Chemical Engineering Journal 428 (2022) 131161

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Efficient removal of microplastics from wastewater by an electrocoagulation process

Maocai Shen [a,b,c,1], Yaxing Zhang [b,c,1], Eydhah Almatrafi [c,1], Tong Hu [b], Chengyun Zhou [a,b,c], Biao Song [b,c], Zhuotong Zeng [a,b,*], Guangming Zeng [a,b,c,*]

[a] Department of Dermatology, Second Xiangya Hospital, Central South University, Changsha 410011, PR China
[b] College of Environmental Science and Engineering, Hunan University and Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, PR China
[c] Center of Research Excellence in Renewable Energy and Power Systems, Center of Excellence in Desalination Technology, Department of Mechanical Engineering, Faculty of Engineering-Rabigh, King Abdulaziz University, Jeddah 21589, Saudi Arabia

ARTICLE INFO

Keywords:
Microplastics
Electrocoagulation process
Aluminum electrode
Surfactant
Removal
Wastewater treatment

ABSTRACT

With the gradual increase of microplastics in the water environment, it is imperative to understand the removal characteristics of microplastics in the current treatment process. Electrocoagulation (EC) is an effective water treatment technology. The purpose of this study is to investigate the removal performance, mechanism and influencing factors of microplastics in wastewater treatment by EC. The impacts of wastewater properties, including initial pH, electrolyte concentration, applied voltage density, anode materials, microplastic type and microplastic concentrations, on the removal efficiency of microplastics by EC were systematically investigated. The findings showed that aluminum anode was better than iron anode in the removal of microplastics, and the removal rate of was above 80% in all experiments, which indicates that aluminum anode EC is an effective method to remove microplastics in wastewater. The removal rate of four microplastics by EC can reach more than 82% in the range of pH 3–10, and the best removal rate was 93.2% for PE, 91.7% for PMMA, 98.2% for CA and 98.4% for PP at pH 7.2. The removal efficiency of fiber microplastics by EC is better than that of granular microplastics. The microplastic removal efficiency increased with the increase of electrolyte concentration and applied voltage density. Additionally, microplastics undergo flocculation and charge neutralization at the same time during EC. The economic evaluation of the reactor operation cost showed that the optimal EC reaction conditions were: 0.05 M of electrolyte concentration, pH 7.2, 10 V of applied voltage density and Al anode. Further research should focus on the possible reactor design and improvement to optimize the process and realize the replication and transfer from the laboratory to the sewage treatment plant.

1. Introduction

Nowadays, microplastic pollution has attracted more attention from the public, and has evolved into a global environmental problem [1–3]. Evidence has shown that microplastics may be stable in the aquatic environment for thousands of years due to their chemical stability [4]. In recent years, microplastics, considered as an emergent pollutant, have caused serious economic losses in marine ecosystem [5]. Because of the large specific surface area, persistence and fluidity, microplastics are easier to adsorb organic chemical pollutants, heavy metals and harmful bacteria, resulting in the increasing impact of microplastics on the water environment and harm to human health [6–9]. In particular, because of its light density and small volume, microplastics are easy to be intake by aquatic organisms after entering the water body, resulting in intestinal abrasion and blockage [2,10]. The toxic chemicals attached to the plastic particles also have great harms to aquatic organisms, and eventually pass to human through the food chain, causing serious health problems.

In view of the increasingly serious pollution of plastics and microplastics in the world, governments have taken measures to reduce the entry of plastic products into the environment [11]. The municipal wastewater treatment plant has been proved to be a major contributor of

* Corresponding authors at: Department of Dermatology, Second Xiangya Hospital, Central South University, Changsha 410011, PR China.
E-mail addresses: zengzhuotong@csu.edu.cn (Z. Zeng), zgming@hnu.edu.cn (G. Zeng).
[1] These authors contribute equally to this article.

https://doi.org/10.1016/j.cej.2021.131161

Available online 6 July 2021

microplastics in the environment [12–14]. Although most of the wastewater treatment plants have effectively removed the microplastics, the municipal wastewater treatment plant is still an important source of microplastics in the environment [15]. In the process of land-based input, a large number of plastic particles (such as plastic beads in daily chemical products, fibers in laundry wastewater, car tire wear debris and other fragmented plastic waste) enter into the sewage pipe network, and then are treated by the sewage treatment plant and discharged into the environmental water body, and finally into the sea. At present, microplastics have not been included in the treatment scope of sewage treatment plants, and the research on the treatment efficiency and discharge of microplastics in the existing process is still in the initial stage. Although most of the microplastics (>90%) can be removed in the sewage treatment plant according to the existing treatment devices, some microplastics can still bypass the sewage treatment plant and enter the water environment with the wastewater [16]. This obviously needs an innovative, cheap and energy-saving solution, which can replace the existing three-stage treatment process to solve the removal of microplastics in sewage. Electrochemical technology, electrocoagulation, provides a cheap three-stage wastewater treatment process, which does not rely on the chemicals or microorganisms used in general chemical coagulation and traditional activated sludge process [17]. Electrocoagulation is a complex process in which metal electrodes produce cations under the action of electric field. The benefits of electrochemical processes include environmental compatibility, low capital cost, energy efficiency, sludge minimization, ease of automation and cost-effectiveness, and have been used to remove other pollutants from water [18,19].

There are three consecutive stages from the generation of ions to the formation of flocs: (1) under the action of electric field, metal cations are separated from anode to form “micro coagulant”; (2) they are combined with suspended particles in water and sink together by colliding flocs; and 3) coagulant forms a sludge layer for retaining suspended solid particles. The research on the removal of microplastics in wastewater by electrocoagulation is still in its infancy. Perren, Wojtasik and Cai [17] studied the removal of microplastics from simulated wastewater by electrocoagulation with Al anode, and the effects of pH value, current density and conductivity on the removal efficiency of microplastics were studied. The results show that the removal efficiency of microplastics in water by electrocoagulation was above 90%, especially 99.24% under neutral condition. However, the removal performance of microplastics by electrocoagulation needs to be further optimized and improved. Anode materials, types and shapes of microplastics, and the concentration of microplastics in wastewater all affect the removal of microplastics by electrocoagulation process.

In this paper, the feasibility of removing different types and shapes of microplastics in wastewater by electrocoagulation technology was systematically explored. Two granular microplastics (polyethylene, PE; polymethylmethacrylate, PMMA) and two fibrous microplastics (cellulose acetate (CA) from cigarette butts; PP from disposable surgical masks) were chosen in this study due to their large proportion in sewage. The addition of cationic surfactant can not only increase the water solubility of microplastics, but also increase the negative charge on the surface of microplastics. The effects of initial pH value of influent, electrolyte concentration (Na_2SO_4), applied voltage intensity, anode material (Al and Fe), and type, shape and concentration of microplastics on the removal of microplastics by electrocoagulation were studied. The feasible operation parameters of electrocoagulation technology in wastewater treatment were put forward according to the removal rate and energy consumption. The application prospect of this method in the three-stage treatment system of sewage treatment plant was evaluated, which provides a powerful support for the transfer and replication from theory to practice.

2. Materials and methods

2.1. Materials and regents

Two granular microplastics (polyethylene, PE; polymethylmethacrylate, PMMA) and two fibrous microplastics (cellulose acetate (CA) from cigarette butts; PP from disposable surgical masks) were chosen in this study. PE and PMMA are two common plastics in the daily life and obtained fromAladdin Chemical Company (Shanghai, China). Evidence has shown that more than 0.28 million tons of microplastic fibers are entering the aquatic environment from cigarette butts. Comparing this value with known sources of microplastic fibers from textile and laundries, it is expected to release about 0.28 million tons of microplastic fibers into the aquatic environment each year [20]. The popularity of COVID-19 has greatly increased the consumption of disposable masks, and has also increased its chances of entering the environment. It is estimated that there are about 129 billion masks used every month in the world during COVID-19, most of which are disposable masks made using microplastic fibers. A fully aged mask could release several billions of microplastic fibers into the aquatic environment once these fragile fragments enter the water without reservation [21]. Cigarette butts were collected from the environment, and masks were placed on the roof for two months of weathering treatment to obtain fiber microplastics [21]. The masks are placed in a transparent glass cover to prevent the interference of other environmental pollutants and the environmental pollution caused by weathered PP fiber. After two months of natural weathering, the materials were carefully recycled. Two kinds of anode materials, aluminum (Al) and iron (Fe), were obtained from the Aladdin Chemical Company (Shanghai, China). Anionic surfactant sodium dodecyl benzenesulfonate (SDBS) was purchased from the Aldrich (USA) and anhydrous sodium sulfate (Na_2SO_4), nitric acid (H_2SO_4) and sodium hydroxide (NaOH) were obtained from the Aladdin Chemical Company (Shanghai, China). The water used in the experiments was ultrapure water extracted by Milli-Q pure water mechanism.

2.2. Preparation of microplastic solution

In order to better understand the impact of wastewater properties, a wastewater simulator generates simulated domestic wastewater conditions while allowing full control variables. Evidence has shown that the particle size of microplastics found in wastewater was greater than 50 μm [15]. In view of the high concentration of microplastics in wastewater with smaller particles, the representative microplastics in granular and fibrous form were emphatically studied, which represent the greatest possibility of escaping from the treatment process such as flocculation, coagulation and sand filtration. The average particle sizes of PMMA and PE granular microplastics were measured to be 6.3 μm and 286.7 μm, respectively, while those of CA and PP fibrous microplastics were about 1–2 mm (Fig. S1). The concentration of microplastics used in this study was controlled at 0.05, 0.1, 0.2, 0.5, 0.8 and 1 g L^{-1}, respectively. PE microplastics are hydrophobic, so to ensure that the microspheres are completely dispersed in water, 20 mg L^{-1} of SDBS surfactant was added to the solution. The surfactant can simulate the average surfactant concentration of domestic sewage and help the microplastics to form a uniform suspension [22]. This concentration did not necessarily represent the concentration of microplastics in real sewage and effluent, but it had a directional effect.

2.3. Microplastic removal experiments

One liter of uniform suspension of microplastics with different concentration was used for each experiment, and the experiment was carried out immediately after preparation. During the reaction, the Al electrode (4 cm × 6 cm) or Fe electrode (4 cm × 6 cm) was used as anode, Cu electrode as cathode, a thickness of 0.1 cm, inserted into the

electrolytic cell as anode, and the plate spacing was 2 cm (Fig. S2). Anhydrous Na_2SO_4 was added to the solution as an electrolyte, and then a voltage was applied to conduct the electrocoagulation experiment. To explore the effect of different current intensity on the electrocoagulation process, the concentration of Na_2SO_4 was controlled at 0.01, 0.02, 0.05 and 0.1 M, respectively. The effect of applied voltage density on microplastic removal was performed under the conditions of 5 V, 10 V and 15 V. The sampling time was 1, 2, 3, 4, 5, and 6 h. The initial pH value (3–10) of simulated wastewater was adjusted by adding H_2SO_4 (1 M) or NaOH (1 M) solution to study its effect on the removal of microplastics by electrocoagulation process. With the development of electrocoagulation, the wastewater solution became more turbid, which is due to the formation of polymer flocs. All the experiments were carried out at room temperature with magnetic stirring speed of 150 rpms. These flocs were relatively easy to disperse in the whole container, and some beads can be seen attached to the flocs. All the solutions can effectively participate in the electrocoagulation reaction. After the electrocoagulation experiment, the solution was evenly stirred with a glass rod, and then the beaker was placed on a clean and closed operating platform and allowed to settle for 16 h [17]. All experiments were repeated three times, and all glass instruments were cleaned with ultrapure water and alcohol three times before use. After each test, the reactor vessel was washed with ultrapure water and ultrasonic assistance, and the electrode was soaked in 1 M H_2SO_4 solution for 30 min. This is to remove most of the oxide layer formed during the experiment, so as to prevent electrode passivation from affecting the removal efficiency.

2.4. Determination

After settling for 16 h, the sludge blanket formed sunk into the bottom of the reactor, taking away most of the microplastics. Compared with the original sample, the remaining liquid volume was significantly clearer. The removal efficiency of microplastics in the process of electrocoagulation was determined after each experimental operation. Briefly, the supernatant after settling was poured into a new clean glass beaker. The sludge was centrifuged at 3000 rpm for 10 mins at room temperature, and the supernatant was collected. All supernatants collected from each sample were filtered by 0.45 μm microporous membrane, and 1 M H_2SO_4 solution was added during the filtration to prevent flocs carried on the surface of microplastics from blocking the membrane. Then the filter membrane was dried at 40 °C for 24 h, and the mass of microplastics of the dried sample was measured by calculating the mass of each filter membrane before and after filtration. During the test, three parallel samples were taken at a time, and the average values of three samples were recorded. The total removal efficiency of microplastics in the electrocoagulation process at each time point was calculated by the following formula:

$$\eta = \frac{m_{in} - m_{end}}{m_{in}} \times 100\% \quad (1)$$

where η is the removal efficiency of microplastics during the electrocoagulation (%); m_{in} is the mass of microplastics in the solution at the beginning of electrocoagulation process (mg); m_{end} is the mass of microplastics in the supernatant after electrocoagulation process (mg).

The centrifuged sludge was further observed by metallographic microscope and scanning electron microscope to investigate the removal mechanisms. The concentrations of Fe^{3+} and Al^{3+} in the final effluent were also measured.

2.5. Quality assurance and quality control

Some quality assurance and quality control measures were used during the experiment. All the experimenters wore cotton clothes in the process of the experiment. All solvents were analytical grade (>95%), and all glassware were pre-cleaned with 30% ethanol solution, thoroughly rinsed with ultrapure water, then heat treated at 400 °C to remove organic impurities. By avoiding the use of any plastic equipment and using only cleaned glassware and metal items, the potential sources of microplastic contamination were minimized. All filters have been cleaned with ultrapure water before use to reduce the external microplastic pollution and its own microplastic pollution. During the experiment, the number of people in the laboratory was controlled. All filtration processes are carried out in a clean and enclosed operating platform.

3. Results

3.1. Variable study on microplastic removal

3.1.1. Effect of anode material

In all the studies in this research, a large number of different microplastics was removed by the electrocoagulation process, and the highest removal efficiency was measured to be >95%–100%. Statistical analysis indicated that the concentration of different microplastics in treated samples was always significantly lower than that in untreated control samples at each sampling time ($p < 0.01$). Different electrode materials have different electrochemical characteristics, so it is crucial to choose suitable electrode materials for improving the efficiency of electrocoagulation. The impact of anode materials on microplastic removal was thoroughly explored by using Al electrode and Fe electrode. The experimental conditions were as follows: the concentration of four microplastics (PE, PMMA, CA and PP) was 0.5 g L^{-1}, the concentration of electrolyte (Na_2SO_4) was 0.05 M, the pH of solution was not adjusted (pH = 7.2), and the applied voltage density was 10 V, respectively. Fig. 1 illustrated the effect of Al and Fe electrode on microplastic removal during electrocoagulation. It can be clearly seen that in the process of electrocoagulation, microplastic removal efficiency of Al as anode material was significantly better than that of Fe as anode material. The flocs produced by Fe electrode have dense precipitation and fast sedimentation, while the flocs produced by Al electrode have fast speed, colorless degree and strong adsorption capacity. With the increase of reaction time and floc concentration in the solution, the removal rate of microplastics was also increasing. When the reaction time continued for 4 h, the removal rate was basically unchanged (Fig. 1). The finally removal efficiency was 93.2% (Al) and 71.6% (Fe) for PE, 91.7% (Al) and 58.6% (Fe) for PMMA, 98.2% (Al) and 85.4% (Fe) for CA, and 98.4% (Al) and 82.7% (Fe) for PP fiber, respectively, which was similar to the results of previous study [17].

For granular microplastics, the removal rate of PE was higher than that of PMMA. The particle size of PE microplastic (286.7 μm) was larger than that of PMMA (6.3 μm), which made it easier to be trapped when the flocs produced by electrode sank and avoided escaping from the gap between flocs. This phenomenon was more common in the electrocoagulation test with Fe as anode, because it was difficult for the small size of the hydroxides to capture more PMMA microplastics in a large range. This also made that the removal efficiency of granular microplastics by electrocoagulation with Al anode was better than that of with Fe anode (Fig. S4–S7). For fibrous microplastics, the removal rate during electrocoagulation was obviously higher than that of granular microplastics (Fig. 1). It was because that the particle size of the two fiber microplastics (CA and PP) in the experiment was significantly larger than that of granular microplastics (PE and PMMA) (Fig. S1). In addition, the density (>1) of the two fiber microplastics was also one of the factors affecting the removal rate. The removal efficiency of fiber microplastics by electrocoagulation with Al electrode was more than 98% and 82% with Fe. The findings showed that electrocoagulation with Al electrode had better removal effect on microplastics, and microplastics with larger particle size and density were easier to be removed.

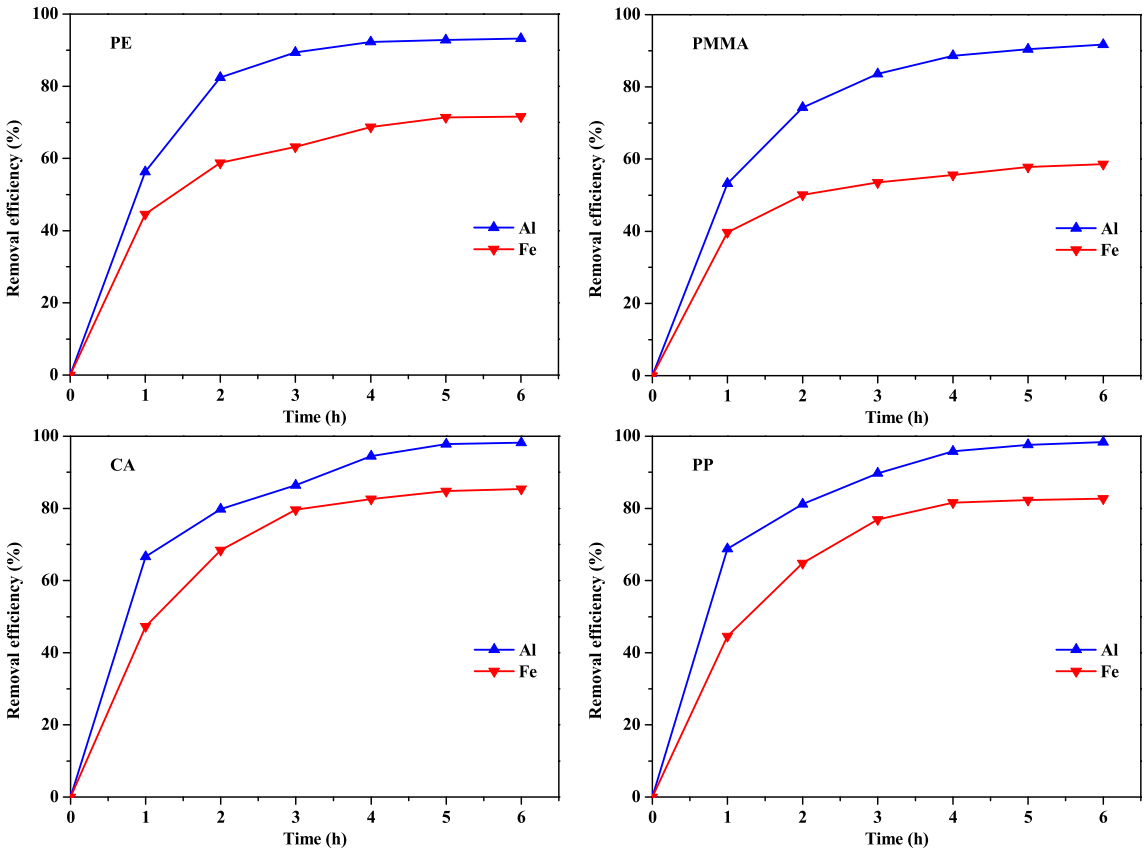

Fig. 1. Effect of Al and Fe electrode on removal of microplastics during electrocoagulation.

3.1.2. Effect of applied electrolyte concentration

Electrolyte concentration is a decisive parameter in electrochemical water treatment technology, which not only has a significant impact on the reaction effect, but also has a crucial impact on electric energy consumption and operation costs. The impact of electrolyte concentration on microplastic removal by electrocoagulation process was thoroughly studied by experiments under the conditions of 0.01 M, 0.02 M, 0.05 M and 0.1 M Na_2SO_4. The other experimental conditions were as follows: the concentration of four microplastics (PE, PMMA, CA and PP) was 0.5 g L^{-1}, the pH of solution was not adjusted (pH = 7.2), and the applied voltage density was 10 V, respectively. Fig. 2 showed the effect of different applied electrolyte concentration on microplastic during electrocoagulation. As shown in Fig. 2, with the increase of electrolyte concentration, the removal rate of microplastics increased significantly. In the same electrolysis time, with the increase of electrolyte concentration, the amount of Al^{3+} and Fe^{3+} dissolved out increased, and the effective Al and Fe species produced increased, thereby increasing the microplastic removal efficiency. The removal rate was 35.4%, 49.7%, 56.3% and 72.1% for PE, 39.4%, 48.9%, 53.2% and 68.4% for PMMA, 51.2%, 56.7%, 66.6% and 70.4% for CA, and 52.2%, 60.1%, 68.8% and 70.8% for PP with the electrolyte concentration of 0.01 M, 0.02 M, 0.05 M and 0.1 M, respectively after 1 h electrocoagulation with Al anode. When the reaction time was extended to 6 h, these removal efficiencies increased to 64.3%, 76.5%, 93.2% and 94.4% for PE, 72.4%, 81.3%, 91.7% and 93.7% for PMMA, 88.2%, 94.3%, 98.2% and 98.7% for CA, and 89.7%, 94.9%, 98.4% and 99.2% for PP with the electrolyte concentration of 0.01 M, 0.02 M, 0.05 M and 0.1 M, respectively. Additionally, the microplastic removal efficiencies also increased with the increase of electrolyte concentration with Fe anode (Fig. 2).

The impact of electrolyte concentration on the cost and operability of electrocoagulation process was also analyzed. The current findings demonstrated that when the electrolyte concentration increased from 0.05 M to 0.1 M, the removal effect of microplastics was not significantly enhanced (Fig. S4–S7). However, the flocs produced in the reaction process obviously increased and the electrode was seriously damaged. In addition, the results showed that the average current density was about 350 mA, 600 mA, 1.52 A and 2.56 A for electrolyte concentration of 0.01, 0.02, 0.05 and 0.1 M, respectively with Al anode, respectively. The energy consumption analysis implied that energy consumption was 91.2 Kwh m^{-3} and 153.6 Kwh m^{-3} after 6 h at 0.05 M and 0.1 M, respectively. The consumption of electrolyte in the process of electrocoagulation cannot be ignored. To ensure the stability of the above electrocoagulation, the minimum operating cost of microplastic removal was 7.1 kg m^{-3} Na_2SO_4 (0.05 M). Consequently, it is necessary to comprehensively consider the microplastic removal effect and the energy consumption of electrode in the actual operation process [17,23].

3.1.3. Effect of initial pH

The initial pH is also the main factor to determine the treatment efficiency, which can be used for the removal of feed water turbidity in the electrocoagulation process. In this study, the experiment was carried out in the range of initial pH value of 3–10 to investigate its effect on microplastic removal efficiency and the other experimental conditions were as follows: the concentration of four microplastics (PE, PMMA, CA and PP) was 0.5 g L^{-1}, the concentration of electrolyte (Na_2SO_4) was 0.05 M, and the applied voltage density was 10 V, respectively. Fig. 3 illustrated the impact of initial pH on microplastic removal during electrocoagulation over time. The findings showed that microplastics in all pH ranges were successfully removed, and the final removal rate was more than 80%, implying that electrocoagulation with Al anode was suitable for microplastic removal from wastewater with a wide range of pH value. The pH applicability of electrocoagulation process meant that it can be effectively used in almost all ordinary domestic wastewater containing microplastics without adding more chemicals to adjust the

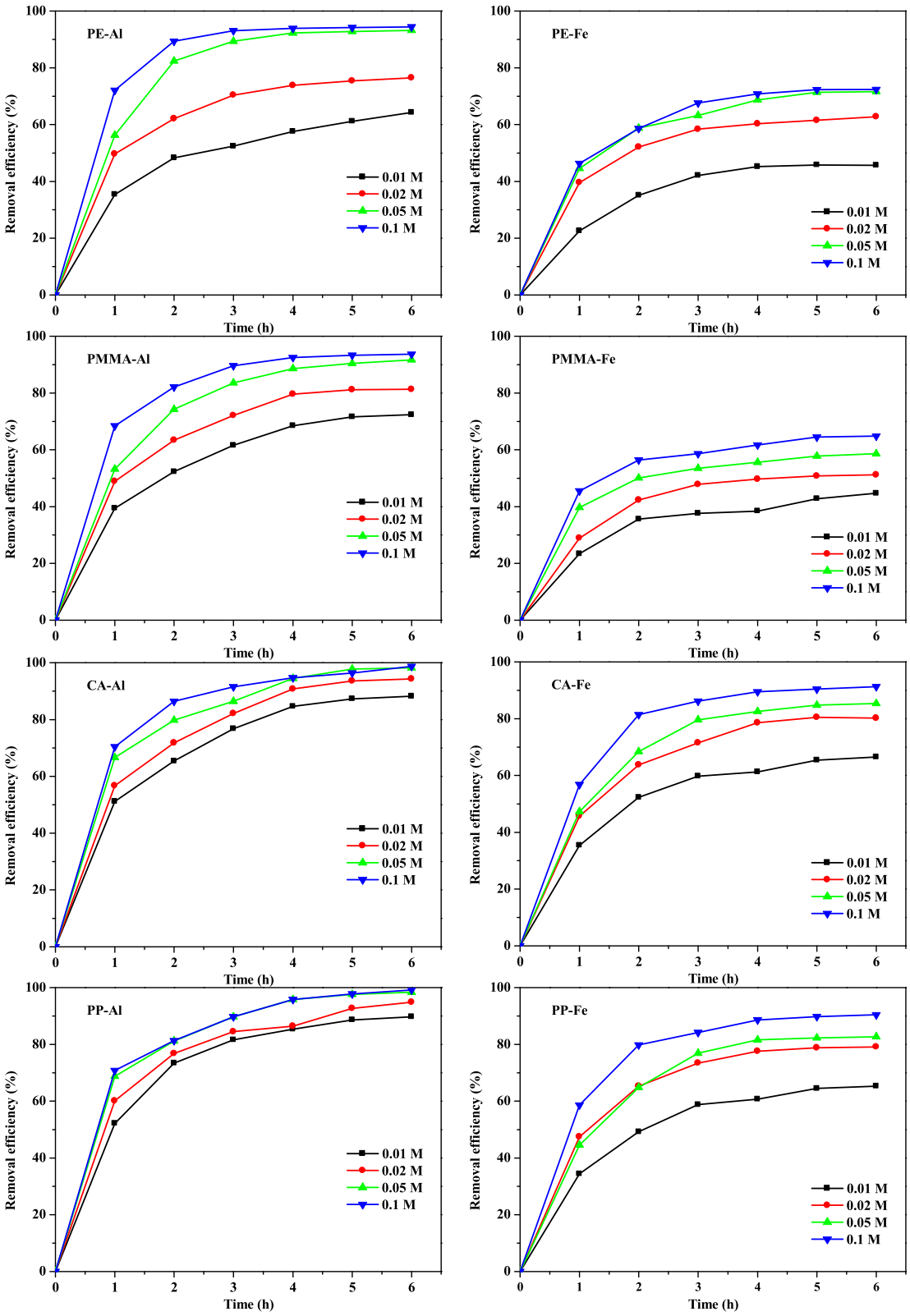

Fig. 2. Effect of different applied electrolyte concentration on removal of microplastics during electrocoagulation.

pH value of wastewater, because the pH value of municipal wastewater is usually 6–9.2 [24]. The final removal rate of each microplastic at pH 3 and pH 10 was lower than that of pH 5 and pH 7.2 (Fig. 3). When the pH value of initial water was 3, floc formation can hardly be observed in the process of Al electrode reaction in a short time, because the pH of Al^{3+} at the beginning of precipitation was higher, and there was little OH^- in the solution at this time, floc formation was not conducive. For Fe anode system, floc formation can be seen almost immediately due to the low initial precipitation pH of Fe^{3+}. These same experimental phenomena have also been reported in other studies [17,25]. With the increase of pH

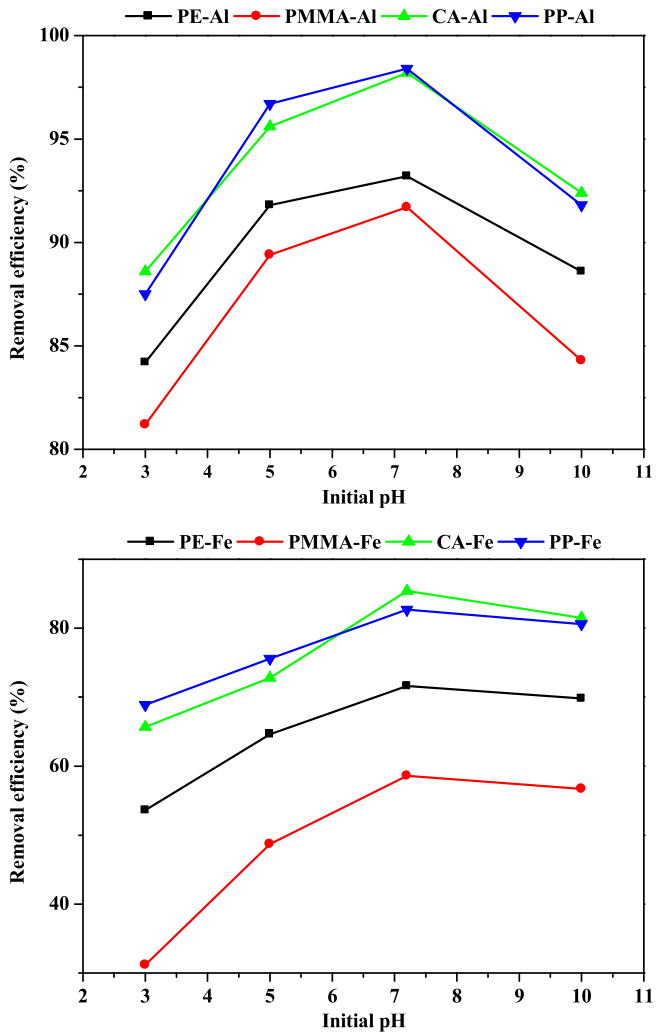

Fig. 3. Effect of initial pH on removal of microplastics during electrocoagulation.

value of water, the flocs formed in the reaction gradually increased, which was more conducive to the formation of $Al(OH)_3$ flocs and improved the removal rate of various microplastics. When the pH value of water was 10, the removal efficiency of microplastics showed a downward trend, which was due to the amphoteric nature of aluminum hydroxide. When the initial pH was 7.2, the final removal rate was 93.2% for PE, 91.7% for PMMA, 98.2% for CA and 98.4% for PP, respectively, and the pH value of simulated wastewater was similar to that of real municipal sewage. In addition, the zeta potential of four microplastics used in this study was shown in Fig. S8. Under neutral conditions, all the microplastics are negatively charged, which is more conducive to combine with the positively charged flocs to remove the microplastics from the water. The findings suggested that neutral pH value was expected to provide better removal effect of microplastics in real sewage treatment because it was conducive to the production of coagulants at neutral pH value during electrocoagulation.

3.1.4. *Effect of applied voltage intensity*

Applied voltage density is a key parameter in the application of constant voltage electrocoagulation, because it is an operating parameter, which can be directly controlled by DC power supply with different test voltage strength. The effect of applied voltage density on microplastic removal was performed under the conditions of 5 V, 10 V and 15 V, respectively, and the other experimental conditions were as follows: the concentration of four microplastics (PE, PMMA, CA and PP) was 0.5 g L^{-1}, the pH of solution was not adjusted (pH = 7.2), and the concentration of electrolyte (Na_2SO_4) was 0.05 M. Fig. 4 showed the removal effect of each microplastic over time under different applied voltage density by electrocoagulation. The removal rate was 34.3%, 56.3% and 64.5% for PE, 29.6%, 53.2% and 61.8% for PMMA, 42.6%, 66.6% and 71.2% for CA, and 44.4%, 68.8% and 73.8% for PP with the applied voltage density of 5 V, 10 V and 15 V, respectively, after 1 h electrocoagulation with Al anode. When the reaction time was extended to 4 h, these removal efficiencies increased to 62.4%, 92.3% and 95.8% for PE, 58.6%, 88.6% and 94.5% for PMMA, 76.3%, 94.5% and 98.6% for CA, and 78.5%, 95.8% and 97.6% for PP with the applied voltage density of 5 V, 10 V and 15 V, respectively. However, at prolonged time of 6 h, these efficiencies have not increased significantly (Fig. 4). Clearly, flocculation and precipitation seemed to be the main mechanisms for microplastic removal when excess flocs have produced during electrocoagulation. This showed that the increase of voltage (from 10 to 15 V) and metal ions would not lead to the increase of microplastic removal in the range of studied voltage density. The dominant time of coagulant excess and flocculation mechanism seemed to be within 4 h, because the voltage density seems to have a more important effect on the removal of microplastics during this period. As shown in Fig. 4, the gradient of the removal efficiency line decreased after 4 h. The findings suggested that compared with the precipitation after 4 h, the excess flocs have been produced when the reactor ran for more than 4 h, which has little impact of the microplastic removal efficiency. At the same time, the long-term operation of the electrocoagulation device would increase the sludge in the system, and increase the consumption of electrode and electric energy [17]. Additionally, based on these results, it was concluded that the increase of voltage intensity would increase the removal efficiency of microplastics. However, when voltage increased to a certain intensity, voltage density did not affect the removal efficiency of microplastics. Therefore, operation under appropriate voltage intensity can not only improve the removal efficiency of microplastics, but also save energy consumption. The start time of excess sludge production in the reaction system would depend on the conditions of the reactor, especially the current density, and can be used to determine the optimal operation time of the reactor concerned.

3.1.5. *Effect of microplastic concentration*

The concentration of microplastics in wastewater is also an important factor affecting the removal efficiency of electrocoagulation. Evidence has shown that the occurrence of microplastics in the influent varied from tens to tens of thousands, which was closely related to the local lifestyle and conditions [15]. Therefore, the concentration range of each microplastics was chosen from 0.05 to 1 g L^{-1} in this research. This concentration did not necessarily represent the concentration of microplastics in real sewage and effluent, but it had a directional effect. Fig. 5 illustrated the impact of microplastic concentration on removal efficiency during electrocoagulation. As shown in Fig. 5, generally, the removal rate of microplastics was decreasing with the increase of the concentration of microplastics. When the concentration of microplastics was selected at 0.05, 0.1 and 0.02 g L^{-1}, the removal rate was 89.7%, 92.7% and 88.6% for PE, 93.4%, 87.8% and 79.8% for PMMA, 86.9%, 85.6% and 83.7% for CA, and 89.2%, 87.3% and 86.5% for PP, respectively, after 2 h electrocoagulation with Al anode, which is similar to that of previous study [17]. The final removal rates of each microplastic at these three concentrations were all more than 97% (Fig. 5), implying that electrocoagulation process with Al anode has excellent removal effect for microplastics with lower concentrations. When the concentration of microplastics increased to 1 g L^{-1}, the final removal rates decreased to 87.5% for PE, 87.1% for PMMA, 91.8% for CA, and 94.3% for PP, respectively. For electrocoagulation with Fe anode, the highest removal rates of microplastics were 84.3% for PP, 68.8% for PMMA, 95.6% for CA, and 94.5% for PP, respectively, when the concentration of microplastics was 0.05 g L^{-1}. The results showed that the removal efficiency of electrocoagulation with Al anodewas much better

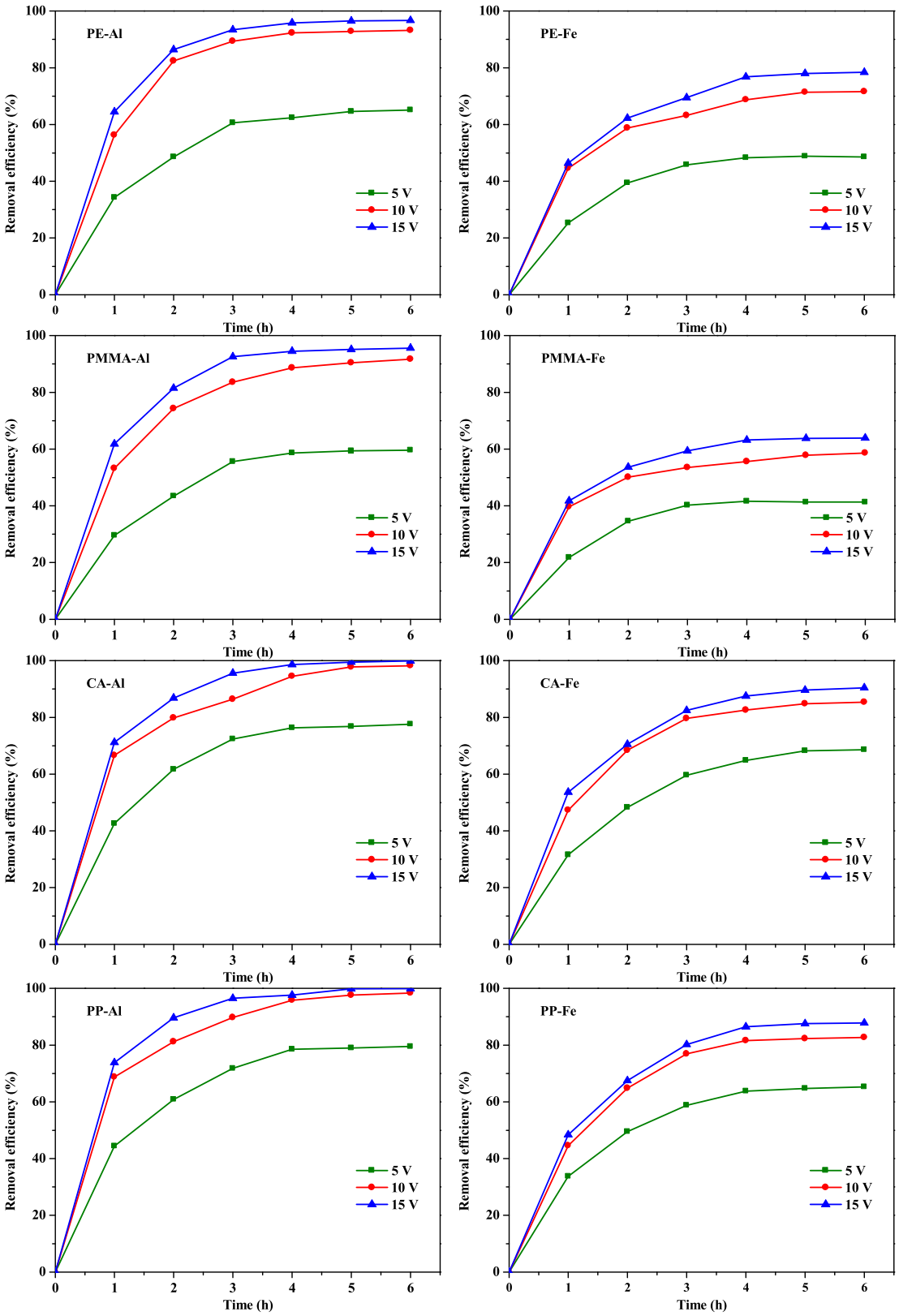

Fig. 4. Effect of different applied voltage intensity on removal of microplastics during electrocoagulation.

than that of electrocoagulation with Fe anode for microplastic removal. Moreover, compared with granular microplastic PE and PMMA, the removal efficiency of fiber microplastic CA and PP by electrocoagulation process was more obvious. Even when the concentration of microplastics has reached 0.8 g L^{-1}, which does not exist in the real sewage, and the final removal efficiency was still more than 95% (Fig. 5). These findings indicated that electrocoagulation with Al anode has a bright future in wastewater treatment, especially in the removal of microplastics.

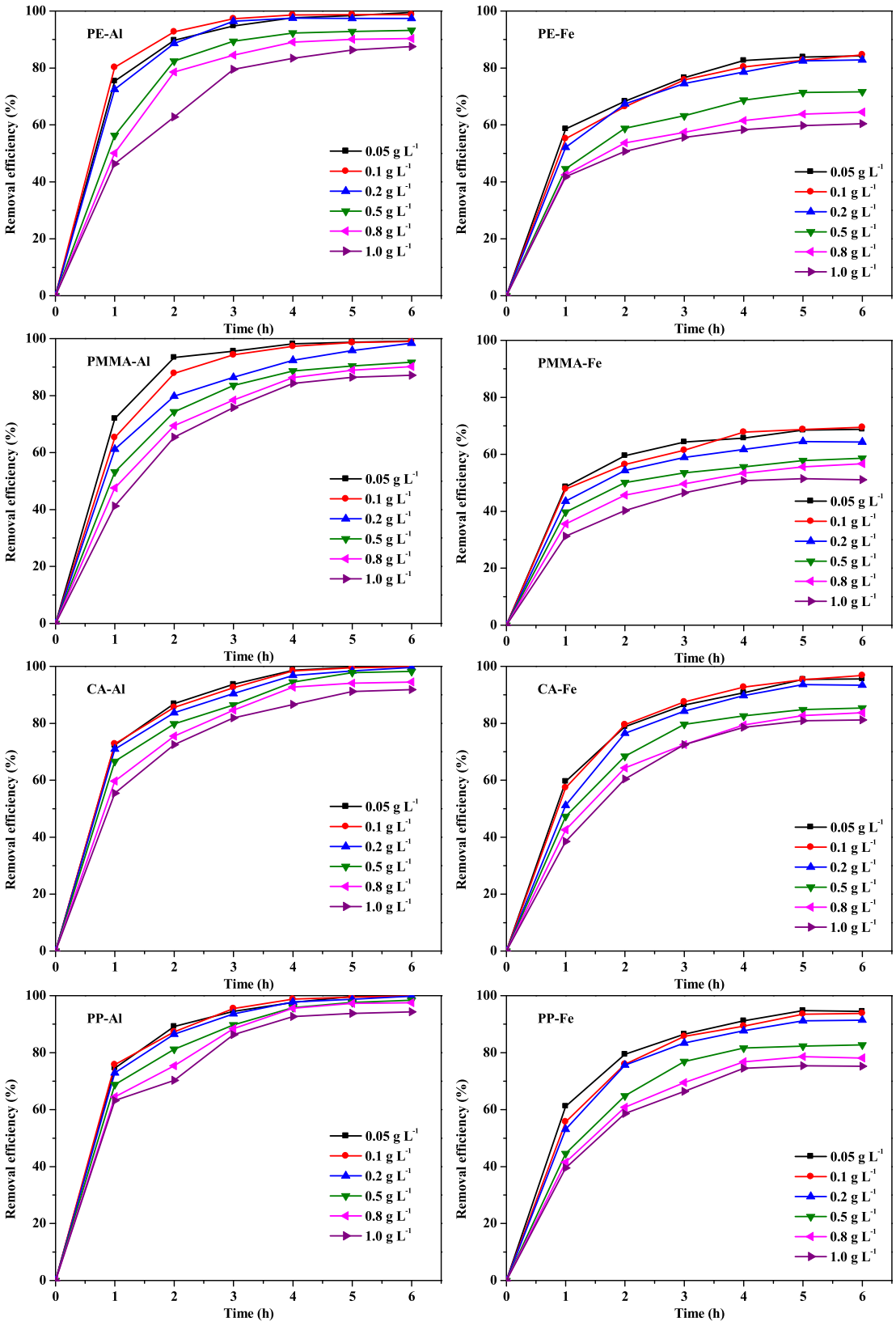

Fig. 5. Effect of microplastic concentration on removal of microplastics during electrocoagulation.

3.2. Removal mechanism

The mechanism and process of microplastic removal by electrocoagulation were well investigated. Firstly, Al^{3+} and Fe^{3+} were produced by anodic dissolution (Equation (2)), and with the increase of pH in the reaction process, Al^{3+} and Fe^{3+} would react with OH^- generated by cathode to from different hydroxides. For Al anode, Al^{3+} produced by electrolysis rapidly exists in the form of hydrated ion Al

$(H_2O)_6^{3+}$ in water and then quickly hydrolyzes to lose H^+, forming a series of mononuclear complexes, such as $Al(H_2O)_5OH^{2+}$, $Al(H_2O)_4OH^{2+}$, $Al(H_2O)_3OH^{2+}$, etc. The retention time of Al^{3+} in water is long, the hydration of Al^{3+} with OH^- is sufficient to form $AlOH^{2+}$, $Al(OH)_2^+$, Al(OH)3 and $Al_3(OH)_4^{5+}$, and the degree of polymerization and yield of polyaluminum are large, which is conductive to the subsequent decontamination (Fig. 6). Due to the increase of hydroxyl aluminum ions, the remaining lone pair electrons and the unsaturation of hydroxyl coordination ability, the hydroxyl group can polymerize with another Al^{3+} gradually to form a hydroxyl bridge structure, forming two hydroxyl bong bridges. As a result, the complex of mononuclear aluminum ($Al_m(H_2O)_x(OH)_n^{(3m-n)+}$) can be slowly polymerized into a network of nuclear polymers with rich hydroxyl groups on the surface, and finally transformed into amorphous flocculant ($[Al(OH)_3]_n$). The low degree of polymerization flocculant can remove the microplastics by adsorption, while the high degree of polymerization flocculant can capture and sweep microplastic particles by netting because of its large surface area and many surfaces groups [26]. Fig. 7 illustrated the morphology of Al flocs and Fe flocs after coagulation with different microplastic particles. It can be seen from the figure that aluminum flocs are distributed in flakes, while iron flocs are distributed in granules. The removal ability of aluminum flocs is better than that of iron flocs. Fig. 7 showed that the aluminum flocculant with high degree of polymerization is formed in the process of electrocoagulation, and the microplastics in the water are removed by net catching and sweeping. The results of flocculation experiment showed that the reactor was completely covered by flocs and there were obvious microplastics adsorbed on flocs.

Aluminum flocculant can polymerize rapidly under weak alkaline conditions, but due to the amphoteric characteristics of aluminum hydroxide, it is easy to dissociate into $Al(OH)_4^-$ when pH is too high. When there is sulfate in water, SO_4^{2-} adsorbed on $Al_m(H_2O)_x(OH)_n^{(3m-n)+}$ can promote the connection of more network polymers due to the attraction of hydrogen bond and charge. For Fe anode, Fe^{3+} mainly exists in the form of hydrated $Fe(H_2O)_6^{3+}$ in water. When encountering with OH^- in water, it will be hydrolyzed into a series of mononuclear hydrolysates, such as $Fe(H_2O)_5OH^{2+}$, $Fe(H_2O)_4OH^{2+}$, etc. Similarly, these mononuclear hydrolysates can polymerize into macromolecular polymers and finally form γ-FeOOH precipitates due to the unsaturation of hydroxyl groups.

Anode:

$$M \rightarrow M^{3+} + 3e^- \quad (2)$$

Cathode:

$$3H_2O + 3e^- \rightarrow \frac{3}{2}H_2\uparrow + 3OH^- \quad (3)$$

$$M^{3+} + 3H_2O \rightarrow M(OH)_3 + 3H^+ \quad (4)$$

In addition, $AlOH^{2+}$, $Al(OH)_2^+$, $Al(OH)_3$ and $Al_3(OH)_4^{5+}$ formed in the reaction process are positively charged, which can adsorb negatively charged ion microplastics. The addition of anionic surfactant can enhance the negative charge on the surface of microplastics in suspension solution [22], which also makes co-precipitate with microplastics in the process of forming $Al(OH)_3$. This is also an important mechanism for the removal of microplastics in the process of electrocoagulation.

Electrostatic adsorption:

$$AlOH^{2+} + 2Microplastics^- \rightarrow AlOH^{2+} - 2Microplastics^- \quad (5)$$

$$Al(OH)_2^+ + Microplastics^- \rightarrow Al(OH)_2^+ - Microplastics^- \quad (6)$$

$$Al_3(OH)_4^{5+} + 5Microplastics^- \rightarrow Al_3(OH)_4^{5+} - 5Microplastics^- \quad (7)$$

In order to ensure the complete and efficient hydration and polymerization of metal ions and flocculation process, the water flow of batch or circulating flow is usually used in electrocoagulation. However, the hydrogen evolution reaction of cathode will produce microbubbles, and the addition of surfactant will make the bubble production more intense [27]. When the formation rate is very high, the polymerization of metal ions will be disturbed, the formation of flocculant will be slow, and the degree of polymerization and density will be small.

3.3. Comparison with other studies

Electrocoagulation provides a simple wastewater treatment process, which does not rely on the chemicals or microorganisms used in general chemical coagulation and traditional activated sludge process [28]. The electrocoagulation process has been successfully applied to remove pollutants from sewage. The research on the removal of various pollutants in sewage by electrocoagulation process was shown in Table 1. Significantly, the available research on the removal of microplastics from wastewater by electrocoagulation process was very limited. Consequently, unquestionably, the comparison between this study and related literature was pale. A research done by Perren, Wojtasik and Cai [17] investigated the removal performance of PE microplastic from simulated wastewater by electrocoagulation. The results showed that the removal rate of PE microplastic was measured to be > 90% under all

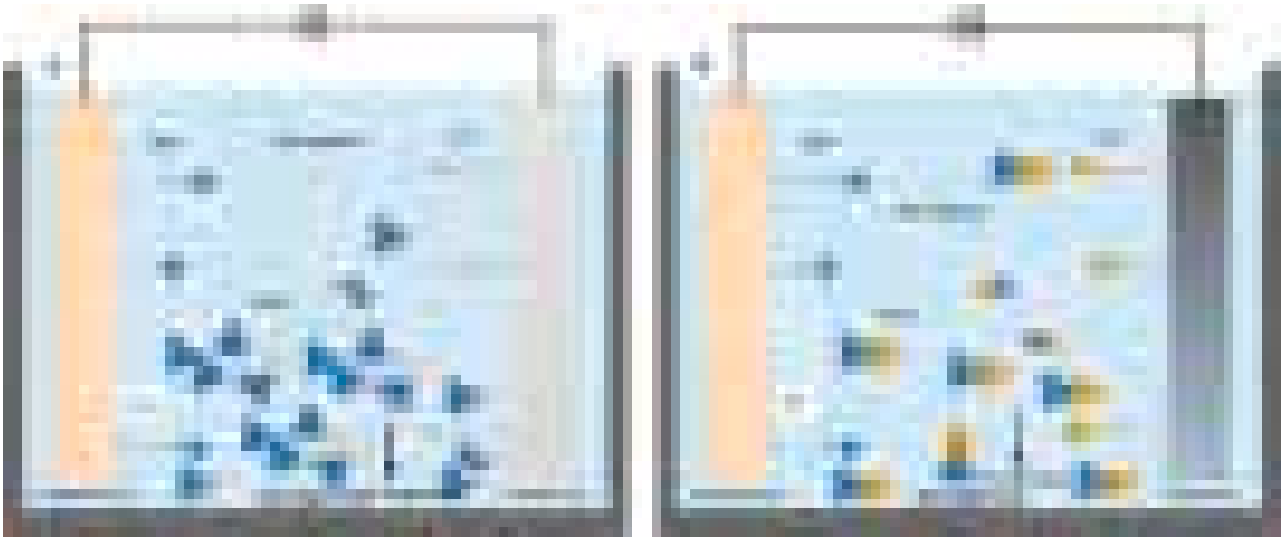

Fig. 6. Mechanism of microplastic removal from water by electrocoagulation process.

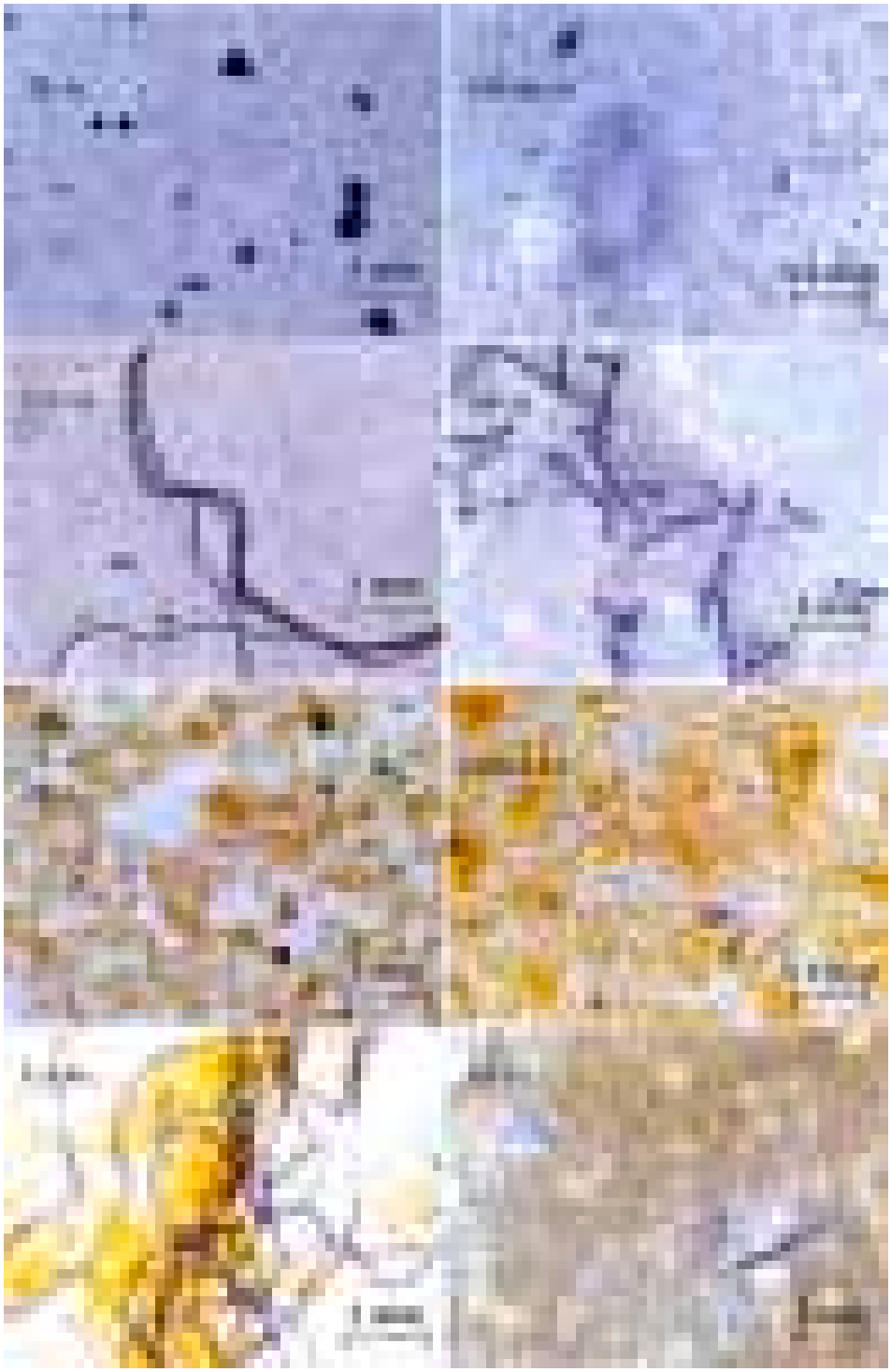

Fig. 7. Morphology of Al flocs and Fe flocs after coagulation with different microplastic particles (PE, PMMA, CA and PP).

simulated conditions, and the best removal rate could reach 99.24% under neutral condition (pH = 7.5). In this study, four microplastics were selected, and the removal efficiency was 98.6% for PE, 99.1% for PMMA, 99.9% for CA and 99.9% for PP under the same experimental conditions (Table 1, Fig. S4–S7). In addition, the removal efficiency of fiber microplastics was obviously higher than that of granular microplastics. This is mainly due to the type of pollutants. The microplastics experienced rapid charge neutralization, which can remove 70% of the initial microplastics. In the remaining operation period, flocculation mechanism played a leading role in purifying wastewater and removing residual microplastics in wastewater. Compared with iron, fluoride, dye, and bleaching effluent (Table 1), this combined mechanism allows the removal of a large number of microplastics. Despite some limitations of this study, considering the removal efficiency and operating costs of microplastics, electrocoagulation technology is transferable and replicable in laboratory and industry. It is also necessary to optimize the working method and operation time to further minimize the power and electrode consumption.

Table 1
Removal effect of different pollutants by electrocoagulation process.

Pollutant	Electrode material	Removal rate	COD removal	TSS removal	Optimuminitial pH	Reference
Domestic wastewater	Al	98.6%	84%	98.6%	–	[34]
Iron	Al	98.5%	–	–	6.0	[35]
Paint manufacturing sewage	Al	–	94%	89%	6.95	[36]
Strontium	Fe	93%	–	–	5.0	[19]
Bleaching effluent	Al	–	90%	94%	7.0	[37]
PE microplastic	Al	99%	–	–	7.5	[17]
PE microplastic	Al	98.7%	–	–	7.2	this study
PMMA microplastic		99.1%	–	–		
CA microplastic		99.9%	–	–		
PP microplastic		99.9%	–	–		
PE microplastic	Fe	84.6%	–	–		
PMMA microplastic		69.5%	–	–		
CA microplastic		96.8%	–	–		
PP microplastic		93.8%	–	–		

4. Discussion

Electrocoagulation technology has been successfully applied in water/wastewater treatment. Charge neutralization and scavenging flocculation are two main flocculation mechanisms, and pH value and current density are also key parameters. It is well known that Al^{3+} and Fe^{3+} show positive charge during hydrolysis, therefore, the characteristics of flocs play an important role in the removal of microplastics in the coagulation process [29]. This study has revealed the removal efficiency and mechanisms of four common microplastics by electrocoagulation with Al and Fe anode. The current findings showed that the removal effect of aluminum anode electrocoagulation technology on microplastics was very obvious (>98.6%), respectively. The average particle sizes of Al flocs and Fe flocs were measured to be 174.6 ± 24.3 μm and 243.8 ± 36.4 μm, respectively (Fig. 7). Small floc size means large specific surface area, and the results showed that Al anode electrocoagulation technology has a high removal rate of microplastics in the coagulation process. Compared with iron anode, aluminum anode was more suitable for water/wastewater treatment. The pH value of wastewater has a significant effect on the removal efficiency of microplastics by electrocoagulation, but the pH value of urban domestic sewage is 6.2–9 [24], which is just in the best pH range for the removal of microplastics by electrocoagulation.

In addition, interestingly, the current findings indicated that electrocoagulation technology has better removal effect on fiber microplastics in wastewater (>99%). Before that, fiber microplastics were often neglected in the research field of microplastics [30]. The fiber microplastics in wastewater mainly come from synthetic fiber in laundry wastewater, discarded cigarette butts and washable wet tissue, etc. Cigarette butts are usually made of cellulose acetate plastic, and one filter is usually consists of 15,000 strands of fibers, which can increase the accumulation of microplastic fibers in the environment in a short period of time [31]. Recently, a study done by Briain, Marques Mendes, McCarron, Healy and Morrison [32] investigated the material composition of various white fibers obtained from intertidal sediments near sewage treatment plants, as well as microplastics in in the sediments from repeated washing of real domestic waste. The findings suggested that the microplastic fibers extracted from these wastes were similar to those extracted from intertidal sediments near the wastewater treatment plant, and the wet wipes and sanitary towels flushed down the drain were an underestimated source of white microplastic fibers in the environment. Nowadays, the consumption of disposable masks has increased sharply with the global outbreak of COVID-19, which also increases the chances of discarded masks entering sewage treatment plants. Evidence has shown that the material of disposable mask is polypropylene [33]. When these plastic fibers enter the water, the plastic fiber strands will gradually separate and release micro plastic fibers. A completely aging mask releases billions of microplastic fibers into the environment, which is a great challenge to the sewage treatment system. The results of this study show that the electrocoagulation technology seems to be able to complete the removal of fiber microplastics in wastewater.

Furthermore, the coexistence of different types of microplastics and other pollutants in the sewage will complicate the removal efficiency in the wastewater treatment process. The relationship between the operation cost and the removal efficiency of pollutants by electrocoagulation technology should be considered. In addition to the advantages of in-situ no secondary pollution, high content of active ingredients, less sludge, simple device and easy to realize automatic control, electrocoagulation also shows the characteristics of wider range of pollutants, higher pollutant removal efficiency and lower energy consumption. Electrocoagulation technology also has some problems that cannot be ignored, which may limit the further development of the technology in microplastic removal, mainly for the following four points: (1) High conductivity requirements. In electrochemical technology, to maintain the electrochemical reaction, that is, the flow of current, the solution must have high conductivity. Therefore, the conductivity of the solution will directly affect the microplastic removal efficiency and operation cost of the whole process. (2) The anode needs to be replaced regularly. Due to the electrochemical dissolution of the anode, it will consume a lot of energy for a long time. If the anode is replaced irregularly, it will easily lead to the destruction of the plate and further cause pollution to the water body. (3) Residual metal ions. The metal ions dissolved in the process of electrocoagulation have a spontaneous hydrolysis process. In addition to the electrocoagulation effect, some metal ions will dissolve in water. For example, the residues of Al^{3+} and Fe^{3+} will lead to high chroma of water and toxic accumulation of human body, so the post-treatment process is generally needed to ensure that the iron and aluminum residues in the effluent meet the standard. (4) Anode passivation: anode passivation is the main factor limiting the application of electrocoagulation technology. The existence of passive film will slow down the dissolution rate of anode, reduce the current efficiency and increase the power consumption. Based on the good removal performance of microplastics, the current electrocoagulation process has good economic benefits and market prospects. However, the surface characteristics of various microplastics, the diversity of wastewater and its impact on the subsequent treatment process need to be further studied. At present, the research of enhanced electrocoagulation is still in the laboratory stage with small scale, and most of the water is synthetic wastewater, which is still a long way from engineering application. In the future, researchers need to pay more attention to the scale-up of the reactor, the treatment efficiency of the actual wastewater, and the economic problems, so as to ensure the early and wider application of the electrocoagulation technology.

5. Conclusion

Recently, with the continuous attention to the behavior and fate of

microplastics in the environment, it is urgent to understand their removal characteristics in the current water/wastewater treatment process. This study systematically explored the removal performance and mechanism of four microplastics (PE, PMMA, CA and PP) in wastewater by electrocoagulation process. The effects of anode material (Al and Fe), pH value, electrolyte concentration, applied voltage and microplastic concentration on the reaction were studied in an electrocoagulation reactor. The findings suggested that electrocoagulation technology is an effective method to remove microplastics from simulated wastewater. Compared with the process with Fe anode, Al anode system has better removal effect on all microplastics (>98.6%). When the initial pH of the influent was neutral (pH 7.2), the removal effect of microplastic reached the highest with the final removal rate of 93.2% for PE, 91.7% for PMMA, 98.2% for CA and 98.4% for PP, respectively. The removal efficiency of fiber microplastics (CA and PP) by electrocoagulation is better than that of granular microplastics (PE and PMMA). The microplastic removal efficiencies increased with the increase of electrolyte concentration and applied voltage density, while the flocs produced in the reaction process obviously increased and the electrode was seriously damaged. The consumption of electrolyte and other energy consumption in the process of electrocoagulation should be considered. In addition, in the process of electrocoagulation, microplastics undergo flocculation and charge neutralization at the same time. Flocs can capture and sweep microplastics by charge attraction and adsorption and the results of flocculation experiment showed that the reactor was completely covered by flocs and there were obvious microplastics adsorbed on flocs. By improving the electrocoagulation reactor, the operation cost of the reactor can be reduced by using these two mechanisms. It is suggested to further study the effect of reducing electrolyte concentration and current density on electrocoagulation operation efficiency and cost. Further research should focus on the possible reactor design and improvement to optimize the process and realize the replication and transfer from the laboratory to the sewage treatment plant.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The study is financially supported by the National Natural Science Foundation of China (82003363, 82073449, U20A20323, 51521006) and the Natural Science Foundation of Changsha (kq2007059).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2021.131161.

References

[1] M. Shen, T. Hu, W. Huang, B. Song, M. Qin, H. Yi, G. Zeng, Y. Zhang, Can incineration completely eliminate plastic wastes? an investigation of microplastics and heavy metals in the bottom ash and fly ash from an incineration plant, Sci. Total Environ. 779 (2021), 146528.
[2] M.F.M. Santana, L.G. Ascer, M.R. Custódio, F.T. Moreira, A. Turra, Microplastic contamination in natural mussel beds from a Brazilian urbanized coastal region: Rapid evaluation through bioassessment, Mar. Pollut. Bull. 106 (1-2) (2016) 183–189.
[3] M. Shen, S. Ye, G. Zeng, Y. Zhang, L. Xing, W. Tang, X. Wen, S. Liu, Can microplastics pose a threat to ocean carbon sequestration? Mar. Pollut. Bull. 150 (2019), 110712.
[4] A. Cozar, F. Echevarria, J.I. Gonzalez-Gordillo, X. Irigoien, B. Ubeda, S. Hernandez-Leon, A.T. Palma, S. Navarro, J. Garcia-de-Lomas, A. Ruiz, M.L. Fernandez-de-Puelles, C.M. Duarte, Plastic debris in the open ocean, Proc. Natl. Acad. Sci. U.S.A. 111 (28) (2014) 10239–10244.
[5] A. Hoffman, K. Turner, Microbeads and engineering design in chemistry: no small educational investigation, J. Chem.l Educ. 92 (2015) 742–746.
[6] J.C. Antunes, J.G.L. Frias, A.C. Micaelo, P. Sobral, Resin pellets from beaches of the Portuguese coast and adsorbed persistent organic pollutants, Estuar. Coast. Shelf Sci. 130 (2013) 62–69.
[7] M. Shen, Y. Zhu, Y. Zhang, G. Zeng, X. Wen, H. Yi, S. Ye, X. Ren, B. Song, Micro (nano)plastics: unignorable vectors for organisms, Mar. Pollut. Bull. 139 (2019) 328–331.
[8] J. Li, H. Liu, J.P. Chen, Microplastics in freshwater systems: a review on occurrence, environmental effects, and methods for microplastics detection, Water Res. 137 (2018) 362–374.
[9] C.D. Rummel, A. Jahnke, E. Gorokhova, D. Kühnel, M. Schmitt-Jansen, impacts of biofilm formation on the fate and potential effects of microplastic in the aquatic environment, Environ. Sci. Technol. Lett. (2017) acs.estlett.7b00164.
[10] B. Slootmaekers, C. Catarci Carteny, C. Belpaire, S. Saverwyns, W. Fremout, R. Blust, L. Bervoets, Microplastic contamination in gudgeons (Gobio gobio) from flemish rivers (Belgium), Environ. Pollut. 244 (2019) 675–684.
[11] D. Hu, M. Shen, Y. Zhang, H. Li, G. Zeng, Microplastics and nanoplastics: would they affect global biodiversity change? Environ. Sci. Pollut. Res. 26 (2019) 19997–20002.
[12] F. Murphy, C. Ewins, F. Carbonnier, B. Quinn, Wastewater treatment works (wwtw) as a source of microplastics in the aquatic environment, Environ. Sci. Technol. 50 (11) (2016) 5800–5808.
[13] L. Yang, K. Li, S. Cui, Y.u. Kang, L. An, K. Lei, Removal of microplastics in municipal sewage from China's largest water reclamation plant, Water Res. 155 (2019) 175–181.
[14] M. Enfrin, J. Lee, Y. Gibert, F. Basheer, L. Kong, L.F. Dumee, Release of hazardous nanoplastic contaminants due to microplastics fragmentation under shear stress forces, J. Hazard. Mater. 384 (2020), 121393.
[15] J. Sun, X. Dai, Q. Wang, M.C.M. van Loosdrecht, B.-J. Ni, Microplastics in wastewater treatment plants: detection, occurrence and removal, Water Res. 152 (2019) 21–37.
[16] S. Ziajahromi, P.A. Neale, L. Rintoul, F.D. Leusch, Wastewater treatment plants as a pathway for microplastics: development of a new approach to sample wastewater-based microplastics, Water Res. 112 (2017) 93–99.
[17] W. Perren, A. Wojtasik, Q. Cai, Removal of microbeads from wastewater using electrocoagulation, ACS Omega 3 (3) (2018) 3357–3364.
[18] M. Behbahani, M.R.A. Moghaddam, M. Arami, Techno-economical evaluation of fluoride removal by electrocoagulation process: optimization through response surface methodology, Desalination 271 (2011) 209–218.
[19] Z.V.P. Murthy, S. Parmar, Removal of strontium by electrocoagulation using stainless steel and aluminum electrodes, Desalination 282 (2011) 63–67.
[20] M. Shen, Y. Li, B. Song, C. Zhou, J. Gong, G. Zeng, Smoked cigarette butts: Unignorable source for environmental microplastic fibers, Sci. Total Environ. 791 (2021) 148384.
[21] M. Shen, Z. Zeng, B. Song, H. Yi, T. Hu, Y. Zhang, G. Zeng, R. Xiao, Neglected microplastics pollution in global COVID-19: Disposable surgical masks, Sci. Total Environ. 790 (2021) 148130.
[22] M. Shen, B. Song, G. Zeng, Y. Zhang, F. Teng, C. Zhou, Surfactant changes lead adsorption behaviors and mechanisms on microplastics, Chem. Eng. J. 405 (2021), 126989.
[23] M. Kiendrebeogo, M.R.K. Estahbanati, A.K. Mostafazadeh, P. Drogui, R.D. Tyagi, Treatment of microplastics in water by anodic oxidation: a case study for polystyrene, Environ.l Pollut. 269 (2021), 116168.
[24] M. Ahsan, A. Riaz, M.J. Jaskani, M. Hameed, Physiological and anatomical response of fragrant rosa species with treated and untreated wastewater, Int. J. Agric. Biol. 19 (01) (2017) 13–22.
[25] Y. Yavuz, Ü.B. Ögütveren, Treatment of industrial estate wastewater by the application of electrocoagulation process using iron electrodes, J. Environ. Manage. 207 (2018) 151–158.
[26] N. Drouiche, N. Ghaffour, S. Aoudj, M. Hecini, T. Ouslimane, Fluoride removal from photovoltaic wastewater by aluminium electrocoagulation and characteristics of products, Chem. Eng. Trans. 17 (2009) 1651–1656.
[27] S. Aoudj, A. Khelifa, N. Drouiche, Removal of fluoride, SDS, ammonia and turbidity from semiconductor wastewater by combined electrocoagulation–electroflotation, Chemosphere 180 (2017) 379–387.
[28] M. Shen, B. Song, Y. Zhu, G. Zeng, Y. Zhang, Y. Yang, X. Wen, M. Chen, H. Yi, Removal of microplastics via drinking water treatment: current knowledge and future directions, Chemosphere 251 (2020) 126612.
[29] B. Ma, W. Xue, Y. Ding, C. Hu, H. Liu, J. Qu, Removal characteristics of microplastics by Fe-based coagulants during drinking water treatment, J. Environ. Sci. 78 (2019) 267–275.
[30] A. Rebelein, I. Int-Veen, U. Kammann, J.P. Scharsack, Microplastic fibers - Underestimated threat to aquatic organisms? Sci. Total Environ. 777 (2021), 146045.
[31] F. Belzagui, V. Buscio, C. Gutiérrez-Bouzán, M. Vilaseca, Cigarette butts as a microfiber source with a microplastic level of concern, Sci. Total Environ. 762 (2021), 144165.
[32] O. Ó Briain, A.R. Marques Mendes, S. McCarron, M.G. Healy, L. Morrison, The role of wet wipes and sanitary towels as a source of white microplastic fibres in the marine environment, Water Res. 182 (2020) 116021, https://doi.org/10.1016/j.watres.2020.116021.
[33] T.A. Aragaw, Surgical face masks as a potential source for microplastic pollution in the COVID-19 scenario, Mar. Pollut. Bull. 159 (2020) 111517–111517.

[34] F. Ozyonar, B. Karagozoglu, Operating cost analysis and treatment of domestic wastewater by electrocoagulation using aluminum electrodes, Pol. J. Environ. Stud. 20 (2011) 173–179.
[35] K.S. Hashim, A. Shaw, R. Al Khaddar, M.O. Pedrola, D. Phipps, Iron removal, energy consumption and operating cost of electrocoagulation of drinking water using a new flow column reactor, J. Environ. Manage. 189 (2017) 98–108.
[36] A. Akyol, Treatment of paint manufacturing wastewater by electrocoagulation, Desalination 285 (2012) 91–99.
[37] R. Sridhar, V. Sivakumar, V. Prince Immanuel, J. Prakash Maran, Treatment of pulp and paper industry bleaching effluent by electrocoagulant process, J. Hazard. Mater. 186 (2-3) (2011) 1495–1502.