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Fouling behaviors correlating to water characteristics during the ultrafiltration of micro-polluted water with and without the addition of powdered activated carbon

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HIGHLIGHTS

- PAC pre-treated water had a relatively low FRs and FIs.
- PAC preferentially adsorbed LMWA&BB as well as HS fractions.
- PCA was used to identify fouling behavior associated with water characteristics.
- LMWA&BB imparted great contribution to membrane fouling.
- PAC controlling fouling was attributed to the reduction of LMWA&BB.

GRAPHICAL ABSTRACT
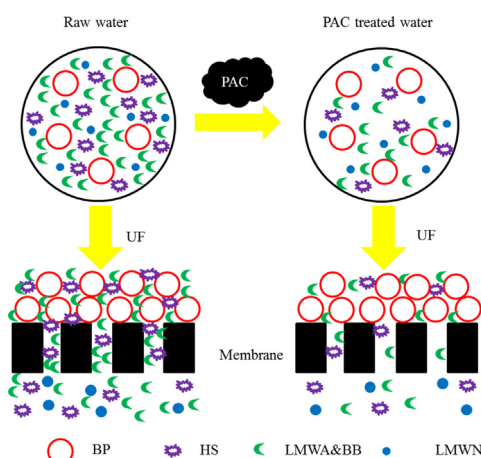
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ABSTRACT

The aim of this paper was to investigate the effects of powdered activated carbon (PAC) pretreatment on the fouling behavior of an ultrafiltration (UF) membrane during micro-polluted water treatment. The results of this study demonstrated that PAC had a positive influence on fouling mitigation. Specifically, PAC-pretreated water had lower fouling resistances (FRs) and fouling indexes (FIs) than raw water. Liquid chromatography combined with peak-fitting analyses of raw water with and without PAC pretreatment suggested that PAC adsorption was much effective in reducing the contents of the low molecular weight acids and building blocks (LMWA&BB) and the humic substance (HS) fractions in raw water. Principal component analysis (PCA) was also used to identify the fouling behaviors associated with the characteristics of the water samples. Significant correlations (r^2) of LMWA&BB with hydraulic reversible resistance (R_{re}), hydraulic irreversible resistance (R_{ir}), total fouling index (TFI) and the hydraulically irreversible index (HIFI) were found (0.7534, 0.8430, 0.6297 and 0.7015, respectively), indicating that LMWA&BB contributed more significantly than biopolymers(BP) and HS to membrane fouling.

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The likely mechanism by which the PAC alleviated fouling was the reduction of the LMWA&BB fraction's presence at the membrane surface and pores. These results suggest that the application of the PAC pretreatment preceding passage through the UF membrane is a promising approach for membrane fouling mitigation.

1. Introduction

Membrane fouling is currently a severe problem that limits the potential use of ultrafiltration (UF) in water purification processes. One of the most hazardous foulants to low-pressure membranes is natural organic matter (NOM) [1]. Some researchers have focused on the molecular weight (MW) distribution and the hydrophobicity/hydrophilicity of the NOM impact on membrane fouling [2–7], while others have devoted efforts to studying related foulants, such as colloidal organics, humic substances (HS), biopolymers (BP), polysaccharides (PS), proteins (PN), etc. [8–12]. To prevent and control membrane fouling, it is necessary to better understand the influence of water characteristics on the mechanism of membrane fouling. Recent improvements in analytical techniques have made it possible to more efficiently analyze NOM. For example, liquid chromatography with organic carbon detection (LC-OCD) is a promising technique for fractionating NOM into LMW neutrals, LMW acids, building blocks (LMW humic-like material), HS and BP [13]. Another relatively new technique, peak-fitting, has proven to be helpful in separating chromatographic peaks into isolated fractions [14–16].

Membrane fouling can generally be divided into two categories: hydraulically reversible and irreversible fouling. In the former, particles are loosely attached to the membrane surface and can be removed through backwashing, but this would lead to an increase in operating costs [17,18]. In the latter, particles are tightly bound to membrane pores, causing a characteristic decline in permeability and requiring elimination by chemical cleaning; this results in both increased operational complexity and reduced membrane lifetime and should therefore be better studied [11,19,20]. Fouling resistances (FRs), which are associated with membrane fouling, can be classified as either a reversible fouling resistance (R_{re}) or an irreversible fouling resistance (R_{ir}), which correspond, respectively, to hydraulically reversible fouling and hydraulically irreversible fouling. Many efforts have been made to investigate the mechanisms of membrane fouling and fouling reversibility [20–24]. However, a clear understanding of the association between the behavior of FRs and fouling components has not yet been achieved.

Several assessment methods, such as the silt density index, the modified fouling index, and the unified membrane-fouling index, have successively been developed to determine the level of membrane fouling [25,26]. Recently, a group of researchers [27] proposed the fouling indexes (FIs), including the total fouling index (TFI) and the hydraulically irreversible index (HIFI), based on the resistance in-series model irrespective of the specific fouling mechanism (e.g., pore constriction, cake filtration, or others). Reports indicated that TFI and HIFI were useful performance indicators for bench, pilot and full-scale filtration systems [28–30]. To the best of our knowledge, the application of FIs in the study of membrane fouling mechanisms coupled with PAC pretreatment has not yet been reported.

To better understand and reduce UF membrane fouling issues, many pre-treatment techniques have been developed and evaluated, including coagulation, pre-adsorption, pre-oxidation and pre-filtration [31,32]. Among these options is PAC adsorption prior to membrane filtration, which is a promising technology for removing organic matter. It is currently unclear whether the addition of PAC may influence membrane fouling behavior. Some studies have testified that PAC could enhance membrane performance. For instance, Gai et al. [33] investigated the effects of PAC on membrane operation and treatment efficiencies and determined that the addition of PAC enhanced filtrate quality and performance efficiency. Li et al. also [34] reported that PAC primarily adsorbed the LMW (<1 kDa) fraction and proved to be effective in mitigating irreversible fouling. Zhang et al. [35] indicated that the structure of the PAC layer was relatively loose and porous, which should positively affect membrane permeability, and Cai et al. [36] determined that a cake layer formed by small PAC ($\sim$1 μm) could act as a profiler and thus facilitate the capture of colloids to reduce total fouling. Conversely, some researchers have published contradictory results. Kang et al. [37] found that the attachment of NOM to PAC particles led to an increase in the resistance of the cake layer. Zhang [38] suggested that NOM binds PAC particles to both one another as well as to the membrane surface, causing the particles to become part of the foulants in the system. Li et al. [39] suggested that the fractions not adsorbed by PAC resulted in higher fouling than the fractions adsorbed by PAC and that PAC pretreatment would thus enhance the removal of organics while simultaneously increasing membrane fouling. These controversial results imply that the influence of PAC addition on membrane performance is still poorly understood and that the mechanism of PAC impact on membrane fouling needs to be more clearly elucidated.

The main objectives of this paper were twofold: first, to identify which fractions in raw water were responsible for membrane fouling and second, to determine whether the addition of PAC into raw water could control or reduce membrane fouling. To address these objectives, hollow-fiber UF of two micro-polluted water samples was continuously carried out for six cycles with periodic backwash, and the water quality characteristics, FRs and FIs were also determined. Additionally, LC-OCD coupled with peak-fitting was used to separate the MW distribution of NOM into isolated fractions. Principal component analysis (PCA) was also performed to evaluate the correlation of FRs and FIs with water characteristics. The results of these experiments were expected to provide useful information for diagnosing and controlling UF membrane fouling during micro-polluted water treatment.

2. Materials and methods

2.1. Source water and PAC

For this study, samples of Sanhaowu lake water (SLW) from the campus of Tongji University, China, as well as Huangpu river water (HRW) from the city of Shanghai, China, were collected to represent surface water polluted by human and industrial wastes.

Two commercially available wood-based carbons were used in these analyses: Carbon M (CM) and Carbon S (CS), supplied by the Fujian Yuanli Active Carbon Co., Ltd and Suzhou Citywater Co., Ltd., respectively. The physical and chemical characteristics (i.e., pore structure, particle size and surface functional groups) of PAC are depicted in Figs. S1–S3.