of *V*_{sp} versus TMP/TMP₀ is shown in (Fig. S3). The TMP/TMP₀ linearly increased as the *V*_{sp} increased. Therefore, the TFI was calculated using linear regression. The HIFI was determined using the TMP of virgin membrane and the TMP of membrane after physical cleaning (two-point method) (Fig. S3) [31].

2.3. Analytical methods

The HA concentration was determined at the wavelength of 254 nm using a UV/vis spectrophotometer (T6, Puxi, China). The particle size of PAC was measured using the Mastersizer 2000 (Malvern, UK). Zeta potential was analyzed using the Zetasizer NS (Malvern, UK). The method to quantify the mass of PAC deposited on the membrane is detailed described in the Supplementary material.

3. Results and discussion

3.1. Combined fouling by HA and PAC

Fig. 1a compares the total fouling and hydraulic irreversible fouling by HA, PAC-treated HA and PAC+HA. HA fouling was

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