Removal characteristics of microplastics by Fe-based coagulants during drinking water treatment

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A B S T R A C T

Microplastics have caused great concern worldwide recently due to their ubiquitous presence within the marine environment. Up to now, most attention has been paid to their sources, distributions, measurement methods, and especially their eco-toxicological effects. With microplastics being increasingly detected in freshwater, it is urgently necessary to evaluate their behaviors during coagulation and ultrafiltration (UF) processes. Herein, the removal behavior of polyethylene (PE), which is easily suspended in water and is the main component of microplastics, was investigated with commonly used Fe-based salts. Results showed that although higher removal efficiency was induced for smaller PE particles, low PE removal efficiency (below 15%) was observed using the traditional coagulation process, and was little influenced by water characteristics. In comparison to solution pH, PAM addition played a more important role in increasing the removal efficiency, especially anionic PAM at high dosage (with efficiency up to 90.9%). The main reason was ascribed to the dense floc formation and high adsorption ability because of the positively charged Fe-based flocs under neutral conditions. For ultrafiltration, although PE particles could be completely rejected, slight membrane fouling was caused owing to their large particle size. The membrane flux decreased after coagulation; however, the membrane fouling was less severe than that induced by flocs alone due to the heterogeneous nature of the cake layer caused by PE, even at high dosages of Fe-based salts. Based on the behavior exhibited during coagulation and ultrafiltration, we believe these findings will have potential application in drinking water treatment.

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Introduction

Plastics have been widely applied worldwide due to the rapid development of industrialization. Previous studies have reported that the global annual production of plastics is around 280 million tons, with the majority for disposable use (Rilling, 2012; Rocha-Santos and Duarte, 2015; Anderson et al., 2016). However, tiny fragments or particles are gradually formed by plastic debris with mechanical action, biodegradation, and photo-oxidation over a long period of time. When the particle size is below 5 mm, they are often defined as microplastics (Wright et al., 2013; Rocha-Santos and Duarte, 2015). Up to now, most microplastics have been detected in the ocean, and cause as high as 13 billion dollars of economic loss annually to marine ecosystems (Raynaud, 2014). As a result, microplastics have been widely investigated as a new kind of emerging pollutant (Cole et al., 2011; Rocha-Santos and Duarte, 2015).

It has been demonstrated that the presence of microplastics can cause various environmental and health problems, which include the following: (1) microplastics are easily mistaken for food, resulting in blockage of the eating organs of aquatic organisms (Nor and Obbard, 2014); (2) light is hindered by microplastics suspended or floating in the ocean due to their low density, decreasing the utilization efficiency of light for aquatic organisms (Li et al., 2018); (3) microplastics are easily used as a carrier for persistent organic pollutants (POPs) because of their large specific surface area and strong hydrophobicity (Ashton et al., 2010; Koelmans et al., 2016). It has been reported that POPs (i.e., polychlorinated biphenyls, polycyclic aromatic hydrocarbons, and organo-chlorine pesticides) on the surface of microplastics were detected in many coastal zones, such as along the USA, China, UK, Japan, etc. (Mizukawa et al., 2013); and (4) heavy metals, even nanoparticles, are also easily adsorbed onto the surface of microplastics, including Fe, Al, Zn, Cu, Pb, and TiO$_2$ (Ashton et al., 2010; Fries et al., 2013).

Although microplastics have gradually attracted global attention, most related research has been focused on marine environments, especially in the coastal areas with human activities (Cole et al., 2011; Oberbeckmann et al., 2015). Current research studies are mainly focused on their sources (Browne et al., 2011; Dris et al., 2016), distributions (Desforges et al., 2014; Klein et al., 2015), abundances (Cole et al., 2011; Eriksen et al., 2013), separation and identification methods (Erker-Medrano et al., 2015; Tagg et al., 2015), adsorption and desorption mechanisms (Wright et al., 2013; Brennecke et al., 2016), and eco-toxicological effects (Avio et al., 2015; Ziccardi et al., 2016). With microplastics gradually being detected in freshwaters (i.e., rivers, lakes), however, little attention has been paid to their corresponding behaviors, which are relevant to human health because of the effect on drinking water treatment to some extent. It has been reported that the abundance of microplastics in the Tamar River estuary of England is around 0.03 items/m$^3$ (Sadri and Thompson, 2014), while it is 0.26 items/m$^3$ in the Goiana River estuary in Brazil (Lima et al., 2014). In comparison with other rivers, the abundance of microplastics is much higher in those of China. Previous studies have reported that the abundance of microplastics in the estuary of the Yangtse River is as high as 4137.3 ± 2461.5 items/m$^3$ (Zhao et al., 2014), while it is 3807.4 × 10$^3$ items per square kilometer in the Three Gorges Reservoir (Zhang et al., 2015).

With the gradual detection and high abundance of microplastics in freshwaters, it is urgently necessary to investigate their behaviors during coagulation and ultrafiltration (UF) processes, which are the main water treatment technologies in current water plants at present and even in the next few decades (Xia et al., 2007; Shannon et al., 2008; Leiknes 2009; Park et al., 2017). Although both Al-based salts and Fe-based salts are widely applied during coagulation (Zhou et al., 2012; Rakruam and Wattanachira, 2014; Chorgbe et al., 2017; Wang et al., 2018), the use of Al-based coagulants may result in residual aluminum in the treated water, which potentially causes adverse human health effects (e.g., residual aluminum in drinking water is suspected to be neurotoxic) (Kimura et al., 2013). In addition, the effect of sweep flocculation plays an important in removing microplastics, while the settling ability of Fe-based flocs is much better than that of Al-based flocs due to the density. As a result, Fe-based salts were chosen in this work. For microplastics, it has been reported that polyethylene (PE) is the main microplastic detected in water (Andrady, 2011). Additionally, the density of PE (0.92-0.97 g/cm$^3$) is very close to that of water, resulting in more severe potential hazards to the water treatment process due to its suspension and flotation properties in water.

Herein, this work systematically investigated the performance of traditional water treatment in the presence of PE. It aims to: (1) understand the removal efficiency of PE by current drinking water treatment processes; (2) explore the corresponding influence factors related to PE removal behavior, such as coagulant dosage, solution pH, polyacrylamide (PAM) addition for the enhancement of coagulation performance (Aguilar et al., 2003; Aboulhassan et al., 2006; Lee and Westerhoff, 2006), etc.; and (3) investigate UF membrane behavior in the presence of PE with/without coagulation.

1. Materials and methods

1.1. Materials

All chemical reagents used were of analytical grade and were obtained from Sinopharm Chemical Reagent Co., Ltd. (China), including FeCl$_3$·6H$_2$O, NaHCO$_3$, HCl, NaOH, PAM, and kaolin. PE plastics were purchased from Yanshan Xintianze Chemical Co., Ltd. (Beijing) and were rinsed with 1 mol/L HCl before experiments to remove residues. Natural organic matter (NOM), represented by Humic acid (HA, Sigma-Aldrich, USA), was dissolved at a concentration of 2 g/L (Yuan and Zydne, 1999; Zularisam et al., 2006). All stock solutions were prepared in deionized water (DI, Millipore Milli-Q, USA) and stored in the dark at 4°C.

1.2. Coagulation with jar tests

A 0.5 L beaker with a stirrer in the bottom was used for comparative coagulation tests. To investigate the removal
behavior, PE particles mechanically disintegrated from large ones were initially sieved with different meshes, with particle diameters \(d<0.5\) mm, \(0.5<d<1\) mm, \(1<d<2\) mm, \(2<d<5\) mm, respectively. During coagulation, the solution pH was adjusted with 0.1 mol/L NaHCO\(_3\) as a buffer, and the pH of the final solution was maintained at 6.7, and 8 by prior addition of a predetermined amount of 0.1 mol/L NaOH/HCl (Ma et al., 2014). The fast mixing speed was maintained for 1 min at 300 r/min (278 S\(^{-1}\)), and then decreased to 100 r/min (18.9 S\(^{-1}\)) for 14 min, followed by 30 min sedimentation. A laser particle size analyzer (Mastersizer 2000, Malvern, UK) was used to measure the dynamic size of flocs in the absence of PE. The floc size was analyzed every 0.5 min and automatically recorded by a computer (Jarvis et al., 2005).

1.3. Ultrafiltration experiments

For UF experiments, flat sheet polyvinylidene fluoride UF membranes (100 kDa, Motimo, China) were used, which was conducted in a Millipore stirred UF cell (8400, USA) (Appendix A Fig. S1). Before experiments, all membranes were immersed in DI water for 24 hr to remove impurities and were operated at 0.15 MPa (nitrogen gas) with 300 mL DI water. During the tests, the pressure was maintained at 0.1 MPa. The normalized specific flux \(J_o\) was used to represent the membrane performance over time, where \(J_o\) represents the initial membrane flux.

During UF experiments, FeCl\(_3\)·6H\(_2\)O stock solutions were diluted with 300 mL feed water in the stirred UF cell, along with 0.1 mol/L NaHCO\(_3\) to give the test solution. During the tests, a predetermined amount of 0.1 mol/L NaOH was pre-added to maintain the final solution pH at 7. The rapid mixing speed at 300 r/min lasted for 1 min while the slow mixing speed at 100 r/min lasted for 14 min to allow floc growth (Ma et al., 2014).

1.4. Characteristics of microplastics and Fe-based flocs

To investigate the morphologies of microplastics and flocs, the samples were taken from below the suspension surface with a hollow glass tube during the test. An optical microscope equipped with a CCD camera (BX51, Olympus, Japan) was used to capture the images. The zeta potentials of the Fe-based flocs before and after adsorption were measured by a nanoparticle sizing and zeta potential analyzer (Delsa Nano C, Beckman Coulter, Ltd., USA). The concentration of iron was measured by inductively coupled plasma mass spectrometry (iCap-Q, Thermo Scientific, USA).

1.5. Measurement method for microplastics

Many methods have been investigated for the detection of microplastics in water treatment. However, no satisfactory method has been established due to the complexity of water quality (Li et al., 2018). In comparison to the currently used methods (i.e., microscopic method, infrared method), the weighing method is much more accurate and was chosen in this work. The specific procedure was as follows: First, microplastics were dried in an air-drying oven (DHG9070, Shanghai Yiheng Scientific Instrument Co., Ltd., China) at 60°C for 12 hr to dehydrate them. Second, a known amount of microplastics was added into the beaker (\(W_{\text{total}}\)), weighed with a precision balance having a minimum range of 1.0 × 10\(^{-4}\) g (Quintix124-1CN, Sartorius, Germany). After sedimentation for 30 min, the supernatant was carefully taken with a 25 mL syringe. Third, the deposits in the supernatant were immersed in 1 mol/L HCl for 1 hr to remove flocs and were then filtered with 0.45 \(\mu\)m membranes. Finally, the microplastics on the membrane surface were carefully scraped down and dried in the air-drying oven at 60°C for 12 hr, followed by weighing after cooling to room temperature (\(W_{\text{dried}}\)). Therefore, the removal efficiency of PE (%) can be expressed as:

\[
\text{Removal efficiency of PE} = \left[\frac{W_{\text{total}} - W_{\text{dried}}}{W_{\text{total}}}\right] \times 100%.
\]

10 mmol/L NaCl was used as the background ionic strength and required turbidity (Turbidimeter, HACH, 2100Q, USA) was obtained with the addition of kaolin. All experiments were conducted three times.

2. Results

2.1. Effects of Fe-based coagulant dosage on PE removal efficiency

The removal performance of PE was investigated with different dosages of FeCl\(_3\)·6H\(_2\)O at pH 7.0. As seen from Fig. 1, the removal efficiency increased with increasing dosage of FeCl\(_3\)·6H\(_2\)O, and it remained stable when the dosage of FeCl\(_3\)·6H\(_2\)O was higher than 1 mmol/L. The smaller the particle size of PE, the higher the removal efficiency was. However, the total PE removal efficiency was low during coagulation, even in the presence of high dosages of FeCl\(_3\)·6H\(_2\)O. The removal efficiencies of small-particle-size PE (\(d<0.5\) mm) were 3.43% ± 0.96%, 6.71% ± 1.26%, 8.24% ± 1.22%, 11.72% ± 0.96%, 13.27% ± 2.19%, and 12.65% ± 1.09% in the presence of 0.1, 0.2, 0.5, 1, 2, and 5 mmol/L FeCl\(_3\)·6H\(_2\)O, whereas those for large PE particles (2 <\(d<5\) mm) were only 0.37% ± 0.16%, 1.02% ± 0.33%, 2.58% ± 0.81%, 2.13% ± 0.74%, 2.29% ± 0.32%, and 2.81% ± 0.69%, respectively.

Fig. 1 – Removal efficiency of PE with different dosages of FeCl\(_3\)·6H\(_2\)O at pH 7.0. Weight of PE particles: 0.1 g.
was lower than 0.36 mmol/L (20 mg/L) during traditional drinking water treatment (Volk et al., 2000; Qiao et al., 2008; Sillanpää et al., 2018). To further investigate the corresponding removal behaviors in detail, the dosages of 0.2 mmol/L (11.2 mg/L) and 2 mmol/L (112 mg/L) FeCl₃·6H₂O were chosen, although the removal efficiencies of different particle sizes of PE were low.

2.2. Effects of solution pH on PE removal efficiency

Actual water conditions are complex, including various kinds of ions, NOMs, and particles. However, the removal efficiency of PE was influenced little by ionic strength (Appendix A Fig. S2), HA (Appendix A Fig. S3) or turbidity during coagulation (Appendix A Fig. S4). It has been demonstrated that solution pH plays an important role in the coagulation performance because of the effect on floc properties (Duan and Gregroy, 2003; Wang et al., 2013). Thus, the removal efficiencies of PE were investigated at pH 6.0, 7.0, and 8.0 (Fig. 2). Results showed that the removal efficiency of PE was also influenced little at the low dosage of FeCl₃·6H₂O (0.2 mmol/L). However, the removal efficiency of PE increased under the high dosage of FeCl₃·6H₂O (2 mmol/L), and the corresponding removal efficiency also increased with increasing solution pH.

In addition, when the particle size is smaller, the higher the PE removal efficiency is, especially for the high dosage of Fe-based salts (Fig. 2b). For 0.2 mmol/L FeCl₃·6H₂O, the removal efficiency of small-particle-size PE (d < 0.5 mm) was 6.03% ± 0.61%, 6.71% ± 1.26%, and 6.67% ± 1.32% at pH 6.0, 7.0, and 8.0, respectively. For 2.0 mmol/L FeCl₃·6H₂O, however, the removal efficiency of small-particle-size PE (d < 0.5 mm) was 11.56% ± 0.98%, 13.27% ± 2.19%, and 17.23% ± 1.72% at pH 6.0, 7.0, and 8.0, respectively.

2.3. Effects of PAM addition on PE removal efficiency

On one hand, the removal efficiency of PE particles was low. On the other hand, plenty of previous studies have demonstrated the by PAM addition (Aguilar et al., 2003; Aboulhassan et al., 2006; Lee and Westerhoff, 2006). Therefore, the removal efficiency of PE was investigated in the presence of cationic PAM and anionic PAM (Fig. 3).

As seen from Fig. 3a and b, both cationic PAM and anionic PAM had little influence on the removal behavior of PE with the low dosage of FeCl₃·6H₂O (0.2 mmol/L) and PAM. It was clearly shown that the removal efficiency of PE increased little, even in the presence of 15 mg/L PAM. However, the removal efficiency of PE significantly increased in the presence of the high dosage of FeCl₃·6H₂O (2 mmol/L) with PAM (Fig. 3c and d). In comparison to cationic PAM, anionic PAM performed much better in terms of the PE removal efficiency. Additionally, the smaller the particle size of PE, the higher the removal efficiency. As seen from Fig. 3d, the removal efficiency of small-particle-size PE (d < 0.5 mm) significantly increased from 13.27% ± 2.19% to 89.23% ± 3.22%, 87.66% ± 1.89%, 85.21% ± 2.12%, 89.32% ± 3.96%, and 90.91% ± 1.01% with 3, 6, 9, 12, 15 mg/L anionic PAM, respectively.

2.4. UF membrane performance with PE

Owing to the excellent performance of UF membranes and their wide application in water treatment (Leiknes, 2009; Gao et al., 2011), the membrane performance in the presence of PE with/without coagulation was further investigated (Fig. 4). In comparison to the average membrane pore diameter (30 nm, manufacturer provided), PE particles can be completely rejected due to their larger particle size (micrometer scale). It was shown that PE particles alone had little influence on membrane fouling and that the J/J₀ was higher than 0.8 after running for 300 s (Fig. 4a, the same below).

For coagulation, flocs were easily formed because of hydrolysis (Leiknes, 2009). Owing to the large particle size of flocs, a loose cake layer was easily formed on the membrane surface and only slight membrane fouling was induced. The higher the dosage of FeCl₃·6H₂O, the more severe the UF membrane fouling after coagulation (Fig. 4b and c). The specific UF membrane flux decreased to 0.69 and 0.55 in the presence of 0.2 mmol/L and 2 mmol/L FeCl₃·6H₂O, respectively. However, it was interesting that the membrane fouling was gradually alleviated after coagulation with PE. The larger the particle size of PE, the less severe the membrane fouling was. The specific membrane flux decreased to 0.78 and 0.70 in the presence of large-particle-size PE (2 < d < 5 mm) after
Fig. 3 – Removal efficiency of PE with different particle sizes at pH 7.0: (a) 0.2 mmol/L FeCl₃·6H₂O and cationic PAM; (b) 0.2 mmol/L FeCl₃·6H₂O and anionic PAM; (c) 2 mmol/L FeCl₃·6H₂O and cationic PAM; (d) 2 mmol/L FeCl₃·6H₂O and anionic PAM. Weight of PE particles: 0.1 g.

Fig. 4 – UF membrane fouling at pH 7.0 (a) without coagulation; (b) with 0.2 mmol/L FeCl₃·6H₂O; and (c) with 2 mmol/L FeCl₃·6H₂O. Weight of PE particles: 0.2 g.
coagulation with 0.2 mmol/L and 2 mmol/L FeCl₃·6H₂O, respectively.

3. Discussion

At present, coagulation is still the main technology in current drinking water treatment due to its excellent ability to remove particles through adsorption (Leiknes, 2009). However, the PE removal efficiency was low during coagulation even with a high dosage of FeCl₃·6H₂O (Fig. 1). The potential reasons were the following: First, the density of PE was low (0.92–0.97 g/m³), resulting in difficulty of settling during coagulation. Second, the average size (μm, which was relatively smaller than that of PE particles (millimeter scale). Therefore, PE particles were not easily adsorbed/trapped by Fe-based flocs even with a high dosage of coagulants, especially the large PE particles.

Owing to the small influence of water conditions (i.e., ionic strength, NOM, turbidity) on floc properties, the removal efficiency of PE particles was influenced little (Appendix A Figs. S2, S3, and S4). However, pH played an important role in floc characteristics because of the strong hydrolysis of the coagulant (Duan and Gregroy, 2003; Ma et al., 2014; Sillanpää et al., 2018). As seen from Fig. 5a, the size of Fe-based flocs gradually increased with increasing solution pH. The specific d₅₀ of the Fe-based floc size was 433.8 ± 22.3, 630.6 ± 28.4 and 761.9 ± 31.7 μm at pH 6.0, 7.0, and 8.0, respectively. In addition, the zeta potential of Fe-based flocs was 1.91 mV ± 0.34 mV at pH 6.0, while it was 0.32 ± 0.13 mV and −3.43 ± 1.18 mV at pH 7.0 and 8.0, respectively. Furthermore, Fig. 5b shows that the zeta potential of FE particles was always negative (Appendix A Table S1, which was lower than 40 mV/L)

In recent years, the UF membrane has been widely applied along with coagulation as a pretreatment during drinking water treatment, of which the effluent is excellent (Xia et al., 2007; Shannon et al., 2008; Leiknes, 2009; Park et al., 2017). Owing to the larger size of PE particles (millimeter scale) compared to the membrane pore diameter (30 nm), a cake layer was easily formed on the membrane surface. The larger the PE particles, the relatively looser the cake layer was and the less severe the membrane fouling was (Fig. 4a). After coagulation, a loose Fe-based floc cake layer was also formed on the membrane surface due to the large floc size. However, the porosity of the cake layer increased due to the influence of PE particles. The larger the PE particles, the larger the porosity of the cake layer was. Thus, less severe membrane fouling was induced, even in the presence of a high dosage of FeCl₃·6H₂O (Fig. 4).

In view of the above-mentioned observations, microplastics alone could be suspended or floated in water because of their similar density to water. Although the smaller the particle size of microplastics, the higher the removal efficiency was, a small amount of microplastics was removed, even with the high coagulant dosage. In comparison with solution pH, anionic PAM played a more important role in removing microplastics due to its large density and opposite charge compared to Fe-based flocs under neutral conditions. However, it should be noticed that the running costs will increase with high coagulant dosage and PAM. For the UF process, slight membrane fouling was induced by microplastics and cake layer formation was the dominant mechanism, even in the presence of high dosage of FeCl₃·6H₂O (Fig. 4).

Fig. 5 – (a) Size of flocs formed by 0.2 mmol/L FeCl₃·6H₂O under different pH conditions; (b) zeta potential of small PE particles (d < 0.5 mm) at pH 7.0.
of small-particle microplastics. After coagulation, a heterogeneous cake layer with large porosity was formed with a low dosage of Fe-based salts because of the influence of PE particles, leading to slight membrane fouling. With increasing dosage of coagulant, the porosity of the floc cake layer could be also influenced by PE particles, especially the large ones. As a result, less severe membrane fouling was induced compared to that with flocs alone. A detailed schematic diagram of microplastic behavior during the coagulation and UF processes is shown in Fig. 7.

Fig. 6 – Morphology of Fe-based flocs before and after coagulation with microplastics at pH 7.0: (a) flocs formed by 0.2 mmol/L FeCl₃·6H₂O; (b) flocs formed by 0.2 mmol/L FeCl₃·6H₂O and 12 mg/L anionic PAM; (c) flocs formed by 2 mmol/L FeCl₃·6H₂O and 12 mg/L cationic PAM; (d) flocs formed by 2 mmol/L FeCl₃·6H₂O and 12 mg/L anionic PAM. Other experimental conditions: weight of PE particles: 0.2 g; particle size of PE: d < 0.5 mm.

Fig. 7 – Schematic diagram of microplastics during coagulation and ultrafiltration processes.
4. Conclusions

The removal behavior of PE, the main component of microplastics, was investigated during the currently used drinking water treatment, including coagulation and UF processes. For coagulation, Fe-based coagulants were used because of their larger density compared to Al-based coagulants. The smaller the particle size of PE, the higher the removal efficiency was. However, low removal efficiency of PE particles (below 15%) was observed after coagulation, even in the presence of high dosage FeCl₃·6H₂O, indicating the difficulty for the traditional coagulation process with regard to microplastics removal.

In addition, PE removal efficiency was little influenced by water conditions, including ionic strength, NOM, and turbidity. In comparison to solution pH, PAM played a much more important role in removing PE particles due to the enhancement of floc adsorption ability. Compared to cationic PAM, anionic PAM performed much better in terms of PE removal efficiency due to its opposite charge to that of Fe-based flocs under neutral conditions.

For UF, PE particles could be completely rejected and only slight membrane fouling was induced because of their large diameter compared to the UF membrane pores. Although membrane fouling was gradually aggravated after coagulation, especially for the small PE particles, it was interesting that membrane fouling was less severe than that induced by Fe-based flocs alone. The potential reason was ascribed to the greater porosity of the cake layer influenced by PE particles, especially the large ones. Based on the removal behaviors of PE exhibited during coagulation and UF processes, we believe these findings have potential application in removal of microplastics during drinking water treatment.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jesz.2018.10.006.

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