



Feasibility study on magnetic enhanced flocculation for mitigating membrane fouling



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ABSTRACT

During coagulation/flocculation-membrane filtration (CF-MF) process, membrane fouling was alleviated more significantly through magnetic enhanced flocculation-membrane filtration (MEF-MF) in the presence of ferromagnetic seeds in coagulants. Porous cake layer with flocs of large size was able to alleviate decline rate of membrane flux. Foulant analysis proved that magnetic enhanced flocculation (MEF) pretreatment was more efficient for the reductions of low and mid-molecular weight (MW) organic structures than CF-MF. Biopolymers with high molecular weight were also effectively removed before filtration. Overall, MEF-MF could provide a novel alternative approach to mitigate membrane fouling for surface water treatment.

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Abbreviations: MEF-MF, magnetic enhanced flocculation membrane filtration; CF-MF, coagulation/flocculation membrane filtration; MEF, magnetic enhanced flocculation; TMP, transmembrane pressure; DOM, dissolved organic matter; COD, chemical oxygen demand; SS, suspended solid; BSA, bovine serum albumin; DOC, dissolved organic carbon; ZP, zeta potential; PVDF, polyvinylidene fluoride; FC, ferric chloride; EEM, fluorescence excitation emission matrix; GPC, gel permeation chromatography; MWD, molecular weight distribution; FTIR, Fourier transform infrared spectroscopy; D_f , fractal dimension.

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Introduction

Membrane fouling is the major constraint in the implementation of membrane processes for drinking water treatment [1], as fouling increases operational costs, reduces permeate production and/or increases transmembrane pressure (TMP) [2,3]. Researches showed that pretreatment of surface water was very important to alleviate membrane fouling [4,5]. Although there were evidences in the literature to demonstrate that conventional flocculation could remove colloidal and dissolved organic matter (DOM) during microfiltration [6,7], significant membrane fouling was still observed according to seasonal conditions with pre-flocculation [8].

Enhanced flocculation pretreatment is one of the efficient techniques for mitigating membrane fouling [7]. It was found that non-reactive chemical additives such as zeolite, chitosan, activated carbon and cationic polymers were applied in pretreatment to reduce the concentrations of foulants in raw water, so as to mitigate membrane fouling [9]. Leo et al. [10] reported that embedded zeolite reduced the fouling by humic acid initiated pore blocking. About 80% permeate flux of membrane was maintained during the filtration of humic acid solution. Lee et al. [11] observed that coagulation using chitosan could remove *Chlorella vulgaris* effectively, which was helpful for membrane fouling reduction. Moreover, pretreatment by coagulation with powdered activated carbon before membrane filtration could form larger and more porous flocs than those formed during conventional coagulation [12]. Overall, the effect on membrane fouling mitigation was achieved by the adsorption of non-reactive chemical additives.

Ferromagnetic seeds enhanced flocculation can rapidly separate compounds from mixtures with high efficiency and minimal initial investment by the magnetic characteristic. The application of magnetic seeding flocculation enhances the collision efficiency and collision frequency of colloidal particles, as well as makes colloidal particles to aggregate into larger flocs due to the decrease of colloidal stability [13]. Thus, the magnetic enhanced flocculation was been applied in wastewater treatment to remove foulants [13–17].

It was found that MEF was efficient to remove COD (94%) and SS (71%) in treating mine water with high turbidity [18]. Liu et al. [19] reported that magnetic-coagulation separation could rapidly and effectively remove algae, chlorophyll-a and other foulants from freshwater. Semblante also applied porous micro-sized magnetite to achieve a maximum adsorption of 5.18 mg/g bovine serum albumin (BSA) and successfully inhibited the protein-induced fouling of a commercial polyvinylidene fluoride (PVDF) membrane [20]. In addition, magnetic nanoparticles in inorganic coagulants and their coagulation performances were studied by Zhang [21]. The performance of magnetic poly-aluminum chloride of basicity 2.0 (MPAC12.0) was better than that of PAC on turbidity and DOC removals. Moreover, large, loose and weak flocs were produced by MPAC12.0, which were preferable to recycle magnetic nanoparticles.

To remove COD, SS, and turbidity, which are main constituents of membrane foulants, MEF process was first designed and applied to mitigate membrane fouling in the ultrafiltration for drinking water treatment. In the study, the performance of PVDF hollow fiber membrane with the addition of magnetic enhanced flocculation was examined for treating surface water. The mechanisms of MEF on mitigating membrane were investigated from the perspective of microcosmic morphology. Furthermore,

the characteristics and formation of flocs were investigated to analysis the performance of cake layer and membrane fouling mitigation.

Materials and methods

Characteristics of natural surface water

The raw water was collected from Luan River in Tianjin, China. The characteristics of the surface water are presented in Table 1.

Experimental apparatus and preparation

The bench-scale experimental setup is shown in Fig. 1. The system consists of a coagulant solution tank, a feeding tank, a membrane reactor and a permeate tank. Coagulant solution was pumped into the membrane reactor together with raw water. Colloidal particles were destabilized and furred in flocs with blending. The membrane module submerged in the mixture, and dead-end filtration was carried out for study membrane fouling phenomenon.

Ferromagnetic seeds (Fe_3O_4) (Kermel, Tianjin, China) with sizes from 20 to 60 μm (refer to Fig. 2) was magnetized in a beaker for 5 min by a permanent magnet (40 mT) put under the beaker. The magnetic induction intensity of those ferromagnetic seeds was 0.01 mT. A novel coagulant was prepared by mixing ferromagnetic seeds in ferric chloride (FC) (Kermel, Tianjin, China) solution. The mass rate of ferromagnetic seeds and FC was 1:4. The novel coagulant with ferromagnetic seeds mixed in was a heterogeneous substance and should be shaken well in order to disperse as uniformly as possible. In this study, FC without any ferromagnetic seeds was adopted for contrast experiments.

For testing the effects of coagulants, a virgin PVDF hollow fiber ultrafiltration membrane module (MOTIMO Membrane Technology, Tianjin, China) was used in each experiment. The effective surface area and pore size of each module were 0.04 m^2 and 0.1 μm , respectively. The operating pressure was remained constant over the filtration period. The module was immersed in deionized water for 24 h before use. After each experimental cycle, the fouled module was soaked in sodium hypochlorite (Kermel, Tianjin, China) solution (500 mg/L as free chlorine) for 10 min and rinsed with deionized water. The permeate tank was used to collect the effluent from the membrane reactor. The tank was placed on an electronic counting scale to measure the mass of permeate and the data were recorded by the computer every 10 min.

Table 1
The characteristics of natural surface water.

Parameter	Unit	Value
pH	–	7.25 ± 0.53
UV ₂₅₄ (abs)	cm ⁻¹	0.074 ± 0.008
TOC	mg/L	8.05 ± 1.78
DOC	mg/L	6.65 ± 0.38
TSS	mg/L	3.85 ± 0.45
Zeta potential	mV	–30.5 ± 0.97
Turbidity	NTU	3.64 ± 0.44
Temperature	°C	18 ± 3
Fe ³⁺	mg/L	0.57 ± 0.05

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