Performance of a novel recycling magnetic flocculation membrane filtration process for tetracycline-polluted surface water treatment

Yufei Wang, Hui Jia, Hongwei Zhang, Jie Wang and Wenjin Liu

ABSTRACT

A recycling magnetic flocculation membrane filtration (RMFMF) process integrating circulating coagulation, magnetic enhanced flocculation and membrane filtration was investigated for the treatment of surface water micro-polluted by tetracycline, a typical pharmaceutical and personal care product. A bench-scale experiment was conducted and several water quality parameters including turbidity, ultraviolet absorbance at 254 nm (UV254), total organic carbon and tetracycline concentration were evaluated, taking coagulation membrane filtration and magnetic flocculation membrane filtration processes as reference treatments. The experimental results showed that at the optimum doses of 20 mg·L⁻¹ ferric chloride (FeCl₃), 4 mg·L⁻¹ magnetite (Fe₃O₄) and 6 mg·L⁻¹ reclaimed magnetic flocs in RMFMF processes, removal efficiencies of above evaluated parameters ranged from 55.8% to 92.9%, which performed best. Simultaneously, the largest average particle size of 484.71 μm and the highest fractal dimension of 1.37 of flocs were achieved, which did not only present the best coagulation effect helpful in enhancing the performance of removing multiple contaminants, but also lead to the generation of loose and porous cake layers favouring reduced permeate flux decline and membrane fouling.

Key words | flux decline, fractal dimension, membrane fouling, particle size, tetracycline

INTRODUCTION

As a broad-spectrum antibiotic, tetracycline, a pharmaceutical and personal care product (PPCP), is the most commonly used medicine in the treatment of human beings’ and animals’ diseases and additive in the feeding stuff for promoting animal growth in livestock farming. It is evidenced that the metabolites and degradation products of tetracycline entering into the environment often have lower activities but stronger toxicities compared with their parent antibiotic (Pomati et al. 2004). With the help of advanced analytical instruments, tetracycline has been frequently detected in surface water resources carrying discharges from agricultural effluents and municipal wastewater treatment plants (Kim et al. 2005). The influence of tetracycline on water safety has raised wide concern in the public and regulatory agencies (Auerbach et al. 2007; Baquero et al. 2008; Le-Minh et al. 2010). Thus, the removal
of tetracycline along with regular contaminants should be put on the agenda of surface water treatment.

The current technologies available for treating surface water include some conventional methods such as coagulation, sedimentation, sand filtration and chlorination; and some advanced processes including ozonation, granular activated carbon, photodegradation and photocatalytic degradation (López-Peña1 et al. 2010; Gómez-Pacheco et al. 2011, 2012). In recent years, the whole treatment train has appeared to be too long and too costly for contaminant removal in the micro-polluted surface water. On the contrary, membrane filtration might provide a promising way to make the treatment train shorter by integrating multiple treatment units, for example, just one membrane unit can replace both sedimentation and sand filtration operations. It is effective in removing particles, microorganisms and some organic matter with relatively smaller footprint and lower cost (Iris et al. 2010; Gao et al. 2011).

Coagulation membrane filtration (CMF) process has been widely used as an effective surface water treatment technology (Jang et al. 2010; Yu et al. 2010). However, the application of CMF process is still hindered by membrane fouling which depends largely on the coagulation conditions and leads to the increase of operational cost with the decrease of specific permeate flux. Therefore, it has become a focus to enhance and optimize coagulation for membrane filtration. Circulating coagulation, in which the flocs after settling return to the coagulating basin, can enhance pollutant removal efficiencies, lower coagulant addition, cause floc morphological changes which have a great impact on the subsequent membrane filtration performance, and improve effluent quality (Zhou et al. 2012). Magnetic enhanced flocculation, applying magnetic seeds or magnetic field in flocculation, is another advanced coagulation technology applied in surface water treatment. Plenty researches have found that magnetic enhanced flocculation could effectively remove multiple contaminants and mitigate membrane fouling (Park et al. 2006; Zhang et al. 2012; Semblante et al. 2013). However, the combination of above two enhanced coagulation technologies which can magnify their individual advantages has rarely been reported.

The objective of this study is to develop a novel hybrid process integrating circulating coagulation, magnetic enhanced flocculation and membrane filtration to deal with tetracycline-polluted surface water effectively. A bench-scale experiment was operated. Performances of pollutant removals, floc generation and membrane permeability were evaluated by taking CMF and magnetic flocculation membrane filtration (MFMF) processes as control groups.

### MATERIALS AND METHODS

#### Water matrices

Raw water was taken from the Luan River, an important drinking water source in the city of Tianjin, northern China. The experimental duration (from March to May) was just in the normal-water-quality period of the river, during which those water quality parameters (see Table 1) seemed relatively stable and suitable for experimental operation.

Background tetracycline concentration in the raw water was especially less than 0.01 μg L⁻¹. For preparing tetracycline stock solution, solid tetracycline (purity > 99%, National Institutes for Food and Drug Control, China) was dissolved to 500 μg mL⁻¹ with ultrapure water (18.2 MΩ cm⁻¹) produced by a Milli-Q gradient system (Milli-Q integral 5, Millipore, Bedford, USA). The stock solution was added into the raw water to synthesize objective water with tetracycline concentration of 50 μg L⁻¹.

#### Membrane module

The membrane module applied in the experiment was composed of polyvinylidene fluoride hollow fiber membrane (Tianjin Motimo Membrane Technology Co., Ltd, Tianjin, China). Each membrane fiber possessed rated pore size, inner and outer diameters of 0.1 μm, 0.60 mm and 1.10 mm, respectively. The total effective filtration area was 0.075 m². Before application, virgin membrane module had been soaked for 24 h in DI water so as to wipe off the protective coating on the membrane surface, and then pre-compacted by filtering DI water for 1 h at the constant pressure (0.02 MPa) and temperature (20 ± 1°C) until a persistent permeate flux was achieved. At the

### Table 1 | Characteristics of raw water

<table>
<thead>
<tr>
<th>Water quality parameter</th>
<th>Minimum</th>
<th>Maximum</th>
<th>Average value</th>
<th>Standard deviation</th>
</tr>
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<tbody>
<tr>
<td>Temperature (°C)</td>
<td>12.5</td>
<td>21.0</td>
<td>16.8</td>
<td>3.5</td>
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<tr>
<td>pH</td>
<td>6.54</td>
<td>7.45</td>
<td>7.00</td>
<td>0.38</td>
</tr>
<tr>
<td>TOC (mg·L⁻¹)</td>
<td>5.692</td>
<td>6.683</td>
<td>6.188</td>
<td>0.249</td>
</tr>
<tr>
<td>UV₂₅₄ (cm⁻¹)</td>
<td>0.088</td>
<td>0.098</td>
<td>0.093</td>
<td>0.004</td>
</tr>
<tr>
<td>Turbidity (NTU)</td>
<td>2.34</td>
<td>3.41</td>
<td>2.88</td>
<td>0.44</td>
</tr>
<tr>
<td>NH₃-N (mg·L⁻¹)</td>
<td>1.014</td>
<td>1.416</td>
<td>1.215</td>
<td>0.165</td>
</tr>
<tr>
<td>Chlorophyll a (mg·L⁻²)</td>
<td>3.34</td>
<td>4.95</td>
<td>4.15</td>
<td>0.66</td>
</tr>
</tbody>
</table>
ambient temperature of 20 °C, pure water flux of the membrane module was 80.0 ± 2.3 L·m⁻²·h⁻¹.

Experimental set-up

A submerged membrane filtration unit was operated in the bench scale. The scheme of experimental setup is illustrated in Figure 1. The main reactor was composed of a 24 L polymethyl methacrylate tank equipped with a stirring paddle (Tianjin Branch Wei Instrument Equipment Co., Ltd, Tianjin, China) connected to a churning motor with adjustable speed (Z93-1A, Tianjin Lihua Weiye Technology Development Co. Ltd, Tianjin, China), a U-shaped hollow fiber membrane module assembled vertically and a liquid level controller (JYB-714, Xinling Electrical Co., Ltd, China). The operating pressure of 0.02 MPa was kept constantly in the whole experiment. Raw water in the feed tank was pumped into the main reactor. Magnetic particles (magnetite, Fe₃O₄, analytically pure, Tianjin Kemiou Chemical Reagent Co., Ltd, Tianjin, China) had been magnetized in a beaker for 5 min by a permanent magnet (40 mT) put under the beaker before being mixed in coagulant (ferric chloride, FeCl₃, analytically pure, Tianjin Yingda Rare Chemical Reagent Factory, Tianjin, China) solution, by which the magnetic induction intensity of those particles reached 0.01 mT (tested by a teslameter, HT201, Shanghai Huntoon Magnetic Technology Co., Ltd, China). The mixture was shaken well in order to disperse as uniformly as possible and spiked into the main reactor. With uniform stirring rate, the mixed liquor in the main reactor was well blended. Water yield of submerged membrane module driven by a peristaltic pump was recorded online for every interval of 10 min by electronic balance (CN-SE, Taiwan Sakura Corporation, China). After each run, the used membrane module was soaked in 0.5% sodium hypochlorite solution for cleaning, which allowed nearly 95% recovery of the initial pure water flux. Flocs containing magnetic particles formed in the main reactor were collected through a drain valve at the bottom and stood over night, and then reclaimed by the magnet and redosed to the main reactor with the mixture of coagulant and magnetite for the next run. For contrast experiments, FeCl₃ with neither Fe₃O₄ nor reclaimed magnetic flocs was adopted in CMF process, while FeCl₃ with Fe₃O₄ only was used in MFMF process.

ANALYTICAL METHODS

Floc morphological analysis

It is suggested that the morphological properties of flocs could influence membrane filtration behavior (Wang et al. 2008). This study employed average particle size and fractal dimension of flocs as key parameters in floc morphological analysis to implicate membrane filtration. Immediately after the whole experimental device began to operate stably, flocs were gently sampled at the membrane module height with wide-mouthed pipette, introduced into a quiescent Petri dish containing distilled water and without cover, and settled for about 2 min. After that, images of complete flocs in the dish were captured by a digital charge coupled device camera (MLM3XMP, OPT, China) with a built-in sensor for
progressive-scan of 1/1.8 inch, maximum resolution of 1,392 (horizontal) × 1,040 (vertical), largest frame rate of 8 bit and 15 frames per second. 100 images were taken for each flock and analyzed by the image analysis software Image-Pro Plus 6.0 for calculating flock average particle size and fractal dimension.

**Tetracycline quantitative analysis**

The water samples were analyzed directly, and an external standard method was applied for quantification. Standard stock solution of tetracycline (500 µg/mL) was prepared by dissolving the reference material in ultrapure water. To draw the calibration curve, six standard solutions (10.0, 50.0, 100.0, 150.0, 200.0 and 250.0 µg/L) prepared by diluting the stock solution with ultrapure water were detected by using a Waters 2478 HPLC system (Waters, USA) equipped with a Venusil ABS C8 chromatographic column (5 µm × 4.6 × 250 mm, Bonna-Agela Technologies Inc., China) and a UV detector (NC T6 UV-visible Spectrophotometer, China). Each standard solution was analyzed thrice in series. All solutions were maintained in 4 °C. The method exhibited significant linearity (R² = 0.9995) in the range of 10.0–250.0 µg/L, with a limit of quantitation of 10 µg/L. Supernatant in the main reactor filtrated through 0.45 µm cellulose acetate membranes (Tianjin Heaion Technology Development Co., Ltd, China) pre-rinsed with ultrapure water and effluent after membrane filtration was collected and preserved at 4 °C as well. Each sample was measured thrice in parallel.

**Other analytical methods**

Quality parameters of water samples such as temperature, pH, total organic carbon (TOC), UV₂₅₄ absorbance and turbidity were measured using a bar mercury thermometer, a pH meter (PHS-25, Shanghai INESA Scientific Instrument Co., Ltd, China), a combustion-type organic carbon analyzer (TOC-Vcph analyzer, Shimadzu, Japan), an ultraviolet spectrophotometer (T6, PERSEE, China) and a turbidimeter (2100N, Hach, Japan), respectively. For the raw water, ammonia-nitrogen (NH₃-N) and chlorophyll a were investigated applying the methods of Nessler's reagent photometry and hot ethanol spectrophotometry, respectively, conducted by the ultraviolet spectrophotometer (State Environmental Protection Administration of the P.R.C. 2002).

**RESULTS AND DISCUSSION**

**Removals of turbidity, UV₂₅₄ and TOC**

Figures 2–4 show the removals of turbidity, UV₂₅₄ and TOC in recycling magnetic flocculation membrane filtration (RMFMF) process comparing with those in CMF and MFMF processes, respectively. For CMF, coagulant concentrations of 5, 10, 15, 20, 25 and 30 mg·L⁻¹ were selected. According to Figures 2(a)–4(a), peak removals of turbidity, UV₂₅₄ and TOC were all achieved at FeCl₃ dosage of 20 mg·L⁻¹, conducting 74.3%, 40.4% and 26.4%, respectively. Therefore, to acquire comparable treatment effects, FeCl₃ dosage of 20 mg·L⁻¹ was adopted in both MFMF and RMFMF processes. It can be seen from Figures 2(b)–4(b) that in MFMF, by adding 8 mg·L⁻¹ Fe₃O₄, the maximum removals of turbidity, UV₂₅₄ and TOC reach 92.1%, 67.0% and 54.2%, respectively, which improves a lot compared with those in CMF. Specifically, the removal efficiencies of turbidity, UV₂₅₄ and TOC in MFMF performed better than those in CMF from an overall perspective. As for RMFMF, best removal effects of turbidity, UV₂₅₄ and TOC were 92.9%, 70.8% and 57.9%, respectively, at the doses of 20 mg·L⁻¹ FeCl₃, 4 mg·L⁻¹ Fe₃O₄ and 6 mg·L⁻¹ reclaimed magnetic flocs (see Figures 2(c)–4(c)), superior to those at the optimal conditions of 20 mg·L⁻¹ FeCl₃ and 8 mg·L⁻¹ Fe₃O₄ in MFMF.

Consequently, it is suggested that RMFMF process can not only improve effluent water quality significantly, but also reduce the dosages of coagulant and magnetic particles by involving reclaimed magnetic flocs, which may save the cost of surface water treatment and has certain economic significance and practical value.

**Removal of tetracycline**

Figure 5 indicates the performance of a RMFMF process on tetracycline removal by taking CMF and MFMF processes as control groups. From Figure 5(a), it can be seen that in CMF process, with the increase of coagulant concentration, detected tetracycline concentration dropped off until flattened out, which meant that the removal ratio of tetracycline rose gradually to stabilize. The removal of tetracycline was enhanced by membrane filtration in combination with coagulation. However, even at the optimal coagulant (FeCl₃) dosage of 20 mg·L⁻¹, the maximum removal ratio of tetracycline was only 35.8%, which demonstrated that CMF process was not efficient in tetracycline removal. By contrast, although it was still less desirable, MFMF process raised the removal efficiency of tetracycline...
to nearly 50% at the dosages of 20 mg·L⁻¹ FeCl₃ and 8 mg·L⁻¹ Fe₃O₄. As for RMFMF process, the peak removal ratio of tetracycline came to 55.8% at the optimal dose of 20 mg·L⁻¹ FeCl₃ with 4 mg·L⁻¹ Fe₃O₄ and 6 mg·L⁻¹ reclaimed magnetic flocs involved (see Figure 5(b)).

It is noteworthy that at the coagulation stage of RMFMF process tetracycline removal ratio was enhanced to 42.6% comparing with 27.6% and 39.2% at the coagulation stages of CMF and MFMF processes, respectively. Some conventional treatment processes like coagulation, sedimentation and sand filtration were not efficient in the removal of PPCPs (Basile et al. 2011). Recycling magnetic flocculation combining with membrane filtration can effectively improve the removal ratio of tetracycline.

**Characteristics of flocs**

Figure 6 shows the variations of average particle size of flocs formed in different coagulation conditions. As shown in Figure 6(a), the average particle size of flocs increased gradually from 300 μm to approximate 400 μm with the growth of FeCl₃ dosage in CMF process. The unremarkable variations of average particle size of flocs in FeCl₃ concentrations over 20 mg·L⁻¹ suggested that an excessive amount of FeCl₃ could not lead to the generation of flocs that had much larger average particle size. It can be found that average particle size range of flocs in MFMF process developed a lot compared with that in CMF process with the increase dose of magnetic floc under the constant FeCl₃ concentration of 20 mg·L⁻¹, from 382.54 μm to 440.23 μm. On the premise of 20 mg·L⁻¹ FeCl₃, five selected Fe₃O₄ concentrations were adopted in the operation of RMFMF process, and with each of them three reclaimed magnetic floc concentrations were given (see Figure 6(b)). In RMFMF process, when reclaimed magnetic floc concentration was set to 3 mg·L⁻¹, average particle size of flocs increased with the rise of Fe₃O₄ concentration, while the decreases in the conditions of 6 and 8 mg·L⁻¹ Fe₃O₄ with
6 and 9 mg·L\(^{-1}\) reclaimed magnetic flocs, respectively, indicated that excessive reclaimed magnetic floc concentration did not contribute to larger average particle size. Among the above three processes, the maximum average particle size of flocs, 484.71 μm, appeared when Fe\(_3\)O\(_4\) and reclaimed magnetic floc concentration were set at 4 mg·L\(^{-1}\) and 6 mg·L\(^{-1}\), respectively, in RMFMF process.

Because of the Lorentz force between magnetized Fe\(_3\)O\(_4\) particles that influenced their collision effects and interaction modes, which might produce more complexations so as to change the flocculation effect, magnetic flocculation could enlarge the average particle size of flocs to some extent (Yiacoumi et al. 1996). Yet higher Fe\(_3\)O\(_4\) concentration did not result in larger average particle size of flocs, which illustrated that there existed a suitable concentration ratio of Fe\(_3\)O\(_4\) and FeCl\(_3\), and the aggregation effect caused by higher Fe\(_3\)O\(_4\) concentration was adverse to effective particle collision. With the appropriate concentration ratio of Fe\(_3\)O\(_4\) and reclaimed magnetic flocs, RMF could increase floc particle size effectively. Magnetic floc recycle and re-addition led to the appearance of destabilized colloids and small floc fragments which could be deposited and adsorbed, enhancing the collisions and aggregations of particles in water so as to form even larger flocs after the breakages of original ones. In addition, the Lorentz force produced by re-magnetized reclaimed magnetic flocs could force the suspended particles in water to generate larger flocs as well. However, it was worth pointing out that the performance of flocculation did not conduct well as the concentration of reclaimed magnetic flocs crossed a particular threshold, which might result in overmuch flocs that created constraints on the collisions and aggregations of flocs and then on the growth of particle size.

Small fractal dimensions of flocs always bring about loose floc structures and lower floc densities (Wang et al. 2008), whereas large fractal dimensions of flocs lead to compact and dense floc structures and better coagulation effects. In CMF process, the fractal dimensions of flocs varied in the

**Figure 1** | \(\text{UV}_{254}\) variations in CMF, MFMF and RMFMF processes: (a) in CMF process; (b) in MFMF process with 20 mg·L\(^{-1}\) FeCl\(_3\); (c) in RMFMF process with 20 mg·L\(^{-1}\) FeCl\(_3\) (R – Raw water; S – Supernatant; F – Filtrate; S\(_3\), S\(_6\) and S\(_9\) – Supernatants by 3, 6 and 9 mg·L\(^{-1}\) reclaimed magnetic flocs, respectively; F\(_3\), F\(_6\) and F\(_9\) – Filtrates by 3, 6 and 9 mg·L\(^{-1}\) reclaimed magnetic flocs, respectively).
range of 1.14–1.20 with the changes of FeCl₃ concentrations. When the coagulant concentration was 20 mg·L⁻¹, maximum fractal dimension of flocs 1.20 was achieved. In MFMF process, at the optimal dosages of 20 mg·L⁻¹ FeCl₃ and 8 mg·L⁻¹ Fe₃O₄, maximum fractal dimension of flocs 1.34 was achieved, 0.14 higher than the
maximum value obtained in CMF process (see Figure 7(a)). As shown in Figure 7(b), in RMFMF process, under the addition of $20 \text{ mg·L}^{-1} \text{FeCl}_3$ and reclaimed magnetic flocs at different concentrations, floc fractal dimension changed in the range of 1.15–1.19 without adding Fe$_3$O$_4$, in the range of 1.25–1.29 with adding $2 \text{ mg·L}^{-1} \text{Fe}_3\text{O}_4$, in the range of 1.32–1.37 with adding $4 \text{ mg·L}^{-1} \text{Fe}_3\text{O}_4$, in the range of 1.33–1.35 with adding $6 \text{ mg·L}^{-1} \text{Fe}_3\text{O}_4$, and in the range of 1.34–1.36 with adding $8 \text{ mg·L}^{-1} \text{Fe}_3\text{O}_4$. Thus, in RMFMF process, through recycling magnetic flocs, newborn flocs obtained not only larger average particle sizes but also higher fractal dimensions.

The smaller fractal dimension floc possesses, the higher porosity it has (Chakraborti et al. 2003). Small flocs of small fractal dimensions are aggregated in random collisions to form large ones that have open and porous structures and are vulnerable to fluid shear stress (Wang et al. 2011). RMF could further promote collisions and aggregations of those flocs possessing contact activity to form flocs of higher densities. However, when the concentration of reclaimed magnetic flocs exceeded some suitable value, floc particles in water got oversaturated, and probabilities of effective collisions between flocs diminished dramatically, which had great impact on floc morphological characteristics. That is why floc fractal dimension tends to become stable with the increase of reclaimed magnetic particle concentration in Figure 7(b). From Figures 6 and 7 it is noteworthy that RMFMF process can enlarge not only average particle sizes but also floc fractal dimensions.
sizes of flocs but also their fractal dimensions. In other words, RMFMF process ameliorates not only floc resistivity to fluid shear stress but also floc properties of coagulation and sedimentation.

Decline of permeate flux

The time-varying changes in permeate flux during membrane filtration in CMF, MFMF and RMFMF processes are shown in Figures 8–10, respectively. From those figures it can be found that the change trend and decline degree of permeate flux are brought into correspondence with those of floc average particle size and fractal dimension, which to some extent confirms the conclusion drawn by Barbot et al. that the formation of large flocs with a strong structure is the optimal pre-coagulation condition for coagulation/membrane filtration process (Barbot et al. 2008). From Figure 8 we can see that, in CMF process, the slowest decline in permeate flux appeared at coagulant dose of 20 mg·L⁻¹. In MFMF process, the drop of permeate flux was alleviated for magnetic filtration with increased Fe₃O₄ dose until touching the bottom with Fe₃O₄ dose of 8 mg·L⁻¹ over which the decline degree of permeate flux intensified, that is, membrane fouling was aggravated. But the decline of permeate flux in MFMP process still performed milder than that in CMF process (see Figure 9). As shown in Figure 10, the most drastic permeate flux decline occurred in the case of absent Fe₃O₄ for RMF. With the adding of Fe₃O₄, the decline of permeate flux began to lighten. It is noticeable that with Fe₃O₄ and reclaimed magnetic floc doses of 4 and 6 mg·L⁻¹, respectively, the decrease of permeate flux was slower than that in any other conditions and the membrane fouling was the slightest. In RMFMF process, although severer permeate flux decline happened in the case of Fe₃O₄ overdosing (6–8 mg·L⁻¹) in comparison with the optimum Fe₃O₄ dose (4 mg·L⁻¹), which meant the aggravated membrane fouling, the permeate flux decline in RMFMF process was still generally slighter than those in CMF and MFMF processes.

Floc adsorbability mainly depends on floc morphology as flocs of large size always have strong adsorbability (Waite et al. 1999). Compared with CMF and MFMF processes, RMFMF process could not only reinforce the removal of organic matters, which eased the burden of cake layer, but also improved the morphologic features of flocs such as particle size and fractal dimension, which strengthened membrane permeability. In RMFMF process, with the presence of reclaimed magnetic flocs, the coupling of FeCl₃ and Fe₃O₄ performed better and produced good-size-distributed flocs which contributed to the generation of porous and loose cake layer (Amjad et al. 2013). Moreover, because of the flocs possessing relatively larger specific surface area, their opportunities of adsorbing tiny particles increased, which could reduce those particles’ probability of blocking membrane pores. In consequence, comparing with CMF and MFMF processes, RMFMF process could mitigate membrane fouling preferably.

CONCLUSIONS AND PROSPECTS

RMFMF process, a hybrid process combining circulating coagulation, magnetic enhanced flocculation and membrane filtration, has been demonstrated to have better capability to deal with the synthetic surface water.
containing tetracycline in comparison with CMF and MFMF processes. At the optimal doses of 20 mg·L⁻¹ FeCl₃, 4 mg·L⁻¹ Fe₃O₄ and 6 mg·L⁻¹ reclaimed magnetic flocs, several contaminants could be removed effectively, including turbidity (92.9%), UV₂₅₄ (70.8%), TOC (57.9%) and tetracycline (55.8%), and floc morphological characteristics showed up as larger particle size (484.71 μm) and higher fractal dimension (1.37). When flocs with the above characteristics accumulated on membrane surface under a given pressure, permeate flux was maintained and membrane fouling was alleviated.

Despite of the superiority of RMFMF process over both CMF and MFMF processes, its efficiency was still unsatisfactory since the removals of above water pollutants were kept at relatively low levels. Aiming at improving the RMFMF technology, several measures, such as employing different coagulant strategies (e.g. other species or dosages), changing coagulation conditions (e.g. pH or stirring rate) and coupling with other pretreatments (e.g. powdered activated carbon or pre-ozonation), can be implemented in the coming work. Nevertheless, it is believed that the RMFMF hybrid process developed in this paper provides some insights into the upgrade of conventional surface water treatment processes.

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