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Advanced Treatment of Laundry Wastewater by Electro-Hybrid Ozonation–Coagulation Process: Surfactant and Microplastic Removal and Mechanism

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Abstract: Laundry wastewater is supposed to be one of the most important sources of surfactants and microplastics in the wastewater treatment plant. Consequently, the aim of the study was evaluating the performance and mechanism of the electro-hybrid ozonation–coagulation (E-HOC) process for the removal of surfactants and microplastics. In this study, the efficiency of the E-HOC process for surfactant and microplastic removal was examined at different current densities and ozone dosages. Under the optimal reaction conditions (current density 15 mA·cm^{-2}, ozone dosage 66.2 mg·L^{-1}), both the removal efficiency of surfactant and microplastic can reach higher than 90%. Furthermore, the mechanism of surfactant and microplastic removal was investigated by electron paramagnetic resonance (EPR) and Fourier transform infrared spectroscopy (FT-IR). The results showed that the E-HOC (carbon fiber cathode) system can produce more reactive oxygen species (ROS), which can significantly improve the removal of the contaminants. In addition, the shape, size and abundance of the microplastics were analyzed. It was found that the shape of the microplastics in laundry wastewater is mainly fiber. Microplastics less than 50 µm account for 46.9%, while only 12.4% are larger than 500 µm. The abundance of microplastics in laundry wastewater ranges between 440,000 and 1,080,000 items per 100 L. The analysis of microplastics by FT-IR showed that most of the microplastics in laundry wastewater were polyethylene, nylon and polyester. These results indicated that the E-HOC process can effectively remove surfactants and microplastics from laundry wastewater.

Keywords: electro-hybrid ozonation–coagulation (E-HOC); laundry wastewater; hydroxyl radical; microplastic; surfactant

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1. Introduction

Domestic laundry wastewater is a kind of greywater [1]. In China, the average annual water consumption of each washing machine is about 10 m^3 [2]. According to statistics, the total amount of laundry wastewater discharged annually in China is about 10.71 billion tons [3]. If such a large amount of laundry wastewater is directly discharged into the environment without treatment, it will cause great burden and harm to the environment. Direct discharge of laundry wastewater may not only cause eutrophication, but also affect wastewater treatment plant performance [4]. Most of the laundry wastewater enters the wastewater treatment plant directly through the municipal wastewater plumbing, which can save water resources if it can be reused after treatment.

Domestic laundry wastewater often contains oils and fats, suspended solids, surfactants as well as microplastics, etc. [1,5]. Due to the addition of detergents, there is an increase in alkalinity and pH in laundry wastewater, which is rich in sodium, nitrogen, phosphorus and surfactants but has a low BOD [6]. Laundry wastewater has a COD, BOD and turbidity level of 375~4155 mg/L, 48~1200 mg/L and 14~400 NTU, respectively [3]. In

addition, surfactants and microplastics are two typical pollutants in laundry wastewater. Surfactants will cause potential harm to the environment after entering the environment. Studies have pointed out that surfactants not only cause damage to the morphology and physiology of plants, but also cause a significant decrease in sludge dewaterability [7,8]. Microplastic is another typical pollutant. The microplastics dropped from the clothes in the laundry process is also an important pollutant. Microplastics are defined as plastic particles and fibers less than 5 mm [9,10]. The study estimated that 9.1×10^{10} microplastic items are discharged from domestic wastewater to wastewater treatment plants every day in China [11]. Studies have shown that when activated sludge is chronically exposed to microplastics, both millimeter sized microplastics and nano-sized microplastics can cause a decline in sludge dehydration capacity [12]. In addition, the presence of surfactants also enhances the migration behavior of microplastics in the environment. [13]. This may contaminate ground water, rivers and lakes, etc. There have been many studies on the removal of surfactants and microplastics, mainly divided into physicochemical (membrane filtration, adsorption and electrocoagulation) and biological (aerobic, anaerobic, biofilter and wetland). They are briefly mentioned below.

For physicochemical treatments, both ultrafiltration and microfiltration membrane processes were highly effective, removing more than 90% of NPs/MPs fragment particles. Although the membrane filtration had a high efficiency, it needed to be cleaned frequently [14]. Using micron iron oxide powder (magnetite and hematite) as an adsorbent can remove 90% of cationic surfactants within 10 min, but a large amount of adsorbents are required [15]. Furthermore, studies have shown that about 90% of the surfactant can be removed by using electrocoagulation/electroflotation [16]. Another study showed that the removal efficiencies of surfactants and microplastics are 94% and 98%, respectively, by the electrocoagulation/electroflotation process [17].

For biological treatments, the aerobic moving bed bioreactor (MBBR) can stably remove 85~96% of the surfactant after 4 months of operation [18]. Moreover, an artificial wetland to treat greywaters was developed. Their research demonstrated 77% removal of LAS after 15 days of hydraulic retention time (HRT) [19]. Meanwhile, the removal of microplastics currently mainly relies on wastewater treatment plants, and studies have shown that oxidation ditch (OD) and MBR can remove 53.6% and 82.1% of microplastics, respectively. This may be due to the improved removal of microplastics by membrane filtration in the MBR [20].

The treatment process of laundry wastewater can generally be divided into three types: physicochemical process, biological process and the combination of physicochemical and biological process [2]. Specific processes include moving bed bioreactor (MBBR) [18], membrane filtration [21–25], solar photo-Fenton [26], electrocoagulation, ozonation [27], membrane bioreactor (MBR) [28], UV/H_2O_2 [29], biological treatment combined with UV/O_3 [30], etc. In these processes, the electrocoagulation process has been widely used because of its simple process, no need for manual dosing, low cost and high treatment efficiency [31]. There have been many studies on the treatment of laundry wastewater by electrocoagulation, which can effectively remove turbidity and chromaticity in water, and has a good removal efficiency on COD [16,17,32–34]. In addition, electrocoagulation process has a very good removal efficiency on microplastics. However, the electrocoagulation process has limited removal efficiency although it is able to remove part of the surfactant. It has been pointed out that the presence of surfactants can reduce the removal efficiency of microplastic fibers during coagulation [35]. Obviously, the single electrocoagulation process could not satisfy the treatment of all the wastewater. If it was combined with other advanced oxidation processes, the desired efficiency could be obtained. Advanced oxidation processes (AOPs) are increasingly used to remove surfactants from wastewater [36–40]. The combination of electrocoagulation process and electrooxidation process realizes the efficient removal of COD and TOC [41,42]. The coupling of the electrocoagulation process and ozone oxidation or electrocoagulation process in a series with ozone oxidation can achieve efficient removal of textile wastewater [43].

The research group has studied the removal mechanism of the relevant electro-hybrid ozonation–coagulation (E-HOC) process in the early stage. The research shows that the synergistic effects between ozone and coagulants (SOC) is helpful to improve the generation of hydroxyl radicals (•OH) in the reaction system [44]. In addition, the efficiency of the E-HOC process for pollutant removal was significantly improved in bipolar connection and the dual-coagulant generation mode [45]. Moreover, the E-HOC (composite anode, CA^+) process achieves high efficiency and simultaneous removal of organics and ammonia in leachate. The reactive chlorine species (RCS) generated in the E-HOC system promoted the generation of hydroxyl radicals [46]. However, the efficiency and mechanism of the E-HOC process for laundry wastewater treatment are still unclear.

In this study, the removal efficiency of C-E-HOC with a carbon fiber electrode as cathode and an aluminium electrode as anode on COD_{cr}, turbidity, LAS and microplastics in laundry wastewater was studied. The microplastics were analyzed by fluorescence microscopy for characterization, shape, size and abundance. The role of the carbon fiber electrode in the E-HOC process for production of reactive oxygen species analysis (ROS) was assessed through electron spin resonance (EPR), Fourier transform infrared spectroscopy (FT-IR) and H_2O_2 quantify to elucidate the mechanism of the simultaneous surfactant and microplastic removal in the C-E-HOC process.

2. Materials and Methods

2.1. Raw Water

Two kinds of raw waters are used in this study. One is synthetic water in the laboratory, and the other is taken from the drainage during the machine washing of household clothes (including laundry wastewater, primary rinsing water and secondary rinsing water). The raw water quality parameters were listed in Table 1 (Sodium linear alkylbenzene sulfonate (LAS) is a widely used anionic surfactant in synthetic detergents [47]). Synthetic water in the laboratory was used for the removal efficiency experiment of COD_{cr}, turbidity and LAS, and another raw water is used for the relevant research of microplastics.

Table 1. Raw water quality parameters.

Raw Water	COD_{cr} (mg/L)	Turbidity (NTU)	LAS (mg/L)
synthetic wastewater	800 ± 50	90 ± 20	100 ± 20
laundry wastewater	800 ± 100	97.5 ± 17.5	800 ± 50
primary rinsing water	140 ± 40	22.5 ± 7.5	15.5 ± 4.5
secondary rinsing water	80 ± 20	10.5 ± 4.5	8 ± 2

2.2. Experimental Agents

Dimethyl pyridine N-oxide (DMPO) used in EPR experiment is superior pure and purchased from Sigma Aldrich (St. Louis, MO, USA). The rest were purchased from Kermel, Tianjin, China. The potassium bromide (KBr) used in the experiment is spectral grade. Other reagents used in this study are analytical grade, such as potassium dichromate ($K_2Cr_2O_7$), mercury sulfate ($HgSO_4$), silver sulfate (Ag_2SO_4), concentrated sulfuric acid, potassium titanium oxalate ($C_4K_2O_9Ti{\cdot}2H_2O$), chloroform ($CHCl_3$), sodium hydroxide (NaOH), sodium dihydrogen phosphate monohydrate ($NaH_2PO_4{\cdot}H_2O$) and phenolphthalein ($C_{20}H_{14}O_4$). Methylene blue is indicator level. All solutions were prepared with ultrapure water.

2.3. The E-HOC Experiment

The experiment was carried out in a cubic glass container with a reaction volume of 500 mL at room temperature. In this system, carbon fiber or stainless steel was used as the cathode and aluminium was used as the anode. The length, width and thickness of the electrodes are 7 cm, 6 cm and 0.1 cm, respectively. The effective reaction area of the electrode was 84 cm^2. The reaction time was 60 min, and slow stirring was always

performed at 100 rpm during the reaction. The ozone generator converts the oxygen source to ozone, which is fed through an aerated head to the reaction container. The ozone generator was purchased from Sankang, Jinan, China. The electrode was connected with a direct current (DC) power supply, which was purchased from ANNAISI, Wuxi, China. A small amount of Na_2SO_4 was added to the water sample before the reaction to adjust the electrical conductivity. The electrocoagulation reaction was started simultaneously with the ozonation reaction. The flocs were allowed to settle sufficiently by standing for 30 min at the end of the reaction before the assay was performed. The removal efficiencies of the E-HOC processes for COD_{cr}, turbidity and LAS were determined at different current density or different ozone dosage.

2.4. Analytical Methods

2.4.1. Determination of Physicochemical Indexes

The COD was analyzed by fast digestion-spectrophotometry [48]. Before the determination, the interference of suspended solids in the sample should be minimized. The appropriate scale was selected according to the water quality of the water samples to be tested. The turbidity was measured by a portable turbidimeter, purchased from HANNA, Padovana, Italy. The LAS was determined by methylene blue spectrophotometry [49]. Water samples need to be pre-filtered by medium-speed qualitative paper filter to remove suspended solids in order to remove insoluble anionic surfactants in the water samples before the determination. H_2O_2 was determined by potassium titanate oxalate spectrophotometry. A sample was taken at 10 min (adjusted pH to 3), and 20 mL of the water sample to be measured was placed in a 25 mL colourimetric tube. Next, 1 mL of 0.05 mol/L potassium oxalate color developer was added to the water sample and subsequently diluted to the scale, shaken well and left for 8 min. The sample was poured into a 10 mm cuvette and measured with an ultraviolet spectrophotometer (UV-4802) at 385 nm wavelength.

2.4.2. Fourier Transform Infrared Spectroscopy (FT-IR) Analysis

The flocs after completion of the reaction were first frozen in the refrigerator and subsequently cold dried in a vacuum freeze dryer until the moisture completely disappeared. The sample was obtained by mixing 10 mg of the cold dried floc and 990 mg of KBr and grinding in an agate mortar. The samples were analyzed using a FT-IR spectrometer (Model Nicolet 6700, Thermo Fisher Scientific, Waltham, MA, USA) at the range from 4000 to 500 cm^{-1}.

2.4.3. EPR Analysis

EPR experiments trap hydroxyl radicals in the reaction system at room temperature by trapping agent DMPO. Experiments were performed on a Bruker EMXmicro spectrometer (Bruker Corporation, Bremen, Germany). Water samples were filtered through a 0.45 μm filter before the determination. Next, 30 mM of the radical trapping agent DMPO were added to each 500 μL of the water sample. The instrument determined the following parameters: resonance frequency of 9.77 GHz, microwave power of 25.18 mW, modulation frequency of 100 kHz, modulation amplitude of 1.0 G, sweep width of 700 G, time constant of 81.92 ms, sweep time of 35 s, and receiver gain of 30 dB.

2.4.4. Microplastics Analysis

In all, 500 mL water samples were passed through a glass fiber filter membrane (0.7 μm. 47 mm Ø, Xingya, Shanghai, China) and the samples were filtered. The filtered filter membrane was placed in a Petri dish and wrapped in aluminium foil. Several fine holes were tied on the surface of aluminium foil with needles, and the dish was placed in an oven (60 °C) to dry for 1 h and then observed under a fluorescence microscope. The microplastics observed under the microscope were classified and counted according to shape and size.

3. Results and Discussion

3.1. Removal Performance

The E-HOC process at the different current densities or different ozone dosages for the removal efficiency of COD_{cr} (a_1, b_1), turbidity (a_2, b_2), as well as LAS (a_3, b_3) in laundry wastewater is shown in Figure 1. According to Figure 1, the E-HOC process resulted in a superior COD_{cr} removal efficiency compared with the electrocoagulation (EC) and ozonation processes. The use of the EC process or ozonation process as the separate treatment process faces important practical limitations. Neither the electrocoagulation process nor the ozonation process can effectively remove COD_{cr}, turbidity and LAS. Obviously, a single treatment process cannot meet the treatment standards [50]. By contrast, the E-HOC process can remove COD_{cr}, turbidity and LAS efficiently. Our previous study found that Al-based coagulants can not only enhance·OH generation during ozonation, but also the addition of ozone promotes the conversion of the Al salt coagulant hydrolysis form to the higher polymerized Al species. Consequently, there are synergistic effects between ozone and coagulants (SOC) in the E-HOC process. [44]. In addition, contrasting Figure 1a_2 with Figure 1b_2, it can be seen that the removal of turbidity was mainly dependent on the EC process. This is similar to the previous research results. EC has an outstanding efficiency on removing turbidity in the water samples [51–54].

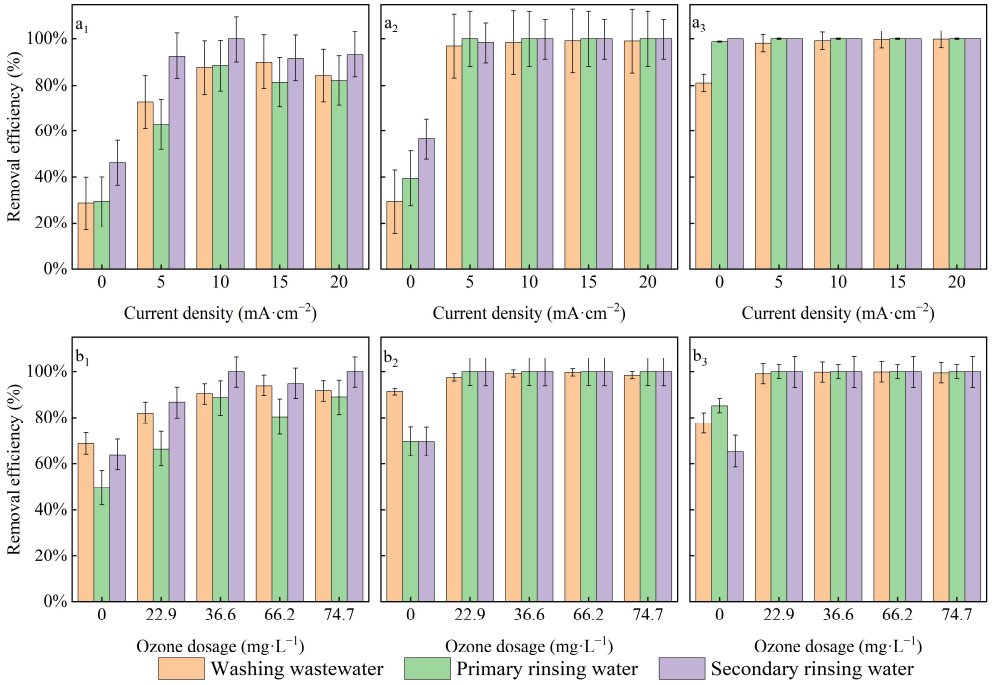

Figure 1. The E-HOC process at different current densities (**a_1**,**a_2**,**a_3**) or different ozone dosages (**b_1**,**b_2**,**b_3**) for the removal efficiency of COD_{cr} (**a_1**,**b_1**), turbidity (**a_2**,**b_2**), LAS (**a_3**,**b_3**).

However, the EC process is less effective for the removal of LAS than the ozonation process. Contrasting Figure 1a_3 with Figure 1b_3, it can be seen that the ozonation process can remove LAS from laundry wastewater with high efficiency. There are two ways for ozone to oxidize organic compounds in laundry wastewater: (i) direct reaction between ozone and organic molecules or (ii) indirect reaction by attack of free radical species produced by ozone decomposition. Studies have shown that ozone has the highest contribution rate in the process of removing surfactant. The radical reaction plays a dominant role in the oxidation of surfactant [55]. In addition, studies have shown that the presence of low concentrations of surfactants can contribute to the ozone dissolution and indirect generation of hydroxyl radicals in the ozone process [56]. Therefore, the removal of LAS mainly depends on the ozonation process.

Ultimately, the optimal reaction conditions of the E-HOC process in three laundry wastewaters were obtained ((washing wastewater, 15 mA·cm^{-2}, 66.2 mg·L^{-1}) (primary rinsing water, 10 mA·cm^{-2}, 36.6 mg·L^{-1}) (secondary rinsing water, 10 mA·cm^{-2}, 36.6 mg·L^{-1})). Under the optimal reaction conditions, the removal rates of COD_{cr}, turbidity and LAS in washing wastewater are 93.9%, 99.7% and 99.9%, respectively. The pollutants in laundry wastewater are mainly long-chain alkanes, aromatic compounds, esters and ethers. Long-chain alkanes are firstly decomposed into short-chain alkanes. These substances subsequently further decomposed into small molecule alkanes as well as small molecule organic acids. Finally, long-chain alkanes, alkenes, ethers and esters have been completely degraded [57,58].

3.2. Microplastic Analysis

3.2.1. Microplastics Removal Performance

The removal efficiency of the EC and E-HOC processes for microplastics from three laundry wastewaters are shown in Figure 2. According to Figure 2, both E-HOC and EC had a significant efficiency in removing the microplastics. In addition, a slightly lower microplastics removal efficiency for the E-HOC process can be seen compared to the EC process at the same reaction conditions. This is probably due to the addition of ozone. Ozone causes the large flocs formed in the EC process to be broken into small flocs, resulting in part in the microplastics falling off the flocs and returning back to the water sample. Compared with the EC process, the flocs produced by E-HOC process are looser and smaller in particle size.

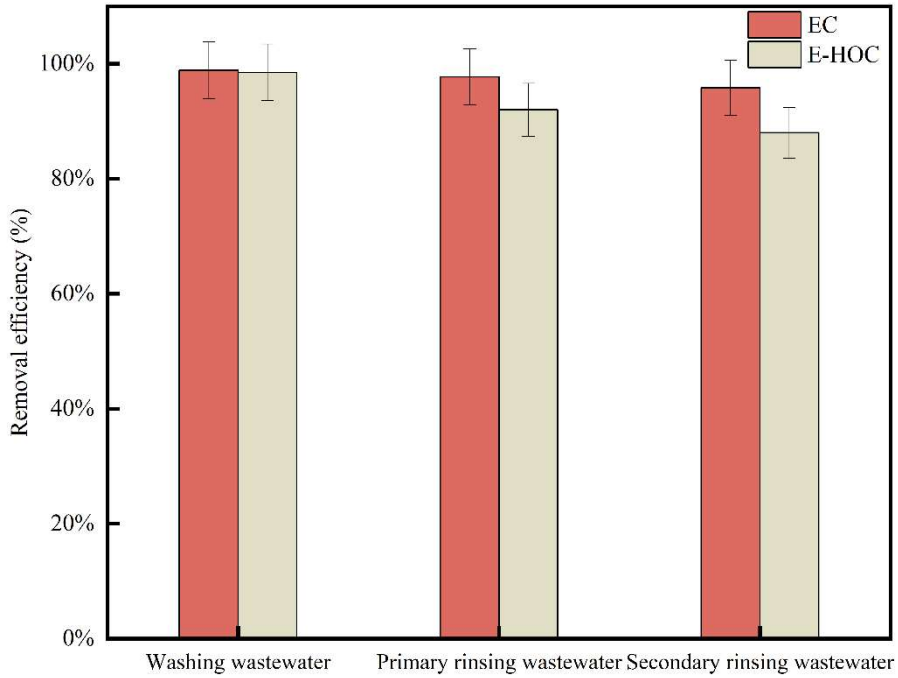

Figure 2. The EC and E-HOC processes on the removal efficiency of microplastics from three laundry wastewaters.

The process of EC can produce aluminium salt coagulant in situ at the Al electrode, which synergistically removes microplastics from water bodies through a variety of mechanisms including charge neutralization, adsorptive bridging and sweep flocculation [59]. There have been many previous studies that have shown that EC has excellent removal efficiency for microplastics. The removal efficiency can reach 96.5% for microplastics under the reaction conditions with a current density of 2.88 mA·cm^{-2} and an initial pH of 4 for the real wastewater samples [60]. In addition, the aluminium anode is superior to the iron anode in removing microplastics. When the pH is within the range from 3 to 10, the

removal efficiency of EC for microplastics can reach more than 82%, and when the pH is 7.2, the maximum removal efficiency for microplastics can reach 98.4% [61]. A considerable number of studies have proved that the removal efficiency of microplastics by the EC process can reach more than 90% [17,62,63].

3.2.2. Shape and Size Analysis

Microplastics are divided into fibers, particles, fragments and films based on the shape [64–67]. Before collecting laundry wastewater, wash the washing machine twice without adding clothes to reduce interference. The shape of the microplastics observed under the fluorescence microscope is shown in Figure 3. As shown in Figure 3, fiber-shaped microplastics account for the vast majority of them. Clothing releases a large number of microplastic fibers during its washing, and laundry wastewater is an important source of microplastic fibers in wastewater treatment plants and the environment [68].

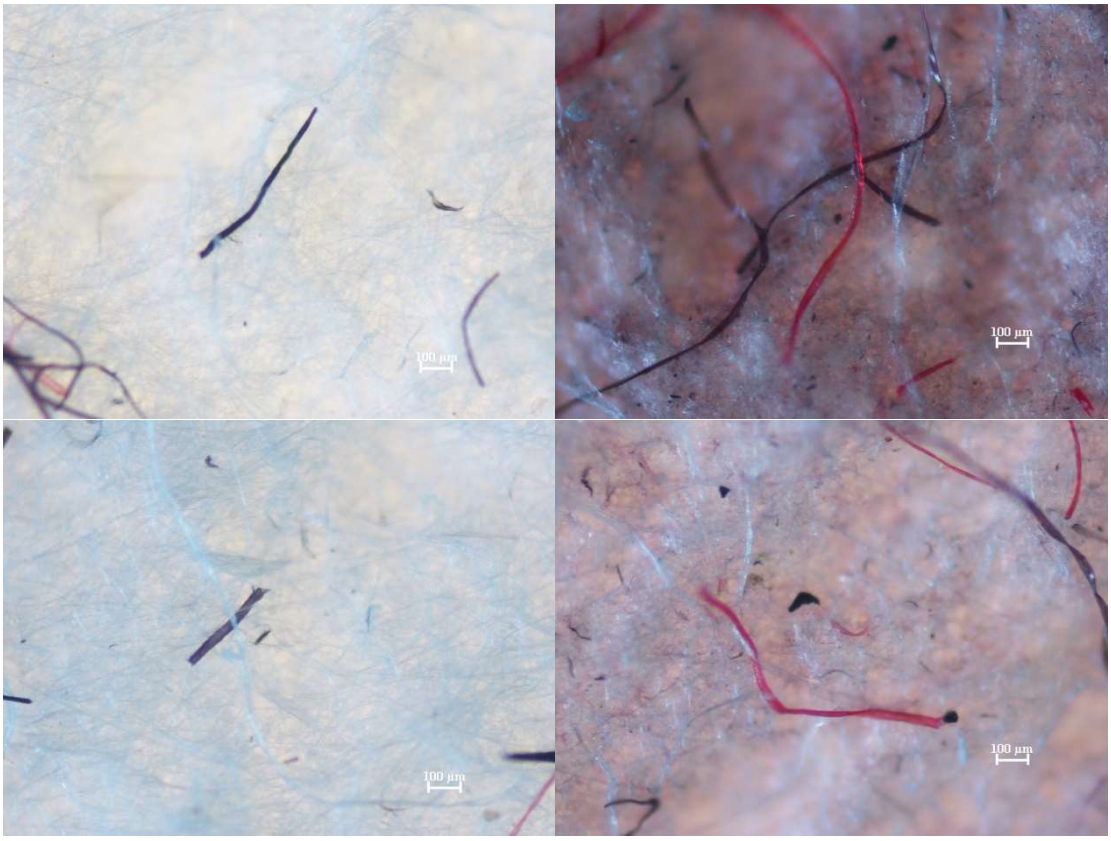

Figure 3. Microplastics observed by fluorescence microscopy.

To further study the characteristics of microplastics in laundry wastewater, the shape and size distribution of the microplastics is shown in Figure 4. As shown in Figure 4, 73.8% of the microplastics detected in laundry wastewater were fibers. According to the microplastics size, it was divided into four ranges: less than 50 µm, more than 50 µm and less than 200 µm, more than 200 µm and less than 500 µm, more than 500 µm. According to Figure 4, 46.9% of the microplastics were less than 50 µm, Only 12.4% of the microplastics were larger than 500 µm. The microplastics were available in sizes ranging from 6.1 µm to 4000 µm. A study found that the average length of the microplastics found in household laundry wastewater was 2258.59 µm [69]. In addition, only 7% of the microplastics fibers were longer than 500 µm, 40% were between 100 and 500 µm and 53% were between 50 and 100 µm [70]. In household laundry wastewater, the study found that 48.64% of the released microfibers were shorter than 5 µm in length, and only 11.49% of the microfibers were longer than 500 µm [71]. This is similar to our findings.

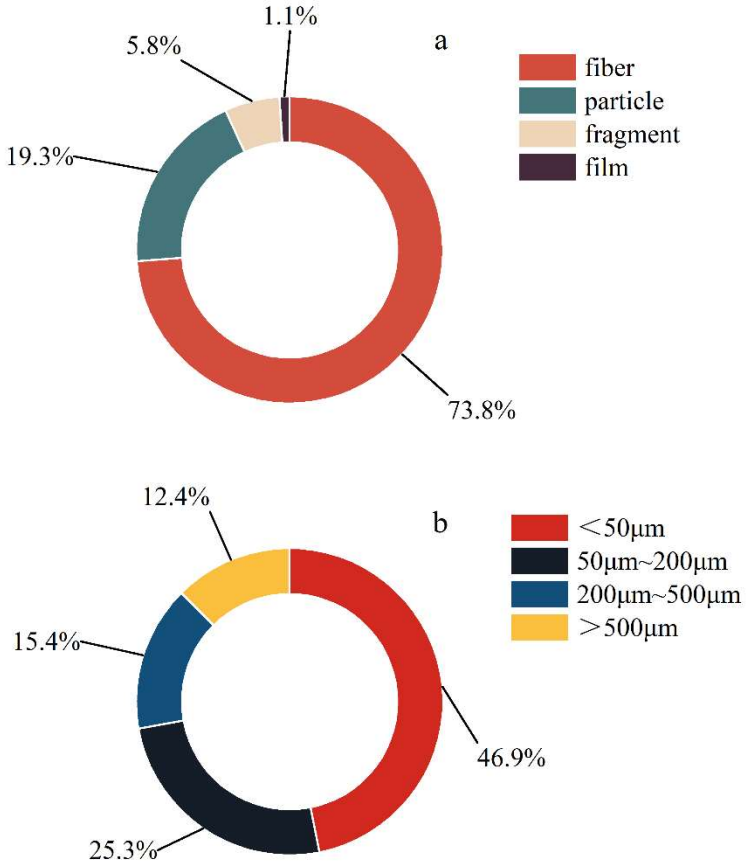

Figure 4. Shape (**a**) and size (**b**) distribution of microplastics.

3.2.3. Quantitative Analysis

Due to the consideration of the factor that the amount of water in laundry wastewater is large, even low concentration of microplastics have many microplastics discharged to the environment. As shown in Figure 5, overall, the amount of microplastics in the washing wastewater was the largest, followed by the primary rinsing wastewater and finally the secondary rinsing wastewater. In terms of the overall quantity, sample 1 released 44 × 104 items of microplastics per 100 L of water, and sample 4 released 108 × 104 items of microplastics per 100 L of water. This can be caused by many factors including the fabric, the textile process as well as the thickness of the clothing. Studies have confirmed that the interlacing coefficient of textiles affects the number of particles in wastewater. The results show that the higher the yarn's density/cm in the fabric, the lower the fiber content in the wastewater [72]. One study has shown that more than 150,000 fibers are released per 1 kg of laundry wash [11]. Cleaning PET knitted fibers in the washing machine releases 4489.93 fibers/L or 368,094.07 fibers per kilogram [73].

It can be seen that the quantity of microplastics is easily affected by many factors, but considering that the total amount of microplastics discharged into the environment is still large, its potential harm to the environment cannot be ignored. We can think of a front-end design to remove microplastics before they enter the wastewater treatment plant or the environment.

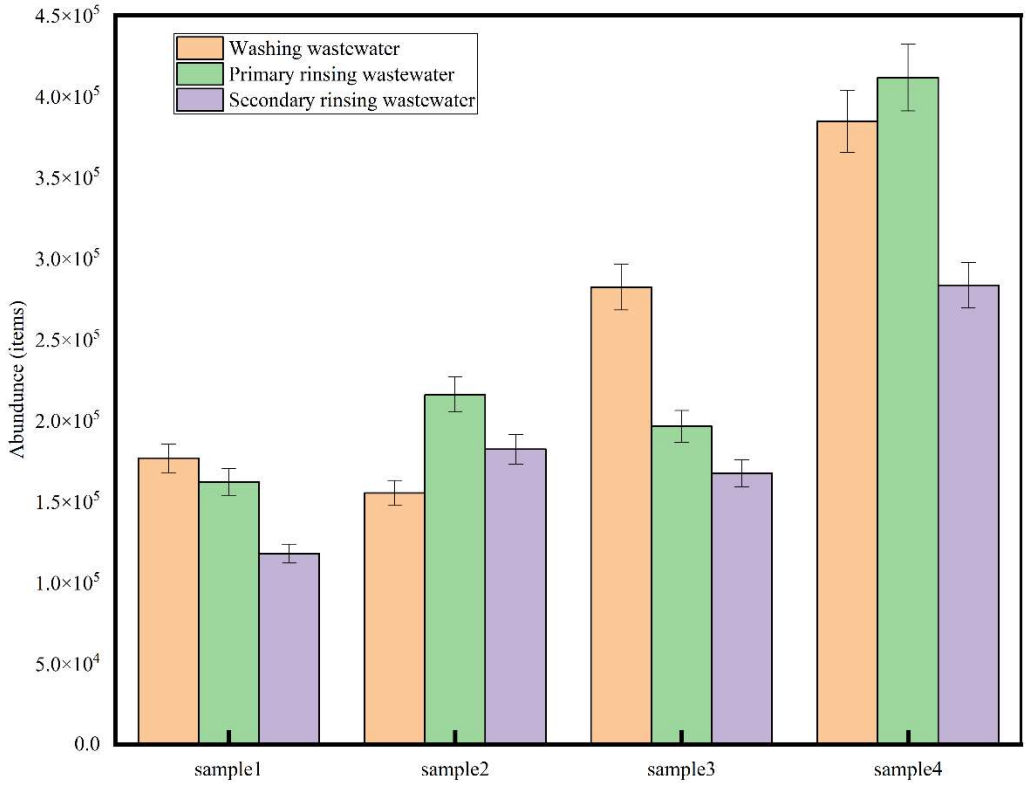

Figure 5. Total drainage microplastics emissions in different stages.

3.2.4. FT-IR Analysis of Microplastics

The FT-IR spectra of the microplastics detected from the different stages of laundry wastewater are shown in Figure 6. According to Figure 6, the kinds of microplastics in laundry wastewater at the different stages did not change obviously. Comparing with FT-IR spectra of standards [74], it is found that the microplastics in laundry wastewater are mainly polyethylene (PE), nylon (Polyamide, PA) and polyester (PET). The peaks of PE, PA and PET are 2914 and 2850 cm^{-1}, 1650 and 1540 cm^{-1}, 1700 and 1060 cm^{-1}, respectively. These are the plastic polymers commonly used in textiles [69]. PE may also come from personal care products such as toothpaste and facial cleanser [75]. This is similar to the composition of microplastic fibers in wastewater treatment plants [76].

3.3. Mechanism Analysis

3.3.1. Reactive Oxygen Species Analysis

The production of •OH in the reaction system was compared by EPR when the cathode was a carbon fiber electrode and a stainless steel electrode, respectively (Figure 7). According to Figure 7, there were four peaks centered at 3500 G in the EPR spectrum with a peak height ratio close to 1:2:2:1, which was a typical DMPO-•OH profile [77,78]. •OH was detected in both laundry wastewater and ultrapure water, but it was not obviously detected in ultrapure water by the carbon fiber cathode system. Compared with Figure 7a,b, the EPR peak of •OH generated by the carbon fiber cathode system is higher than that of the stainless steel cathode system, which is because the carbon fiber cathode will generate H_2O_2 in the system (Equation (1)) [79], and H_2O_2 will further react and convert into •OH. This indicates that more •OH is produced in the carbon fiber cathode system than in the stainless steel cathode system. Previous studies of our group have shown that $•O_2^-$ can also be produced in the reaction system [44,45], which can be further converted to •OH in the system (Equations (2) and (3)) [80,81]. The change of pH in the system before and

after the reaction was measured. The carbon fiber system changed from 8.3 to 8.9, and the stainless steel system changed from 8.26 to 8.78.

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2, \quad (1)$$

$$\bullet O_2^- + H^+ \rightarrow H_2O_2 + O_2, \quad (2)$$

$$\bullet O_2^- + H_2O_2 + H^+ \rightarrow O_2 + \bullet OH + H_2O, \quad (3)$$

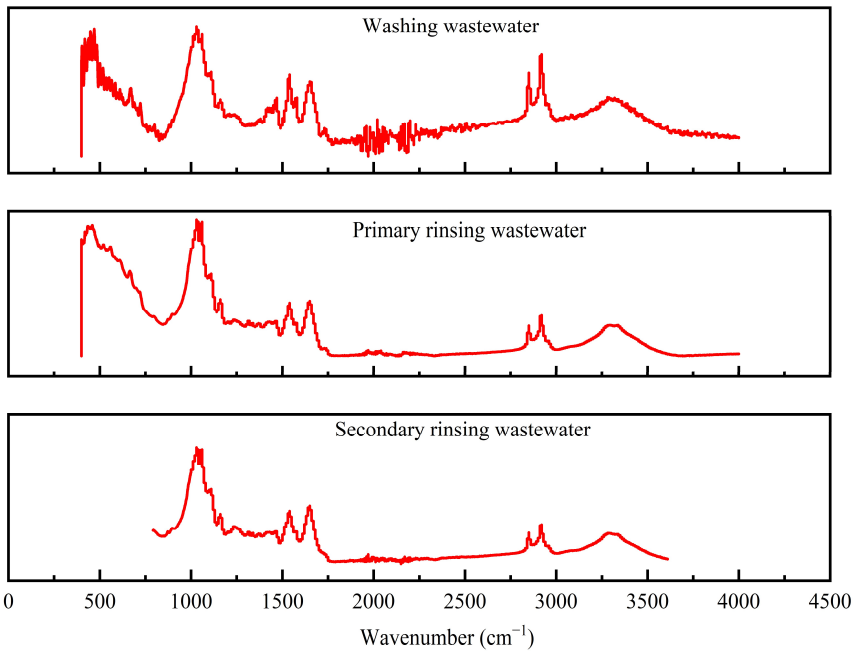

Figure 6. FT-IR spectra of microplastics from laundry wastewater.

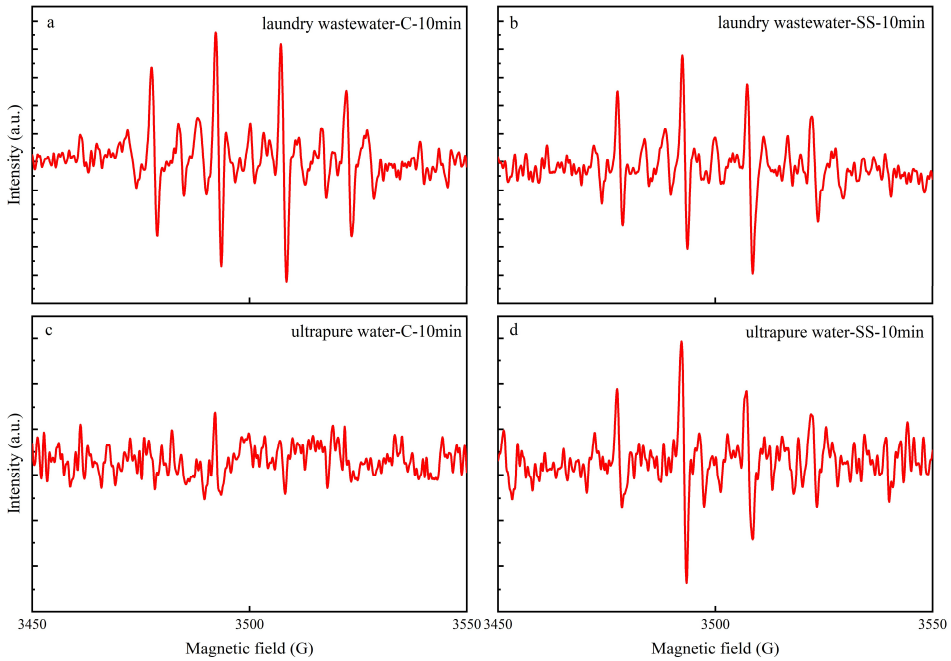

Figure 7. EPR spectra detected in the E-HOC processes. Reaction conditions: current density 15 mA·cm^{-2}, ozone dosage 66.2 mg·L^{-1}. Samples for EPR test were taken at the 10th min during the reactions. Experimental condition: raw water (laundry wastewater (**a**,**b**), ultrapure water (**c**,**d**)), cathode (carbon fiber (**a**,**c**), stainless steel (**b**,**d**)).

To further verify the ability of the carbon fiber cathode system to produce H_2O_2, the concentration of H_2O_2 in the reaction system at different current densities or under different reaction conditions was determined in the washing wastewater. There are two groups of reactions. One is to keep the ozone dosage constant, only changing the current density, while the other is to keep the current density constant, only changing the ozone dosage.

Figure 8 shows the ability of the carbon fiber cathode and the stainless steel cathode to produce H_2O_2 under different current densities or different ozone dosages by using titanium salt photometry. According to Figure 8, under different reaction conditions, the carbon fiber cathode had a stronger ability to produce H_2O_2 in the reaction system than the stainless steel cathode. The best reaction conditions were further demonstrated by the fact that the carbon fiber electrode has the highest capacity to produce H_2O_2 in the reaction system with a current density of 15 mA·cm^{-2} or ozone dosage of 66.2 mg·L^{-1}. Under the optimal reaction conditions (current density of 15 mA·cm^{-2}, ozone dosage of 66.2 mg·L^{-1}), the carbon fiber system produced H_2O_2 at a concentration 32.2% higher than that of the stainless steel system. This also verifies the result that the peak height of the carbon fiber cathode system is higher than the stainless steel cathode system in EPR experiments.

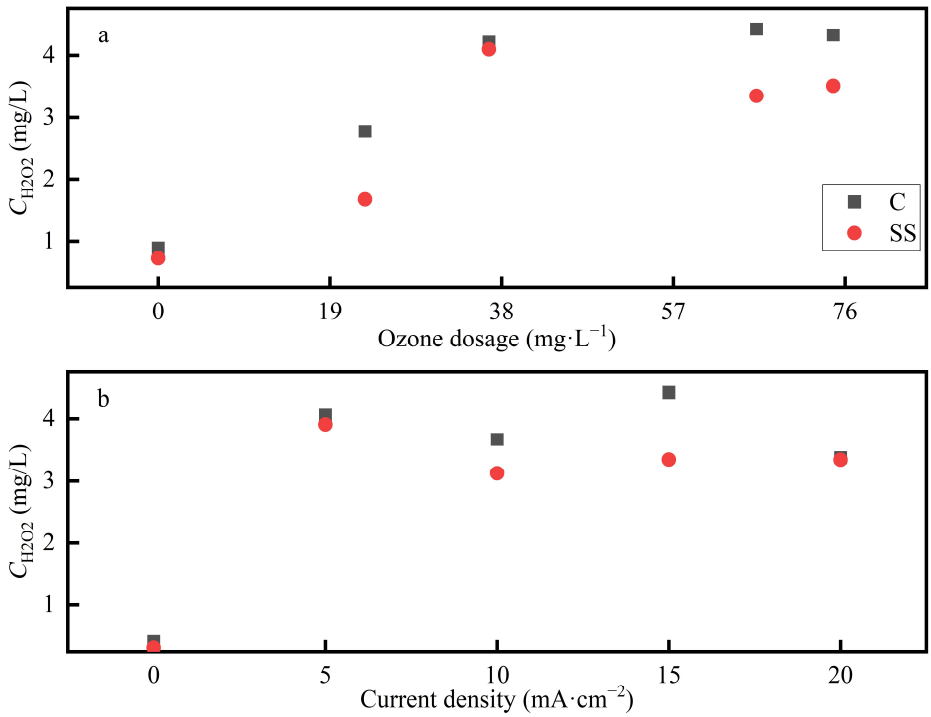

Figure 8. Concentration of H_2O_2 under different cathode electrodes (**a**) at different ozone dosages (0, 22.9, 36.6, 66.2 and 74.7 mg·L^{-1}) (**b**) at different current densities (0, 5, 10, 15 and 20 mA·cm^{-2}).

Based on the previous research, •OH, H_2O_2 and $•O_2^-$ were generated in the E-HOC process. The •OH quenching experiences indicated that •OH accounted for approximately 90% contribution for the E-HOC process, which indicated •OH played a dominant role in pollutant removal [45].

3.3.2. FT-IR Analysis of Flocs

In order to further define the characteristics of the E-HOC process with the carbon fiber electrode as the cathode, the functional group changes of the flocs after the reaction of the contrasting carbon fiber system with the stainless steel system were determined. As shown in Figure 9, the peak around 1630 cm^{-1} is mainly caused by the stretching vibration of the -C=C- or O-H bending of the absorbed water molecules [82–85]. The peak around 3400 cm^{-1} results from the stretching of the O-H bond of the hydroxyl group [86]. The

peak around 1400 cm^{-1} may be due to the formation of the H bond with the aluminium hydroxide precipitates [87].

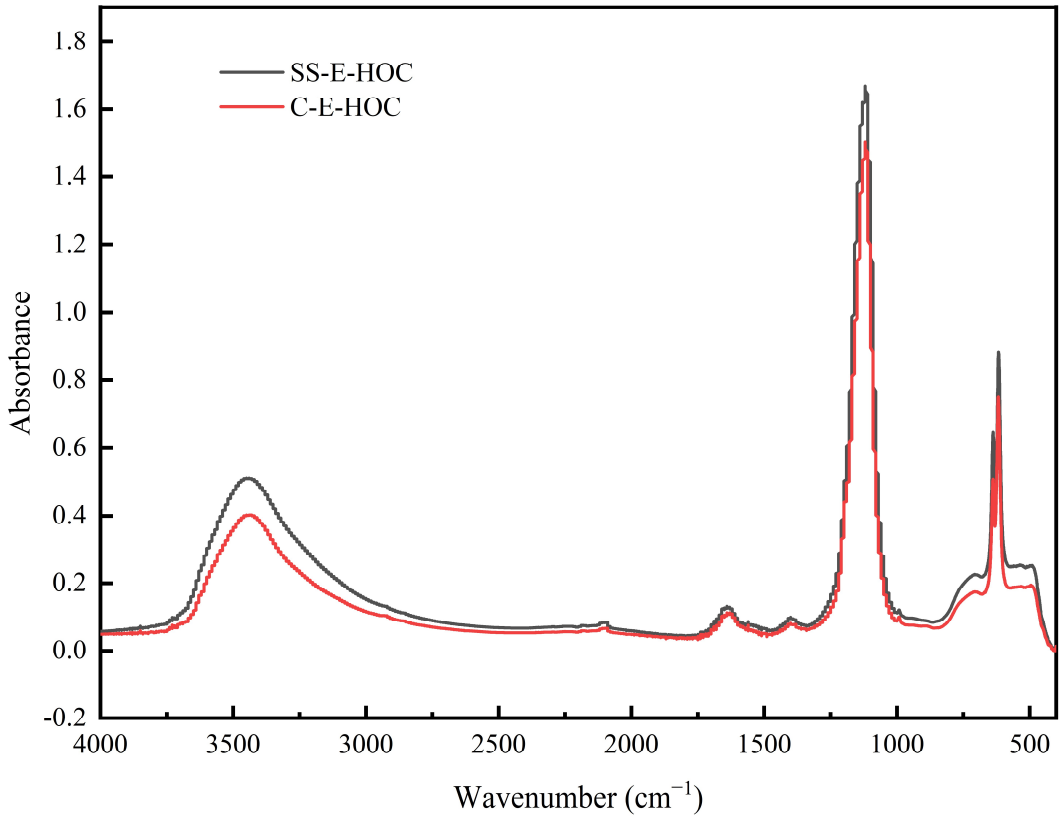

Figure 9. FT-IR spectra of flocs in the reaction system of washing wastewater treatment by E-HOC process under different cathode electrode conditions.

Comparing the FT-IR spectra of the flocs between the carbon fiber cathode system and the stainless steel cathode system, the peaks of both basically coincided, but the peaks in the carbon fiber cathode system was a little lower than that in the stainless steel cathode system. In a previous study, the O_3 addition during the EC process decreased the peak intensity of hydroxyl groups on the surface of the aluminium salt coagulants generated in situ at the anode, indicating that there is an interaction between the surface hydroxyl groups and O_3, and that there is a complexation between the coagulant and the surface hydroxyl groups of organics in the system [88]. Since the carbon fiber cathode system showed higher removal efficiency for the contaminants than the stainless steel cathode system with a large consumption of reactive oxygen species, the intensity of the peaks would be lower than that of the stainless steel cathode system.

4. Discussion

The removal efficiency of COD_{cr}, turbidity and LAS in the C-E-HOC process is higher than that in the traditional electrocoagulation process and the ozonation process. The optimal reaction conditions of C-E-HOC for washing wastewater are a current density of 15 $mA \cdot cm^{-2}$ and an ozone dosage of 66.2 $mg \cdot L^{-1}$. The removal efficiency of microplastics by the C-E-HOC process is slightly lower than that by the EC process because ozone aeration causes the large flocs to break down, causing a portion of the microplastics to fall off from the flocs. In addition, 73.8% of the microplastics were shaped as fibers, and nearly half of the microplastics were less than 50 μm in size. In the process of household laundry, 440,000 to 1,080,000 microplastics will be released to the environment for per 100 L of water. According to FT-IR analysis, the microplastics in laundry wastewater are mainly polyethylene, nylon and polyester. By comparing the E-HOC processes of different types of

cathodes, the C-E-HOC process generated more •OH during the reaction compared with SS-E-HOC. In addition, more H_2O_2 could be generated from the carbon fiber cathode in the system, and H_2O_2 would be further converted to •OH.

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