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Review

A Review of Treatment Technologies for Perfluorooctane Sulfonate (PFOS) and Perfluorooctanoic Acid (PFOA) in Water

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Abstract: Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) are a category of persistent, non-degradable pollutants that are widespread in the environment and in humans. They have attracted considerable attention due to their high bioaccumulation, multiple toxicities, long-term stability, and, in particular, their effects on human health. Therefore, there is an urgent need for highly efficient technologies and systematic mechanisms for the degradation of PFOS and PFOA. Therefore, we summarize four mainstream technologies for the degradation of PFOS and PFOA in water and their research progress in this review, namely adsorption, advanced oxidation processes, microbial treatment, and membrane separation. Among them, adsorption technology is the earliest and relatively mature, the advanced oxidation process has relatively high treatment efficiency, there are deep and broad development prospects for microbial treatment in the future, and membrane separation technology can recycle raw materials and help to save resources. Based on the analysis of the advantages and disadvantages of the existing technologies, the development prospect of reasonable optimization technology is prospected, with a view to providing a practical scientific reference for the research and engineering practice of PFOS and PFOA pollution control in China.

Keywords: PFOS; PFOA; adsorption; advanced oxidation processes; microbial treatment; membrane separation

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1. Introduction

Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) are commonly considered to be the most representative and widely used perfluorinated compounds (PFCs), while their structures are as we see in Figure 1 [1].

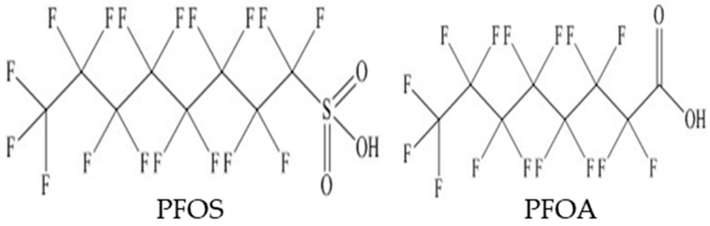

Figure 1. The structure diagram of PFOS and PFOA.

PFCs refer to C-F bond compounds formed by replacing all the hydrogen atoms connected to carbon in organic compound molecules with fluorine atoms. The general formula is $F(CF_2)_n$-R, in which R represents a hydrophilic group (such as sulfonic acid group, carboxylic group, etc.) [2]. Since their discovery by 3M Co. in the 1950s, PFCs are known for their excellent thermal stability, surface activity, and hydrophobic and oil-repellent properties, and as such have been employed worldwide in many areas including chemical plating, paper making, textile, food, leather and fire-fighting foam [3–7]. Furthermore, because of having the best surfactant characteristics and lowest surface tension, PFOS and PFOA are applied in fields closely related to humans, such as coating additives and non-sticking utensils [8,9]. It is found to be widely distributed on land and in water owing to its excellent chemical stability and high solubility in water [10]. It is reported that their concentrations typically vary from ng/L to µg/L in aqueous environments [11]. They are also found in plants, wildlife, and human blood samples [12–14].

PFOS and PFOA are also extremely widely distributed worldwide. North America was one of the first regions to be concerned about PFOS and PFOA contamination, with areas currently tested and reported on involving at least rivers in the New York area of the USA, the Tennessee River system, marine waters in the Sarasota Bay area of Florida, marine waters in Charleston Harbor, Virginia, the Lake Winnipeg and Lake Manitoba areas of Canada, drinking water, sewage in cities in the USA and Canada, etc. Loewen et al. [15] analyzed fluoride contamination in stormwater from Lake Winnipeg and Lake Manitoba in Canada and found an average PFOS concentration of 0.59 ng/L, but no PFCAs were detected. Sinclair et al. [16] investigated PFOS-like contaminants in rivers in the New York area and confirmed the presence of contamination. Later it was also found that PFOA concentrations in wastewater treatment plant effluent ranged from 58–1050 ng/L and PFOS concentrations ranged from 3–68 ng/L. PFOS and PFOA contamination testing in the European region covers a wide range of major river systems and bays in Europe, such as the Rhine and Ruhr in Germany, the Po in Italy, and the Nordic region, where PFOS and PFOA concentrations generally range from a few nanograms to several hundred nanograms per liter of water, with high levels reaching several thousand nanograms. Skutlarek et al. examined PFAs in shallow surface water and drinking water from the Rhine and Ruhr River systems in Germany [17]. The upper reaches of the Ruhr reached 446 ng/L and the main tributaries up to 4385 ng/L, with PFOA as the main component. PFOA concentrations in drinking water samples from related water systems were approximately 519 ng/L. The levels of PFOS and PFOA contamination in the Jinzu River, Tama River, Yodo River, and Tokyo Strait have been tested in Japan and, with the exception of a very few locations where the levels are abnormally high, they generally range from a few ng to over 100 ng per liter of water, which is lower than in Europe and the USA. Saito et al. detected the concentrations of PFOS in 142 shallow groundwater from different sources, including the Jinzu River, Tama River, and coastal areas in Japan, using an established LC/MS technique [18]. The results showed that the river samples averaged 2.37 ng/L, the median value was 1.68 ng/L, with a range of 0.3–157 ng/L; the geometric mean value for coastal water samples was 1.52 ng/L, the median value was 1.21 ng/L, with a range of 0.2–25.2 ng/L. Most of the samples were found to be lower than the US data at that time, except for the Jinzu River. The PFOS concentrations in the Jinzu River and the Tama River were higher, at 13,510 ng/L and 157 ng/L, respectively, and these two rivers are densely inhabited. The PFOS concentrations in the three rivers used as drinking water sources, including the Tama River, ranged from 13.0 to 38.5 ng/L (Ara River), 0.7 to 157.0 ng/L (Tama River), and 0.9 to 27.3 ng/L (Yodogawa River), respectively. In recent years, with the deepening of research on PFOS and PFOA, people have gradually realized that they are not only accumulating in living organisms, triggering biotoxicity and having a highly negative impact on the organism's liver, immunity and, above all, fertility, but are also potentially carcinogenic [19]. Hence, numerous countermeasures are already being implemented. As early as 2000, the United States Environmental Protection Agency announced the prohibition of PFOA and PFOS to avoid environmental pollution and potential health risks [20]. PFOS and its salts were added to Annex B of the Stockholm Convention in 2009 and amended in 2019; meanwhile, PFOA and its salts had been added to Annex A in 2019 [21,22]. In 2018, the European Food Safety Agency issued tolerable weekly intakes (TWIs) for PFOS (13 ng/kg) and PFOA (6 ng/kg). However, the quantities in the actual survey exceeded the TWIs [23]. The European Commission has announced restrictions on the content of four “permanent chemicals” in food that pose a threat to human health from 2023, including PFOS and PFOA. In most developed countries, the manufacture and application of PFOS and PFOA were limited or eliminated, but they and their related substances are still widely produced and applied in China due to the lack of cost-effective alternatives. It is estimated that from 2003 to 2011, the cumulative historical production of PFOS-related compounds in China was close to 1800 tons, the industrial emissions of PFOS may be underestimated because of the lack of sufficient detailed information on PFOS consumption in China [24]. The concentrations of PFOA and PFOS in the seven major aqueous systems in China are listed in Table 1 [25–31].

Table 1. Concentrations of PFOA and PFOS in seven major aqueous systems in China.

Location	Concentration (ng/L)		Data Sources
	PFOS	PFOA	
Pearl River	0.90–99	0.85–13	[25]
Yangtze River	0.01–14	2.0–260	[26]
Huaihe River	4.7	18	[27]
Yellow River	82.3–261.8	-	[28]
Haihe River	2.0–7.6	4.4–42.0	[29]
Liaohe River	0.089–9.5	4.38–77.01	[30]
Songhua River	0.06–8.04	0.02–2.68	[31]

Although these policies targeting PFOS and PFOA are helpful to reduce their presence in the environment, the potential threats they pose to the ecosystems and biological survival and health remain. As a result, further research on treatment technologies to remove PFOS and PFOA from the aquatic environment is of increasing urgency.

Previously, the literature had only reviewed source removal techniques or redox degradation techniques for PFOS and PFOA, but it is obvious that this is not enough from the point of view of the complexity of PFOS and PFOA as well as the comprehensive demand for treatment technologies. Therefore, in this paper, we provide a comprehensive review of PFOS and PFOA treatment technologies and summarize the main removal technologies for PFOS and PFOA, not only the degradation technologies, but also the adsorption technologies, biological treatment technologies, and membrane separation technologies. We analyzed their removal effects and principles in detail and introduced the current research progress. At length, the outstanding issues and future challenges are prospected based on summarizing their advantages and disadvantages, with a view to providing practical scientific references for the pollution control research and engineering practice of PFOS and PFOA in China.

2. Existing Technologies

2.1. Adsorption

Adsorption is a kind of effective technology that applies carbon materials, mineral materials, metal oxides, ion exchange resins, and other adsorbents to selectively adsorb PFOS and PFOA to remove them in wastewater [32]. The nature of the adsorbent and its substituents has an important influence on the adsorption on porous surfaces, not only the concentration of the adsorbent affects its adsorption capacity, but also the removal mechanism of the adsorbent varies depending on the type of functional group. Table 2 shows the adsorption efficiency of some adsorbents for PFOS and PFOA [33–36].

From Table 2, adsorbents of different material types generally have good adsorption effects. It can be found that the same adsorption material has different adsorption effects on different types of PFCs, which may be due to the different spatial structures of different types of PFCs, and the different number of adsorption sites contacted during the adsorption process, leading to different adsorption effects. In addition, the hydrophobic interactions between different types of molecules vary, which has a certain impact on the adsorption effect. It can also be seen that for the same PFCs, different adsorbents have different adsorption effects. This may be due to the fact that adsorption occurs not only through ionic interactions between PFCs and the functional groups of the adsorbent material but also through multi-layer adsorption mechanisms, with different adsorbents of different materials having different adsorption mechanisms.

Table 2. The adsorption efficiency of different adsorbents for PFOS and PFOA.

Absorbent Materials	Types of Adsorbed Substances	Initial Concentration/(mg/L)	pH	Adsorption Capacity/(mg/g)	Data Sources
Powdered activated carbon	PFOA	20–300	5–7	175–524	[33]
Anion exchange resins	PFOS PFOA	20–400 20–250	3–5 5	210–2575 1206	[34]
Non-ionic resins	PFOS PFOA	0.01–5 0.01–5	6.4–6.9 6.4–6.9	37–41 37–41	[35]
BSSOs	PFOS PFOA	100–400 100–400	7 7	23–907 421–846	[36]

2.1.1. Activated Carbon (AC)

Due to its developed pores, excellent adsorption performance, high mechanical strength, and easy regeneration, AC adsorption has been proven to be an efficient and feasible method for removing PFOS and PFOA in an aqueous solution [37]. The adsorption of ordinary activated carbon has been discussed extensively. By far the best removal method in terms of both economic efficiency and removal effectiveness is GAC adsorption [10]. Nevertheless, it generates solid waste and is less efficient in the presence of other organic pollutants typically present in wastewater. Therefore, the main research is to improve the performance of activated carbon modified by compounds to remove PFOS and PFOA more effectively. Meng et al. [38] prepared ultrafine magnetic activated carbon (MAC) composed of Fe_3O_4 and powdered activated carbon (PAC) by ball milling. The best adsorption of PFOS and PFOA was achieved by MAC when the mass ratio of Fe_3O_4 and PAC was 1:3, and the adsorption capacity could be realized at 1.63 and 0.90 mmol/g, respectively. The waste MAC could be detached by using a magnet, and only a small amount of methanol was used for regeneration, avoiding environmental pollution. The regenerated MAC had good reproducibility and could be operated more than five times at most. Du et al. employed homemade bamboo-based activated carbon to remove PFCs from perfluoro octane sulfonyl fluoride laundry wastewater at pH 2–9 [39]. Through experiments, they found that the removal of PFOA decreased rapidly as the pH increased from 2 to 4, but when the pH was higher than 5, it tended to stabilize, which may be because pH affected the existing forms of bamboo-based activated carbon and PFCs in water. As pH increased, the electrostatic attraction between PFCs and activated carbon weakened, and hydrophobic interactions gradually played a dominant role in the adsorption process. Numerous studies have shown the adsorption efficiency of PFCs reduced with increasing pH, and the removal effect is better under acidic conditions. Tang et al. [40] investigated the removal effect of ZnO-coated activated carbon (ZnO/AC) on PFOS and PFOA in the effluent from a wastewater treatment plant. The results showed that the removal of PFOS and PFOA reached 82.1% and 86.5%, respectively, and the removal order was positively correlated with the length of the C-F chain. The kinetic investigations indicated that the removal model for PFCs was generally elucidated through the Lagergren quasi-second-order kinetic model. In addition, the saturated ZnO/AC still retained excellent performance after ultrasonic regeneration for 3 h, which was sufficient to prove that it was a highly feasible removal technique.

The current application of AC adsorption is more comprehensive, not only for the removal of PFCs in drinking water, but also developed for the removal of PFCs in groundwater technology, the shortcomings are the simultaneous removal of various PFCs when the removal of short chains is not effective, and the regeneration of activated carbon is not as good as the resin, the use of organic solvents regeneration required when the cost is high, so the future should focus on the study of some new methods of regeneration of activated carbon and treatment of highly polluted water containing PFCs.

2.1.2. Mineral Materials

Research of minerals with rich pores and large specific surface area used as adsorbent have been taken for a long time. In spite of the electrostatic attraction, the adsorption of PFOS and PFOA by mineral materials is also characterized by hydrogen bonding force and surface complexation, which together contribute to the excellent effect of mineral materials [41]. Zhao et al. [42] researched the adsorption effect of PFOS and PFOA on montmorillonite, kaolinite, and hematite. The sorption on the minerals followed an order of hematite > kaolinite > montmorillonite, which was due to the weakening of electrostatic attraction, the increase in electrostatic repulsion and solution pH, resulting in the weakening of mineral adsorption capacity for PFOS and PFOA. In addition to electrostatic attraction, other interactions (hydrogen bonding and surface complexation) also significantly affect the adsorption of PFOS and PFOA on minerals.

Ordonez et al. [43] investigated the efficiency and mechanism of PFOS and PFOA removal from surface water employing two types of filtration media, both iron filings-based green environmental media (IFGEM) and sand-based filtration media (CTS) consisting of clay, tire crumb, and sand, through a dynamic fixed bed column. The findings showed that IFGEM had a larger adsorption area, faster rate, and greater capacity than CTS and was therefore superior to CTS in the removal of PFOS/PFOA. Both media were more efficient in the removal of PFOS than PFOA due to the better affinity between Al_2O_3 and PFOS functional groups; furthermore, the adsorption of PFOS was influenced by the hydrophobic and electrostatic interactions. The hydrophobic and electrostatic interactions with the media components governed the removal of PFOS. In addition, the excellent adsorption effect of IFGEM may be interpreted by the higher iron content. The adsorption behavior of PFOS and PFOA on sediments obtained from river systems was investigated by Fagbayigbo et al. [44]. The sediment was applied as an adsorbent material after a series of pre-treatments such as drying and sieving. Experimental data were obtained by removing as much of the diffusion effect as possible. The time required for equilibrium was between 200 and 400 min for the compounds. The contents of PFOS and PFOA in the sediments ranged between 0.50–248.14 ng/g dw and 30.01–191.96 ng/g dw, respectively. Physical and chemical indicators of the sediment, such as the size of the sediment, could significantly affect the efficiency of the sediment adsorption of PFOS and PFOA. Moreover, the adsorption behavior of PFOS and PFOA was significantly affected by the decrease in solution pH and the increase in Ca^{2+} concentration.

Due to the large dosage and long adsorption equilibrium time of mineral materials, the adsorption of PFOS and PFOA by mineral materials is currently limited to the laboratory research stage and is still far from practical large-scale application. However, studying the adsorption of mineral materials helps to understand the distribution and transport of PFCs in groundwater when minerals are encountered in practical situations. Mineral adsorption still has broad research prospects.

2.1.3. Anion Exchange (AE) Resin

Compared with activated carbon, which can be more efficient in the removal of long-chain PFCs, the advantage of AE resin is that the removal efficiency is not so reliant on the length of the carbon chain and the treatment efficiency is higher regardless of the length of the carbon chain; for this reason, the application range of AE resin is wider [45]. Otherwise, the treatment effect of various AE resins on different PFCs varies significantly because of the physical and chemical indexes of different resins and the characteristics of different polymers such as matrix and functional groups [46]. The competitive adsorption of AE resin IRA910 on PFOS and PFOA and the effect of co-existing compounds on their adsorption were studied by Maimaiti et al. [47]. Although the IRA910 had a high adsorption capacity for PFOS and PFOA, which can reach 3.5 mmol/g, their competitive adsorption was completely different. The differences in hydrophobicity and functional groups between the two resulted in an order of PFOS > PFOA. The non-ionic organic compounds had no interference effect on adsorption, while ionic ones could reduce their adsorption capacity and their influence was concentration dependent. There were multiple effects in the adsorption process, such as diffusion, competitive anion, screening effect, salting-out effect, etc. The literature data indicate that IX isotherm uptake capacity for PFOS and PFOA can reach 5 mmol/g on marketable IX resins such as IRA 958 and IRA 67 [48]. Dixit et al. [49] investigated the application of a strongly basic AE resin to remove PFOS and PFOA for water reuse and drinking applications. In addition to the concentration of PFOS and PFOA and resin dosage affecting the adsorption effect, the background organic matter (OM) properties, more specifically the charge density and molecular weight distribution of OM in the source water, were another factor attracting attention. Furthermore, the AE resin was capable of removing more than 60% of dissolved organic carbon (DOC) while removing PFOS and PFOA completely.

So far, the application of AE resin adsorption is more comprehensive, including not only the removal of various PFCs in simulated wastewater, but also in drinking water sources, especially industrial wastewater, and the adsorption capacity is large and the regeneration research is extensive, but the regeneration of the resin often requires the use of organic solvents such as methanol, which is not very feasible if applied from an economic point of view, so future research is still needed.

2.1.4. Carbonaceous Nanomaterials (CNMs)

The advantages of CNMs are their large specific surface area, the excellent adsorption capacity for both anions and cations as well as their thermal stability [50]. CNMs can break through many limitations related to their corresponding bulk materials due to their special properties at the nanometer level [51]. Therefore, CNMs are free from the constraints of traditional adsorption methods, such as regeneration of adsorbents, harmless disposal, low adsorption efficiency, etc. Not only that, CNMs can be optimized by various techniques to improve their physical and chemical indexes, thus enhancing their adsorption efficiency [52]. Therefore, it had great application prospects for rapid and efficient treatment of wastewater containing PFOS and PFOA, however, their large-scale release into the natural environment causes enormous negative impacts on ecological stability and human health if they are not used properly. As a consequence, the development of a facile process to separate CNMs from pretreated solutions should be aimed at [50]. Li et al. [53] explored the effect of multi-walled carbon nanotubes (MWCNTs) grafted with different metal nanoparticles on the adsorption of PFOA. By analyzing the adsorption of PFOA at low concentrations, it was found that the adsorption capacity of MWCNT-carrying copper nanoparticles increased 17 times compared with the original, which was attributed to the enhanced ligand exchange capacity and electrostatic forces.

In addition, studies on the removal of PFCs by adsorption using CNMs are often limited to laboratory-prepared water, with little research on actual water bodies. Competitive adsorption of PFCs with different carbon chain lengths is not sufficiently studied, and efficient regeneration of CNMs is rarely mentioned, and future research should focus on these aspects.

2.2. Advanced Oxidation Processes (AOP)

AOP is a treatment process based on producing strong oxidants (such as hydroxyl radical, ·OH) with sufficient concentration in situ to effectively treat water. The hydroxyl group is one of the strongest oxidizing agents (E_0 = 2.33 V) while it is also the radical with the best reactivity and the most applications. It can react with a wide range of organic molecules and the reaction rate constant can reach 10^6~10^{10} mol/ (L · s). ·OH can be generated in the reaction process using oxidant (O_3, H_2O_2), radiation (UV, ultrasound, microwave γ-ray, and accelerated electron beam). Table 3 lists recent studies of AOP for real-world wastewater treatment, where electrochemical treatment is less influenced by other substances in the water, photocatalytic oxidation is more influenced by other organic substances due to the presence of catalysts, and ozone oxidation is more demanding on the treatment environment but has high removal rates [54–60]. Although the quality of the actual wastewater is very different from that of the laboratory water, it was found that the actual wastewater treatment also conforms to primary kinetics. In the practical application of advanced oxidation for the removal of perfluorinated compounds from wastewater, the appropriate method should be selected according to the influence of the type of ions and organic matter in the water on the removal rate. The four technologies applied to generate ·OH from AOP to remove PFOS and PFOA are described below.

Table 3. Removal efficiency of PFCs from real wastewater by different AOP.

Treatment Methods	Wastewater Sources	Wastewater Quality	Removal Efficiency (%)	Data Sources
EO (Nano ZnO as electrodes)	Wastewater treatment plant effluent.	Large range of PFCs, low levels of TOCs, etc.	Partial PFCs: 60.	[54]
EO (Ti/RuO_2 as anodes)	Contains AFFFs Fire base groundwater.	High sulphate concentration, contains many PFCs.	PFOS:90, PFOA:90.	[55]
EO (Ti_4O_7 as anodes)	Ion exchange resin regeneration waste liquids.	High TOC/chlorine contents.	PFOS:100, PFOA:100.	[56]
EO (BDD as anodes)	Underground water.	Low TOC content, neutral.	PFOA:90.	[57]
	Industrial wastewater treatment plants.	Wide range of PFCs, high content, neutral.	PFHpA:90, PFHxA:99, PFPeA:97, PFBA:75.	
	Waste leachate.	High COD/ammonia/chlorine content.	PFOS:75, PFOA:80.	
UV/PS photocatalytic oxidation	Sewage treatment plants.	-	PFOS:50.	[58]
Ozone oxidation	Semiconductor industry wastewater.	-	PFOS:100, PFOA:92.	[59]
Ultrasonic oxidation	Groundwater at landfills.	Contains small amounts of organic compounds, neutral.	PFOS:90.	[60]

"-" indicates that no data were obtained.

2.2.1. Electrochemical Oxidation (EO)

EO can produce strong oxidizing hydroxyl radicals in aqueous solution at the condition of high current density and close to ambient temperature and pressure [61]. The anode material is the main factor that affects the oxidation effect. It is necessary to employ anode electrode materials having high oxygen evolution potential and stable performance, or a large number of compounds containing oxygen will be precipitated, leading to reduced current efficiency and ineffective function [62,63]. Zhang et al. employed aluminum, stainless steel, titanium, and titanium coated with nano zinc oxide as anodes to construct an electrochemical system to remove PFCs from the sewage of the Beijing Miyun Sewage Treatment Plant [54]. The results showed that nano zinc oxide improved the degradation efficiency of the electrochemical reactor for PFCs in wastewater, becoming the best among the four anode materials, and the removal rate of some PFCs could reach 60% or more, which is consistent with the experimental results obtained from simulating wastewater in previous experiments. The degradation of PFOS and PFOA in soil under low voltage direct current conditions was researched by Hou et al. [64]. About 33% and 51.7% of PFOS and PFOA were decomposed under conditions of 24 V and 467–690 mA, respectively. Moreover, the fluoride removal rates reached 23% and 44.7%, respectively, and no intermediates were observed, e.g., short-chain perfluoro carboxylic acids, which indicated that PFOS and PFOA were removed through their destruction. The large amount of electron migration caused the solid phase PFOS and PFOA to be destroyed and entered the aqueous phase. This significant discovery could help to accumulate, attract, and degrade PFOS and PFOA in contaminated soil. Hwang et al. [65] performed the removal of PFOS and PFOA by poly (acrylic acid) (PAA)/poly (allylamine) hydrochloride (PAH) hydrogel fibers EO electrodes embedded with silver (Ag) and gold (Au) nanoparticles (NPs). Compared with the ceramic nanofiber membrane (CNM) electrode, the NP-loaded PAA/PAHs showed better charge transfer performance and faster PFAS removal rate, which may be attributed to its denser pores and larger area–volume ratio. The removal rates of PFOS and PFOA by Ag/Au-PAA/PAH electrodes were 91% and 72%, respectively, and the removal rates of PFOS and PFOA reached 0.0093 min^{-1} and 0.0046 min^{-1}, respectively, which were the highest among the PAA/PAH electrodes loaded with NPs. The rapid removal of PFOS may be explained by the stronger redox reaction of $SO_4^{\bullet-}$ than the electron transfer of ·OH or the redox reaction of the sulfonate. Overall, the NPs-embedded hydrogel nanofibers were a type of catalyst for electrochemical oxidation with high efficiency, promising applications, and environmental sustainability. Trzcinski et al. investigated a combination of both graphite intercalation compounds (GIC) adsorption and EO techniques for the removal of PFOS [66]. When the two techniques were combined, 99% of PFOS was removed while having a half-life of 15 min. If adsorbed on GIC alone, the removal efficiency is only 42–68%. This study may open a new way of thinking about the use of EO technology not only as a single technique but also as an adjunct to other techniques to improve the removal of PFOS and PFOA.

Despite the complexity of the actual wastewater quality, the above studies have shown that the EO method can completely remove some PFCs from the wastewater with high treatment efficiency, and the removal process follows primary kinetics, which is consistent with the results of previous laboratory water distribution studies, but with high energy consumption.

2.2.2. Fenton Oxidation

Fenton oxidation, in which the interaction of Fe^{2+} and H_2O_2 and the generation of hydroxyl radicals in several continuous reactions, has aroused the enormous interest of researchers for decades due to its potential in the treatment of organic pollutants [67]. It is worth noting that the reagent of Fenton works under acidic conditions. According to a large number of experiments, the removal rate is greatest when the pH is around 3~4. At this time, the organic matter degradation rate is fast and can be degraded in just a few minutes [68,69]. The main reactions of Fenton oxidation are as follows:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + \cdot OH \tag{1}$$

In addition, it also participated in another reaction:

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + H^+ + \cdot HO_2 \tag{2}$$

The ·OH alone cannot completely decompose PFOS and PFOA since the C-F bonds of PFOS and PFOA are highly stable. Nevertheless, ·OH has a strong influence on the effect of Fenton oxidation of PFOS and PFOA [70]. Decomposition of PFOA in the UV–Fenton systems was investigated by Tang et al. [71]. It was found that the removal of PFOA consisted of two steps: degradation and defluorination. In the step of degradation, PFOA was broken down at a fast rate within 1 h, as a result of the ·OH produced by the simultaneous action of Fe^{2+} and UV irradiation, leading to a degradation rate of 87.9% and a defluorination rate of 35.8%. After reaching the defluorination step (>1 h), H_2O_2 was almost completely consumed, Fe^{3+} (primarily produced by the step of degradation) and UV continuously produced ·OH to remove the remaining PFOA. The PFOA degradation rate slowly increased to nearly 100%, and the fluoride removal efficiency further reached 53.2%. Santos et al. [72] investigated the removal of PFOA using the Fenton reagent with the addition of humic acid (HA). Quantitative removal of PFOA was achieved within 100 min at 25 °C. In this process, Fenton reagent oxidized and precipitated HA while PFOA was irreversibly immobilized in the solid phase. This technique can be applied to the complete immobilization of PFOA.

Fenton oxidation is currently only at the laboratory stage due to the harsh experimental conditions and the high susceptibility to Fe^{2+} and H_2O_2 concentrations, and more research is needed in the future to apply it to real water bodies.

2.2.3. Photochemical Oxidation

After absorbing solar energy, PFCs jump to the excited state, and after a series of chemical changes, they become substances that are less harmful or even harmless to the environment, which is called direct photolysis [73]. This method will not produce secondary pollution, but the degradation of pollutants such as PFOS and PFOA is generally a slow and inefficient process due to the stability of chemical bonds. Hence, some oxidants or chemical reagents can be added to enhance the treatment efficiency, which is called photochemical oxidation [74]. Yuan et al. [75] explored the significantly accelerated defluorination of PFOA through UV/sulfite treatment (UV/sulfite–nitrate) and coexistence nitrate (20 mg/L). Just after the first 30 min, PFOA defluorination was at an initial stage with a low removal rate, which corresponded to the removal of co-existed nitrate. After the end of the initial stage (>30 min), the rate of PFOA defluorination was significantly accelerated, achieving nearly 100% defluorination within 2 h. Ultimately, the complete defluorination of PFOA and denitrification of nitrate were achieved simultaneously. In a word, the kinetics of PFOA decay, defluorination, and transformation product formations in UV/sulfite–nitrate treatment were greatly enhanced. The coupling of hydrochar (H-BC) with iodide (KI) was carried out by Hu et al. to degrade PFOS and PFOA under UV light [76]. After 60 min of photolysis, PFOS and PFOA were removed from aqueous solutions by 99.5% and 94.1%, respectively. These values were higher than the total when using UV under H-BC or KI conditions only, suggesting that the co-use of both H-BC and KI had a mutually reinforcing effect. An explanation for this can be found in the unique structure and nitrogen-containing functional groups of H-BC. H-BC was capable of reducing iodide oxidation products to I^- and absorbing PFOS and PFOA. The combination of all these factors enhanced the reaction between PFCs and hydrated electrons. Qian et al. used ultraviolet/persulfate (UV/PS) to remove PFOA from disinfected effluent from wastewater treatment plants, with a removal rate of about 67% [58]. In comparison, the removal efficiency of PFOA in surface water with low chloride ion content was found to be about 80% and the reaction time was relatively short, which was consistent with the experimental results obtained from the previous treatment of experimental simulated wastewater. Considering the effect of chloride ions on the photodegradation efficiency, chloride ion pretreatment should be carried out to eliminate the effect if this method is applied to actual wastewater treatment.

2.2.4. Ultrasonic Oxidation

The specific mechanism of ultrasonic oxidation is that the cavitation bubbles can release huge energy during the collapse process, resulting in a local high temperature and high-pressure environment (4000 K, 182 MPa) in the nearby small space, and then the H_2O can be naturally decomposed into hydroxyl radicals and active hydrogen while entering the environment [77]. Shende et al. [78] evaluated the ultrasonic degradation of PFOS and PFOA at different power densities in the presence or absence of argon. It was found that the removal of PFOS and PFOA was enhanced when the temperature of the water samples was high (30 °C). At an ultrasonic frequency of 575 kHz, the application of oxidizing agents such as iodate and persulfate could remarkably strengthen the removal of PFOA in the presence of an argon environment, but could not improve PFOS degradation, which indicated that the highly active radicals formed during the oxidation process cannot destroy the strong C-S bond of PFOS. Shende et al. [79] investigated the ultrasonic breakdown of PFOS and PFOA under conditions of different ultrasonic frequencies, pH, water temperature, and gas. The impact of different ultrasonic frequencies on the degradation of PFOS and PFOA was not remarkable by laboratory standards. When the volume of solutions was reduced from 500 mL to 200 mL for the same concentrations, the removal of PFOS and PFOA increased from 1.8 times to 4.5 times under different ultrasonic frequency conditions, with a higher increase for PFOA. Meanwhile, the degradation rate of PFOA was also higher than that of PFOS over a range of sonication frequencies, which can be attributed to the higher number of active cavities collapsing at higher temperatures. In the air environment, an increase in the ultrasonic degradation rate of PFOS and PFOA was observed at higher pH and higher bulk water temperature, while in an argon environment, a decrease in them upon increasing pH. Furthermore, the addition of extra gas decreased the degradation rates of PFOS and PFOA. Cheng et al. investigated the degradation of PFCs in landfill groundwater by ultrasound [60]. The presence of volatile organic compounds in the water samples resulted in lower liquid surface temperatures, which inhibited the degradation of PFCs. The treatment of PFOA by ultrasound/persulfate (US/PS) was studied by Xiong et al. [80]. It was observed that 100% fluoride removal was achieved after only 4 h of US treatment at a power of 900 W, i.e., complete defluorination of PFOA was achieved and its harmless end products were CO_2, H_2O, and F^- ions. PS addition, however, suppressed defluorination. A Langmuir-type adsorption model fitted by kinetics demonstrated that the addition of PS increased competition with PFOA for adsorption sites. The most important aspect of this work is that it will further facilitate the implementation of treatment technologies for PFCs in order to achieve the goal of zero fluorine pollution.

2.3. Microbial Treatment

Microbial treatment is the process of treating target degradation products by employing physiological processes such as dissolution, enzymatic hydrolysis, and phagocytosis of bacteria, fungi, and other microorganisms [81]. Huang and Jaffé [82] studied the treatment of PFOS and PFOA with the microbial strain A6 which acted via the Feammox pathway (Equation (3)). The removal of PFOS and PFOA was as high as 60% during incubation, while DOC decreased slightly as time progressed. The phenomenon indicated that some products were metabolized by other heterotrophic bacteria with the occurrence of PFOS and PFOA defluorination. Therefore, the A6 strain can be employed to remove PFOS and PFOA by applying ammonium or hydrogen as an electron donor while reducing iron.

$$6(Fe_2O_3{\cdot}1/2\ H_2O) + 20H^+ + 2NH_4{}^+ \rightarrow 12Fe^{2+} + 17H_2O + 2NO_2{}^- \tag{3}$$

Albert et al. [83] investigated the functionalization of iron nanoparticles on living diatoms (Dt) to achieve the removal of PFOS and PFOA. The degradation efficiency reached 89% and 93.7%, respectively, within 24 h. Dt provided oxygen-produced endogenous reactive oxygen species (ROS) while serving as an effective support for adsorbing solids, which together promoted the decomposition of PFOS and PFOA. Its reusability (85% decomposition efficiency was still achieved after reuse) has been proved by simple magnetic separation. Current results also indicated that these functionalized diatom cells performed consistently and sustainably, especially since no cytotoxicity was observed. The diatom-assisted bioremediation strategy demonstrated the potential and promise of green and sustainable environmental technologies to tackle pollution by PFCs, while the used-up materials may be applied to the development of agriculture.

Wang et al. investigated the removal of PFCs by different treatment processes in four different wastewater treatment plants in Liaoning Province [84]. The results showed that the removal of PFCs in the four WWTPs mainly depended on activated sludge adsorption, which was not closely related to the specific microbial treatment process. Among them, the removal rate of short-chain perfluorinated compounds was up to 85%, while the removal rate of long-chain perfluorinated compounds was less than 10%.

2.4. Membrane Separation

The mechanisms of removal of PFOS and PFOA by polyelectrolyte (PE) functionalized UF membranes were investigated by Olimattel et al. [85]. It was found that this treatment led to a reduction in membrane molecular weight cut-off (MWCO, defined as the molecular weight of a solute with a retention rate of at least 90%, the standard unit of measurement is Dalton) and porosity of about 38% and 9.2%, giving an increase in the removal of PFOS and PFOA of about 30%, with size exclusion being the main reason. The synergistic effect of cations and HA led to a remarkable increase in the removal of PFOS and PFOA, including a maximum of 23%, which was due to the increase in the size of the macromolecular complexes and the enhancement of electrostatic repulsion resulting from the interactions of PFOS and PFOA with the solutions and UF membranes. The removal rate was further increased by 14% when the cation concentration was increased to twice the original concentration, however, an increase in HA concentrations had no significant effect. Wang et al. [86] researched the removal of PFOA through the RO membrane. It was found that the removal of PFOA was more than 99% at 1.5 MPa with a permeate flux of 55 $L/m^2 \cdot h$ from the RO membrane. In addition, the retention rate remained stable after the concentrated solution of PFOA was treated by a multistage reverse osmosis unit. The regeneration capacity of the RO membranes could still reach 85% after physical rinsing. The high removal of PFCs by polyamide membranes in reverse osmosis membranes, which could exceed 99%, was due to the ability of polyamides to form strong hydrogen bonds with them [87]. The removal of fluorinated compounds by RO membranes is due to the multiple mechanisms of dissolution–diffusion mechanisms, electrostatic repulsion, and pore rejection, due to the fact that PFOA has highly linear molecules with extremely small molecular sizes compared to aromatic and aliphatic organic compound molecules. Therefore, the shape of the molecule determines the pathway of the membrane, and the more densely structured RO membranes can be greater in removing PFOA [88,89].

The essence of the membrane separation process is to use physical methods to treat PFCs in water, which does not affect the structure of the contaminants. It can also recycle raw materials while purifying wastewater. However, the membrane separation method is prone to membrane fouling in practical applications, which will reduce the treatment efficiency. Therefore, it can be applied together with other treatment methods to treat industrial wastewater. In addition, the actual industrial wastewater has complex components, and the dissolved organic matter may affect the membrane flux in terms of the large particle sizes. Consequently, pretreatment should be performed to improve the removal rate of PFCs in practice.

3. Discussion and Perspectives

To this end, we have summarized the degradation principles and research cases of PFOS and PFOA from the aspects of adsorption, advanced oxidation process, microbial treatment, and membrane separation technology, all of which own their pros and cons.

For adsorption, an amply studied process and the most used remediation strategy have been applied in the field and it is a highly efficient and cost-effective removal technology. At the same time, it is low in consumption, simple to operate, does not generate excess by-products, and is easier to manage in practice in water plants. However, its treatment of PFCs only transfers from one medium to another, failing to fundamentally disrupt the C-F bond and degrade it and it treats smaller volumes of water with higher losses. The future research focus should be on how to improve the recycling rate of adsorption materials, and how to safely release adsorbed PFOS and PFOA to avoid secondary pollution.

For advanced oxidation processes, the efficiency of removing PFCs is relatively high, with a maximum of 99%; furthermore, the reaction time is short, and it is easy to automate and control in the actual operation of the water plant. However, not only are the operations complex, energy consumption is high, certain raw materials such as electrode materials are expensive, and overall operating costs are high, but also the removed portion of PFOS and PFOA only has a shorter carbon chain length, and some still have similar toxicity, which means that the defluorination rate is not high and the degradation products still require further treatment. Therefore, how to improve technology to improve the defluorination rate still needs further research in the future. In addition, many processes, such as ultrasonic oxidation, only stay at the laboratory scale, and further research is needed to assess the feasibility of these technologies under larger and more challenging conditions.

For microbial treatment, the degradation cycle is long and incomplete, and some intermediate products have high toxicity. The degradation mechanism is still under further study. Currently, the removal of PFCs in actual wastewater mainly depends on the adsorption of sludge. Therefore, this technology is not mature yet, and there are still many difficulties in the biodegradation of perfluorinated compounds. Based on the previous studies, how to further elucidate the biodegradability of PFCs mechanistically and to improve their biodegradation rate will be a key issue to be addressed in subsequent studies.

For membrane separation, there is no impact on the structure of pollutants during treatment, the fluoride removal is complete, the quality of the effluent is high and raw materials can be recycled while purifying wastewater. However, in practical applications, membrane pollution can easily occur, resulting in reduced treatment efficiency. Therefore, industrial wastewater can be treated in combination with alternative technologies. In addition, the actual industrial wastewater has a wide variety of compositions as well as the dissolved organic matter may affect membrane flux, requiring pretreatment to reduce the impact of other organic substances. For the daily operation of water plants, the running costs are high, and the average water production rate is low, with 20–30,000 tons of water being discarded for a 30,000 tons per day water plant. Therefore, membrane separation is currently only suitable for industrial ultrapure water production and bottled water, not for large and medium-sized water plants. Overall, in terms of efficiency, the advanced oxidation process has the highest efficiency in removing PFOS and PFOA, which can reach 99%, followed by adsorption, followed by membrane separation technology, with microbiological treatment being the least efficient. Of course, this is not absolute, different processes correspond to different treatment targets, and different categories of treatment within the same process can produce different results. PFOS and PFOA are extremely stable chemicals that do not undergo hydrolysis, photolysis, or biodegradation under typical environmental conditions. Therefore, it persists in the environment and is difficult to repair. In recent years, the pollution problem of PFOS and PFOA has become increasingly prominent. The technology for removing them from water has attracted worldwide attention and many new technologies or combination technologies have been developed. Based on the above advantages and disadvantages, future work should be carried out in the following aspects:

1. Up to now, most of the studies on removal technologies for industrial wastewater have only observed the changes and removal rates of PDCs throughout the process. However, in practical applications, not only the removal of other pollutants from wastewater by the process should be studied, but also the impact of treatment technology on wastewater quality should be fully considered.
2. Considering the advantages and disadvantages of different technologies, multiple technologies can be considered for coupling in the future to achieve the effect of improving processing efficiency or reducing side effects.
3. Due to the late start of research on PFCs in China, the research on PFCs removal by Chinese scholars is relatively limited. Most of the removal research is also limited to common PFCs such as PFOS and PFOA. Next, we can increase the research on the removal of other types of PFCs.
4. With the development of industry, the environmental pollution situation is becoming increasingly serious, and groundwater has been polluted to varying degrees. However, currently, various types of research on PFOS and PFOA are concentrated in laboratory-

simulated water bodies or surface water bodies, with little research on groundwater. It is recommended to increase the detection of PFCs in groundwater, establish simple and economical PFCs detection methods, and provide research data for the treatment of PFCs in groundwater.

5. Currently, there is a huge demand for PFCs substitutes in many fields, and increasing research on PFCs substitutes can alleviate the current environmental pollution caused by PFCs.

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