



## Ultrafiltration (UF) membrane fouling caused by cyanobacteria: Fouling effects of cells and extracellular organics matter (EOM)

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### ABSTRACT

The ultrafiltration (UF) membrane fouling caused by cyanobacterial cells and extracellular organic matter (EOM) was investigated in this study. Flux decline and reversibility of fouling caused by cyanobacterial cells (including live cells and cell fragments), EOM and their combination were compared. UF fractionation and XAD resin adsorption were employed to characterize the molecular weight (MW) distribution and hydrophilicity/hydrophobicity of EOM, respectively. Attenuated total reflection-Fourier infrared spectroscopy (ATR-FTIR) spectroscopy and scanning electron microscope (SEM) were utilized to characterize the membranes fouled by cyanobacterial cells and EOM. Results showed that cells and cell fragments caused not only reversible but also irreversible fouling of UF membrane. Besides, the membrane fouling caused by the cells and cell fragments was characterized by secondary sharp flux decline which was related to the compression of cake layer. Cyanobacterial EOM also caused serious flux decline due to the deposit of macromolecular organics such as proteins and polysaccharides on membrane. Moreover, EOM could lead to serious irreversible membrane fouling probably due to adhesion of proteins which were characterized by hydrophobicity. Additionally, UF membrane fouling was exacerbated when cells and EOM were filtered together, but no synergistic fouling occurred in this study.

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### 1. Introduction

Reservoirs are the important sources of public water supply in many cities. However, many reservoirs are subjected to eutrophication which usually results in seasonal algae blooms. The presence of algae in source water, especially for cyanobacteria which is commonly found in algae blooms, has become a worldwide problem for drinking water treatment [1]. Cyanobacteria can cause not only undesirable taste and odor but also the release of toxins and organics which may serve as precursors to form disinfection by-products (DBPs) during chlorination [1–3]. Besides, algae-rich water usually requires higher coagulant dosage and backwash frequency of rapid sand filter [4]. These may dramatically increase the operational cost of water treatment process. The traditional water treatment process is ineffective in treating algae-rich water, and thus some chemical oxidation technologies are usually employed to enhance the removal of algae [4]. However, this may cause more cell lysis leading to the release of intracellular organic matter (IOM) and toxins [5]. Ultrafiltration (UF) is increasingly applied in production of drinking water and reclamation of wastewater in recent years [6,7]. The technology is considered as a good choice for the treatment of algae-rich water for water quality concerns, because it can completely remove algal cells

by size exclusion without destroying them [8]. However, it has been proved in many researches that the deposit of algal cells and extracellular organic matter (EOM) on membrane surface can cause severe membrane fouling, leading to remarkable decrease of membrane permeability [9–11].

In recent years, many studies have been carried out to investigate the membrane fouling caused by algae. As a kind of particle, algal cells can deposit on membrane and form thick cake layer which would reduce the permeability. It has been reported that the accumulation of the algal cells on membrane can result in a rapid increase of transmembrane pressure [9]. As a source of organics, algae can release EOM into natural water and cause severe membrane fouling [12,13]. Chiou et al. compared the membrane fouling potentials of three different algal species and found that algae species with more amount of EOM led to lower critical flux and more severe fouling [14]. In their study of membrane fouling caused by natural organic matter (NOM), Lee et al. found that autochthonous NOM, which contained proteins and polysaccharides, caused much more severe membrane fouling than allochthonous NOM [15]. The release of EOM due to shear or preozonation was demonstrated to dramatically exacerbate the membrane fouling during UF or microfiltration (MF) of algae-rich water [16,17]. In addition, it has been reported that particles and organics can cause synergistic membrane fouling when filtered together [18]. Similarly, algae-rich water is also a mixture of particles (cells) and organics (EOM). As the cells and EOM are usually filtered together in the studies conducted with natural algae-rich water and

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lab cultured algae solution, the fouling effects of naked cells have seldom been reported [8–10]. Thus, it is not clear that whether there exist synergetic effect in the fouling caused by the cells together with EOM.

By UF experiments and characterization of separated cyanobacterial cells and EOM solutions, this study tried to get more insights into the membrane fouling caused by cyanobacteria. Moreover, the interactions between cells and EOM during the formation of the membrane fouling are also investigated.

## 2. Materials and methods

### 2.1. Algae culture and EOM extraction

*Microcystis aeruginosa* which was a common species of cyanobacteria was selected for this study. It was purchased from Institute of Hydrobiology, Chinese Academy of Sciences. Axenic cultures were carried out in batch mode in 1 L conical flasks with BG11 medium [19]. The conical flasks were placed in an incubator and cyanobacteria were cultured at the temperature of 25°C with illumination of 5000 lx provided for 14 h every day. Cyanobacteria were harvested at the stationary phase with culture time between 15 and 28 days. Centrifugation was usually used in the extraction of extracellular polymeric substances (EPS) from activated sludge or EOM from algae solution [20]. In this study, algal EOM extraction included two steps: 1) centrifuging the cell suspension at 10,000 g and 4 °C for 15 min to make the EOM released into the supernatant, 2) filtering the supernatant by a 0.45 μm mixed cellulose ether membrane (Taoyuan Co. Ltd, China) to remove the residual cell in the supernatant [12]. A high speed refrigerated centrifuge (GL-21B, Anting, China) was used for EOM extraction. Subsequently, the cyanobacterial cells remained in the bottom of centrifuge tube and those on the surface of 0.45 μm membrane were collected and resuspended with sodium chloride solution (0.6%). Sodium chloride was added to maintain osmotic equilibrium and to prevent the cell breakage [21].

### 2.2. Membrane and UF experiment

Fig. 1 presents the schematic diagram of the UF system. Flat polyethersulfone (PES) UF membranes (OM100076, Pall, USA) with the molecular weight cutoff (MWCO) of 100 kDa and the surface area of  $4.5 \times 10^{-3} \text{ m}^2$  were used in present study. Membrane filtration experiments were carried out in a stirred cell (Amicon 8400, Millipore Corp., USA) and in the mode of downflow dead-end filtration (the stirring paddle inside the stirred cell was not used). UF membrane was placed in the bottom of the stirred cell with its glossy side towards the bulk solution. Nitrogen gas (0.03 MPa) was used to

drive the feed solution through membrane. Permeate flowed into glassware on the electronic balance which was connected to a personal computer. The weighting data were automatically logged every 5 s. New membranes should be carefully rinsed beforehand, because there were plenty of preservatives on them. They were soaked in Milli-Q water for 48 h and then filtered with Milli-Q water under the pressure of 0.1 MPa until the effluent exhibited a comparable level of dissolved organic carbon (DOC) concentration as Milli-Q water.

In the current study, cyanobacterial solution was classified into the cell solution (with EOM) and the resuspended cell solution (without EOM) according to whether EOM was previously separated. UF experiments of resuspended cell solutions with different cell concentration were carried out to investigate the membrane fouling effects of cyanobacterial cells. Four levels of cell concentration were chosen including  $5.0 \times 10^5$ ,  $1.0 \times 10^6$ ,  $2.0 \times 10^6$  and  $4.0 \times 10^6 \text{ cells mL}^{-1}$ . In order to gain more insights into the membrane fouling caused by cyanobacterial cells together with EOM, the fouling caused by cell solution, resuspended cell solution and individual EOM solution were also compared. Concentrations of the cell solutions and the resuspended cell solutions were both  $2.0 \times 10^6 \text{ cells mL}^{-1}$ , and EOM solution was extracted from cyanobacteria solution at this concentration.

In UF experiments, 400 mL of cell or EOM solutions were fed into the stirred cell with 75% of them filtered (i.e., 300 mL). The experiments were carried out under neutral conditions ( $\text{pH } 7.0 \pm 0.1$ ). Prior to each experiment, the pure water flux of membrane was measured by filtering 100 mL milli-Q water and named as  $J_{p(0)}$ . Every UF experiment contained 4 continuous filtration cycles. Each filtration cycle included 3 steps: 1) filtration of 300 mL feed solution, 2) back-wash for 2 min by placing the reverse side of membrane upwards and 3) filtration of 100 mL Milli-Q water (used to analyze the reversibility of fouling). The initial and final flux in the filtration of feed solution were named  $J_{i(n)}$  and  $J_{f(n)}$ , respectively. The average flux in filtration of Milli-Q water after rinse was named  $J_{p(n)}$ . The number  $n$  represented the cycle number. Then, reversible fouling (RF), irreversible fouling (IF) and total fouling (TF) can be calculated as follows according to Jermann et al. [18].

$$IF_n = \frac{J_{p(n-1)} - J_{p(n)}}{J_{p(0)}} \quad (1)$$

$$TF_n = \frac{J_{p(0)} - J_{f(n)}}{J_{p(0)}} \quad (2)$$

$$RF_n = TF_n - IF_n \quad (3)$$

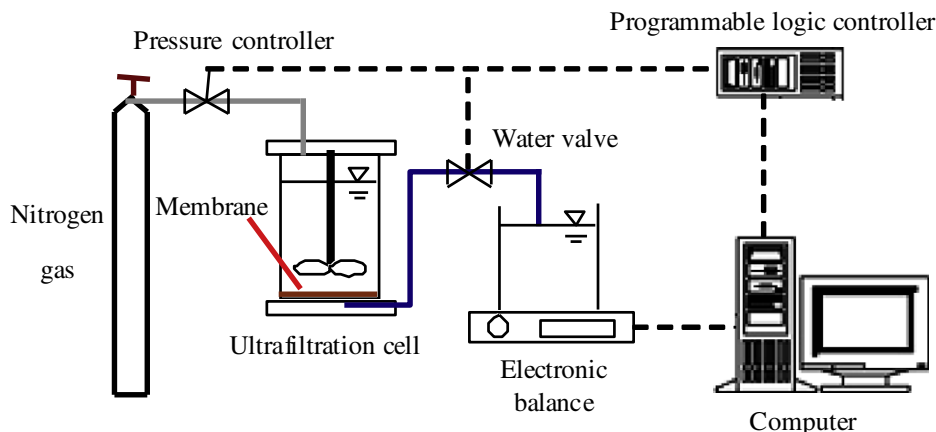


Fig. 1. Schematic diagram of UF system.

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