water

Review

A Review of the Treatment Process of Perfluorooctane Compounds in the Waters: Adsorption, Flocculation, and Advanced Oxidative Process

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Abstract: Perfluorinated compounds (PFCs) are recognized as a new type of refractory organic pollutants. Due to the persistent environmental pollution, bioaccumulation, and biotoxicity of PFCs, they have received extensive attention in recent years. To deal with the environmental risks caused by PFCs, the pollution and distribution of PFCs in the aquatic environment are discussed in detail, mainly for the most widely used PFCs—perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS). The latest progress in the current processing technology of PFOA and PFOS is comprehensively introduced. It includes a variety of physical techniques to remove PFCs such as adsorption and flocculation. It has been confirmed that various adsorbents can play a key role in the enrichment and removal of PFCs through high specific surface area and hydrophobic interaction. In addition, traditional degradation processes are often unsatisfactory for PFCs, prompting the search for more efficient and cost-effective methods, with great progress having been made in advanced oxidation processes (AOPs) based on radical decomposition of pollutants. This review also integrates multiple advanced oxidation processes (AOPs) such as photocatalysis, electrochemical processes, ozone, the Fenton process, and ultrasound. This paper provides an overview of the various PFCs removal techniques and discusses their efficacy. It also explores future possible developments for PFCs elimination technologies for water treatment.

Keywords: perfluorinated compounds (PFCs); pollution and distribution; adsorption; flocculation; advanced oxidation processes (AOPs)

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1. Introduction

Since being developed by 3M Company in the 1950s [1], polyfluorinated compounds (PFCs) have been widely used in many fields such as chemical electroplating [2], coatings [3], textiles [4,5], leather [5], synthetic detergents [6], cookware manufacturing [7], and fire-fighting foam [8], due to their excellent thermal stability, surface activity, and hydrophobic and oleophobic properties. In recent years, with the in-depth study of PFCs, in addition to excellent chemical properties, it has been found that PFCs also have the characteristics of refractory degradation [9], long-distance migration [10], and bioaccumulation [11], and even show a variety of toxic effects [12].

If the accumulation of PFCs reaches a certain concentration threshold in animals or humans, there will be corresponding toxic effects. The resulting toxicity includes organ toxicity, neurotoxicity, immunotoxicity, reproductive toxicity, and carcinogenicity [13,14]. However, due to the need for industrial development, the production and use of certain PFCs are still increasing. Therefore, in the future, scientists will have to study the amount of pollution, how PFCs break down, and the health risks they pose in different environments.

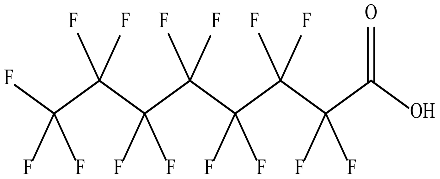
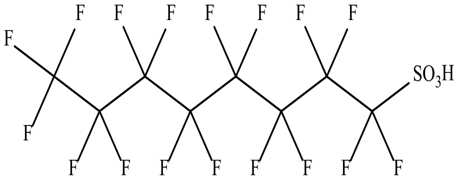
Several reviews [15–20] on the removal of PFCs by different processes have been reported. These research results include degradation and removal methods of PFCs such as adsorption [17] (activated carbon, carbon nanotubes), ion exchange resins [17], filtration [21] (RO and NF), electrochemical oxidation [18], and chemical oxidation and reduction [22]. However, despite the wide range of perfluorinated compounds discussed in most articles, there is a lack of detailed analysis of PFCs' contamination status in different regions and treatments. For example, the oxidation and reduction processes of different chemical degradations are not precise. In particular, the introduction of the flocculation process is often neglected. The effect of simultaneous mineralization and final degradation of PFCs is often suboptimal, or the treatment cost is relatively high.

This research examines the benefits and limitations of PFCs treatment. The most widely used PFOA and PFOS were selected as research targets, and the source and distribution of PFCs in the water environment from different countries and regions are discussed. The biological inertness caused by the special structural C-F bond of PFCs makes it difficult to be thoroughly mineralized by native microorganisms in nature. Currently, relevant studies have shown the feasibility of biodegradation, and the main work is the screening of dominant strains and the exploration of the metabolic degradation mechanism [23]. However, reports on the biodegradation of PFCs are still limited, with physical and chemical treatment methods having significant advantages in research depth and operational feasibility. Therefore, this paper focuses on evaluating physical treatment methods (adsorption and flocculation) and chemical treatment methods represented by advanced oxidation technology. The characteristics and treatment effects of various adsorbent materials were introduced, with adsorption mechanisms such as surface hydrophobicity and pore structure being especially analyzed. The flocculation process has not been well represented in previous reviews, although the treatment effect is remarkable, and this paper summarizes the effect and mechanism of flocculation to remove PFCs. Regardless of physical treatment methods such as adsorption or flocculation, PFCs are not fundamentally degraded, and the problem of secondary pollution still exists. Therefore, it is an important research direction to completely mineralize PFCs and gradually defluoride them. Photocatalysis, electrochemical oxidation, Fenton, and other AOPs also showed good effects on the degradation of PFCs, and the effect of free radicals played a crucial role in the cleavage of C-F bonds. In this study, the removal effects and typical degradation mechanism of various AOPs were studied, along with the impacts of various water quality characteristics and experimental conditions on the degradation process. PFCs' primary issues regarding water pollution were better defined. The effects and limits of the current methods are looked at, with suggestions made to improve the removal of PFCs or accelerate the process of degradation.

2. Distribution and Pollution of PFCs in Water Environment

PFCs are a class of highly fluorinated organic compounds in which all hydrogen atoms connected to carbon atoms in the molecule are replaced by fluorine atoms. PFOS and PFOA are the most productive PFCs. According to statistics, from 1951 to 2004, the global output of PFOA was about 3600 to 5700 t [24]; from 1958 to 2015, the emission of PFOS was 1228 to 4930 t [25]. The basic properties of PFOA and PFOS are shown in Table 1 [26]. In the molecular structure, adjacent fluorine atoms are helically distributed along the carbon chain and repel each other. The C-C bond is surrounded by a series of stable fluorine atoms, so it is not easy to break. The C-F bond is highly polar due to the high electronegativity (-4.0) and low polarity of fluorine. The C-F bond is also one of the covalent bonds with the largest bond energy in nature (the bond energy is about 460 $kJ \cdot mol^{-1}$). As a result, PFCs have high thermal and chemical stability and a more stable molecular structure [27].

Table 1. The basic chemical and physical properties of PFOA and PFOS.

	PFOA	**PFOS**
CAS number	335-67-1	1763-23-1
Molecular formula	$C_7F_{15}COOH$	$C_8F_{17}SO_3H$
Molecular Structure		
Molecular weight	414.07	500.13
Solubility	13.6 g/L	7.5 g/L
Density	$1.84\ g{\cdot}cm^{-3}$	$1.25\ g{\cdot}cm^{-3}$
Melting point	45–50 °C	≥400 °C
Boiling point	260 °C	189~192 °C
pKa	0.50	−3.27

Due to their excellent physical and chemical properties, PFCs are widely used in industrial production and daily necessities. PFCs in environmental media are difficult to degrade by physical, chemical, and biological actions, and can remain stable even under extreme conditions such as some strong oxidants and strong acids and bases. The high stability naturally also leads to the long-distance mobility of PFCs. As shown in Figure 1, a large number of PFCs have been cyclically migrated into the environment, daily necessities, and even people since the beginning of production. Currently, PFCs can be detected in air [28], water [29], sediment [30], and soil [31]. PFCs contamination has spread all over the world in various ecosystems, and they can even be detected in polar ice sheets and animal tissues [32]. Industrial, agricultural, and municipal effluent have become an important source of PFCs pollution in surface water, groundwater, and even drinking water. The production and use of PFOS and PFOA have been banned or curtailed. However, due to their large amounts of use and refractory degradation, many studies have found that PFCs pollution in the environment is still dominated by these two substances.

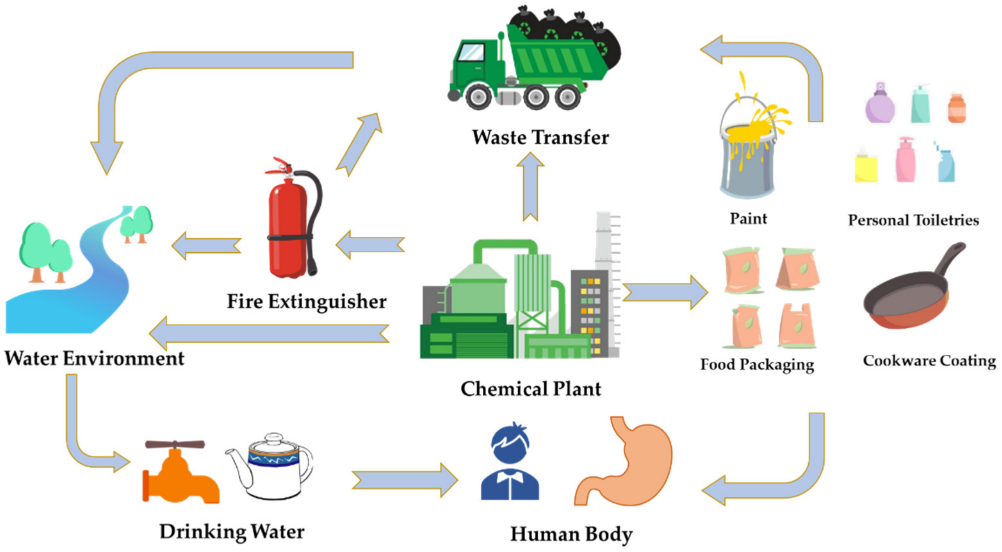


Figure 1. Migration process of PFCs in the environment.

Table 2 summarizes the concentrations of PFOA and PFOS in the water environment of some areas of the world. The detection rate of per-and polyfluoroalkyl substances (PFASs) in water samples in the United States exceeds 70%. The content of PFASs in drinking water treated by a specific process is at a low level. However, the content of perfluorohexanoic acid (PFHxA) in individual sites is still as high as 62 ng·L^{-1} [33]. In a sample analysis of major rivers in the United States from 2008 to 2019, the highest PFOA and PFOS content ranges were n.d.~204 and n.d.~297 ng·L^{-1}, respectively [34–39]. These are all below the drinking water thresholds specified by the US Environmental Protection Agency (200 ng·L^{-1} for PFOA and 400 ng·L^{-1} for PFOS) [33].

PFCs pollution is also common in China's water environment. Different concentrations of PFOA and PFOS have been detected in the Yangtze River, Yellow River, Pearl River, and other river basins where the economy is more developed. The pollution level of PFCs in some industrially developed areas is relatively high. The detection rate of PFOA in the Yangtze River water was the highest, with a concentration range of 2.0~260 ng·L^{-1} [40]. The study found that PFCs pollution in the Yangtze River mainly comes from wastewater discharged from local sewage plants [40]. The purpose of sewage plants is to reduce or eliminate pollutants in wastewater, but studies have found that the concentration of PFCs in the final discharge water of some sewage plants is still at a high level. There are high concentrations of PFOS in the surface waters of the Pearl River [40] and the Yellow River [41], with the highest detected concentrations in the Pearl River and the Yellow River being 99 and 261.8 ng L^{-1}, respectively. Likewise, untreated industrial wastewater and effluent from sewage treatment plants are considered to be the main sources of PFCs pollution in these regions.

The Bohai Sea is one of the most polluted sea areas by PFCs in China. It is in a semi-closed and semi-isolated state, and the water body has poor self-purification ability. The PFCs pollution in the Bohai Sea has regional differences, with the total concentration of PFCs detected in the most polluted areas three orders of magnitude higher than that in the least polluted areas [42]. Fluorine chemicals used in industrial parks may be the main cause of the problems in the areas listed [43]. To reduce the pollution of PFCs in the aquatic environment, it is necessary to effectively treat the industrial wastewater in the basin.

Surface water quality analysis was conducted in the estuarine area, with the concentrations of PFOA and PFOS being 2.95~68.6 ng·L^{-1} and 4.11~450 ng·L^{-1}, respectively [44]. The results of Guo also show that domestic sewage is the main source of PFCs pollution in the Korean water environment, with the PFCs in sewage being mainly PFOA and PFOS. The PFOA concentrations in the influent and effluent of the WWTP were 2.3~615 ng·L^{-1} and 3.4~591 ng·L^{-1}, respectively, and the PFOS concentrations were n.d.~68.1 ng·L^{-1} and n.d.~8.9 ng·L^{-1}, respectively [45]. The treatment at the WWTP decreased the amount of PFOS in the effluent, while the amount of PFOA increased.

Sampling and analysis of the Yodo River basin and nearby sewage treatment plants in Japan was performed from 2004 to 2005. The contents of PFOS and PFOA ranged from 0.4~123 ng·L^{-1} and 4.2~2600 ng·L^{-1}, respectively. It was found that the surface water samples of the basin and the effluent from the sewage treatment plant had the highest concentrations. This indicates that wastewater from WWTPs is an important source of PFCs pollution in local surface water [46].

Table 2. Pollution of PFOA and PFOS in the water environment in different countries and regions.

Sampling Location		**Sample Type**	**Time**	**Concentration (ng/L)**		**Refs**
				PFOA	**PFOS**	
USA	Georgia River	surface water	2008	n.d.~204	n.d.~297	[36]
	Mississippi River	surface water	2008	n.d.~125	n.d.~245	[37]
	Rhode Island	surface water	2014	0.3~56	<0.1~27.5	[39]
	Truckee River	surface water	2019	1.6~19.2	n.d.~17.4	[34]
	Tennessee River	surface water	2015	<10~120	<10~220	[38]
	New Orleans	groundwater	2010	3.900	4.025	[35]

Table 2. *Cont.*

Sampling Location		Sample Type	Time	Concentration (ng/L)		Refs
				PFOA	PFOS	
France	Orge River	surface water	2010	9.4	17.4	[47]
	Freshwater	surface water	2012	<0.08~36	<0.06~173	[48]
Spain	Llobregate basin	surface water	2010	0.07~146	0.07~27,110	[49]
Germany	Rhine upstream	surface water	2008	2.22~4.07	3.03~7.34	[50]
	Ruhr area	drinking water	2006	n.d.~519	n.d.~22	[51]
	Ruhr area	surface water	2006	n.d.~3640	n.d.~193	[51]
Canada	Ontario Lake	surface water	2006~2007	14	22	[52]
Sweden	Lake in Lulea	surface water	2015	60	-	[53]
China	Pearl River	surface water	2004~2005	0.85~13	0.90~99	[40]
	Yangtze River	surface water	2004~2005	2.0~260	0.01~14	[40]
	Huai River	surface water	2011	18	4.7	[54]
	Yellow River	surface water	2011	-	82.3~261.8	[41]
	Haihe River	surface water	2010	4.4~42.0	2.0~7.6	[55]
	Liao River	surface water	2012	4.38~77.01	0.089~9.5	[56]
	Bohai Sea	surface water	2013	n.d.~83.4	n.d.~6.80	[43]
Korea	Estuarine area	surface water	2008	2.95~68.6	4.11~450	[44]
Japan	Kamo River	surface water	2005	102	150	[57]
	Yodo River	surface water	2004~2005	4.2~2600	0.4~123	[46]
Australia	Sydney Harbour	surface water	2009	4.2~6.4	7.5~21	[58]

In addition to surface water and groundwater, PFCs contamination in drinking water is also a cause of concern. Drinking water is an important source of PFCs in the human body. Studies have shown that the level of PFOA in drinking water is directly proportional to the level of PFOA in the serum of local residents [59]. A thorough analysis of the water quality in the German Rhine River basin showed that PFCs are more concentrated in the water reservoirs. The PFOA levels in the blood of residents who drink this water source are five times higher than those in other areas [51].

3. Adsorption Process and Flocculation Process

3.1. Adsorption

Adsorption technology is an efficient and feasible method to remove perfluorinated compounds. Activated carbon, ion exchange resins, carbon nanotubes, mineral materials such as zeolites, and organic polymeric materials are often used as sorbents.

The C-F bond energy of PFCs is very high (about 110 kcal·mol^{-1}) and the chemical stability is extremely strong. Therefore, PFCs are difficult to degrade and can withstand traditional chemical action, microbial action, and their biological metabolisms [22]. This brings great challenges to the development of its control technology. At present, the technologies for effectively removing PFOS mostly focus on physical separation and chemical degradation. At the same time, new degradation technologies such as ultrasonic degradation and photochemical decomposition have also made preliminary progress. However, these technologies still need to be improved, have high energy consumption, are complicated operations, and are a potential threat of secondary pollution. Surfactants such as PFOA cannot be properly disposed of with traditional treatment methods because they use a lot of energy, are hard to run, and could cause secondary contamination.

The adsorption method is a common and important treatment method in water treatment, and has the characteristics of low cost, high efficiency, easy operation, and recyclability. A large number of scholars have conducted research on the process and mechanism of PFCs adsorption. Table 3 summarizes the adsorption performance of various adsorbents for PFCs, including their adsorption conditions and the fitting parameters of the second-order adsorption kinetics. The adsorption ability and mechanism of different types of adsorbents are not completely the same. The adsorbents will have different adsorption mechanisms for PFCs. Thus, the previously reported adsorbents for the degradation of PFCs were sorted, and the interactions and adsorption process between PFCs and adsorbents were explained.

3.1.1. Activated Carbon

Activated carbon (AC) is a low-cost, high-capacity adsorbent that is currently widely used in the removal of water pollutants. Previous studies have confirmed that granular activated carbon (GAC) and powder activated carbon (PAC) have strong adsorption capacities for PFOA and PFOS [60,61]. Yu et al. [62] compared the adsorption performance of PAC and GAC when treating PFCs. It was found that the adsorption capacities of PAC for PFOA and PFOS were 0.67 and 1.04 mmol/g, respectively, and the adsorption equilibrium time was about 4 h. The adsorption capacities of GAC were 0.39 and 0.37 mmol/g, respectively, and the adsorption equilibrium time was at least 168 h. Activated carbon's adsorption rate is affected by the size of its particles. Since PAC has a larger surface area than GAC, it is better at removing perfluorinated compounds.

The time required to reach adsorption equilibrium for PFCs treated with activated carbon is too long, while in the actual treatment process, the hydraulic retention time of GAC is limited. The lower adsorption rate makes it easy for PFOA and PFOS to penetrate the adsorption layer before adsorption saturation. It has been shown that per- and polyfluoroalkyl substances (PFASs) in drinking water can rapidly penetrate activated carbon filters, thereby hindering the removal of pollutants. Some smaller-chain PFASs (4 or 6 carbons) may pass through the GAC faster than longer-chain PFASs [63]. Activated carbon has good adsorption properties for PFOA and PFOS, and their adsorption capacities are strong. However, researchers are constantly working on the most effective techniques for recycling and reusing adsorbent materials. Future research will focus on this.

3.1.2. Resins

Compared with GAC, some ion exchange resins showed higher adsorption efficiency for PFCs. Carter et al. [64] found that the adsorption equilibrium time (10 h) of resin for PFOS was much shorter than that of traditional GAC (50 h). PFOS reached >95% of the equilibrium absorption of the resin after 4 h, while the absorption of GAC took 15 h to reach >95% of the equilibrium absorption. Zaggia et al. [65] investigated the adsorption properties of three anion resins, A532E (high hydrophobicity), A520E (generally hydrophobic), and A600E (non-hydrophobic), for PFOA and PFOS in drinking water. The adsorption equilibrium capacities of the three resins for PFOA were 142.1, 134.7, and 125.2 mg/g, and 260.5, 210.4, and 186.2 mg/g for PFOS, respectively. These data indicate that the more hydrophobic the resin, the stronger the adsorption, which is directly related to the surfactant properties of PFCs. Senevirathna et al. [66] compared the adsorption effects of GAC, ion exchange resin, and non-ion exchange polymer on PFOS and found that at a lower PFOS concentration (100 ng/L), the adsorption capacity of non-ion exchange polymer was higher than the other adsorbents. At concentrations >1000 ng/L, GAC was the best filter material for PFOS removal. Ion exchange resins, however, might be employed to remove PFOS in the range of ng/L when the adsorption isotherm and adsorption kinetics are considered.

3.1.3. Carbonaceous Nanomaterials (CNMs)

Carbonaceous nanomaterials (CNMs), especially carbon nanotubes (CNTs) and graphene, have a high specific surface area, porosity, and adsorption capacity for anions and cations. Their excellent mechanical properties, as well as chemical and thermal stability, etc., give them great application potential in environmental remediation [67]. Chen et al. [68] researched the adsorption kinetics and equilibrium adsorption capacity of PFOS on willow charcoal, corn stover ash, and CNTs. It was found that CNTs reached equilibrium within

2 h, much faster than willow charcoal (384 h) and corn stover ash (48 h). This is due to the high adsorption available space of CNTs on the cylindrical outer surface. The adsorption mainly depends on the electrostatic attraction and the hydrophobic interaction of PFCs. Bei et al. [69] investigated the adsorption mechanism of six PFCs on different kinds of CNTs. When these six PFCs share the same functional groups, the adsorption capacity of CNTs increases as the C-F chain length increases. The adsorption capacity of CNTs functionalized with hydroxyl and carboxyl groups (CNT-OH, CNT-COOH) was much lower than that of pristine CNTs. It shows that the hydrophobic interaction is dominant, while the electrostatic repulsion inhibits the adsorption of PFCs on CNTs. The fact that the rate of adsorption decreases as pH increases indicates that the nature of hydrogen-bonding interactions is not strong. CNTs' ability to bind to PFOS may be significantly influenced by the concentration of certain anions and cations in the solution.

The removal rate of PFOS by CNTs decreased by 15–30% as the concentration of anions such as SO_4^{2-}, Cl^-, and $Cr_2O_7^{2-}$ in the solution increased [70]. However, when the concentrations of Cu^{2+} and Pb^{2+} increased, the removal rate first decreased and then increased. The possible reason for the increase in removal rate was that Cu^{2+} and Pb^{2+} played a bridging role between PFOS and CNTs. In addition, divalent metal ions may be directly complexed with negatively charged PFOS. CNMs have special properties at the nanoscale, which can break through many limitations associated with them (such as high surface activity, quantum size effects, surface/interface effects, etc.). Additionally, CNMs may be functionalized and have their surface morphology modified in different ways to enhance performance. CNMs are not restricted by the limitations of conventional adsorbents (such as reuse after adsorbent regeneration, slow adsorption speed, difficult functionalization, etc.). Therefore, CNMs are essential for the fast and effective treatment of wastewater containing PFCs.

3.1.4. Minerals

Mineral materials refer to natural minerals with one or several available physical and chemical properties (or modified to achieve the above conditions). They have the characteristics of economic benefits, large reserves, variety, and wide application [71]. Johnson et al. [70] tested the adsorption behavior of PFOS using five well-characterized materials (goethite, kaolinite, Ottawa sand, iron oxide-coated sand, and lake bottom sediments). The results showed that the adsorption performance decreased with the increase in solution pH. This is consistent with the findings of Zhao [72]. Zhao researched the adsorption effects of four PFCs, including PFOA and PFOS, on montmorillonite, kaolinite, and hematite. It was found that the adsorption capacity was hematite > kaolinite > montmorillonite, and the adsorption capacity decreased with the increase of solution pH. This is because the point of zero charge (pH_{PZC}) values of montmorillonite, kaolinite, and hematite are 7.2, 3.6, and 5.9, respectively. When the solution pH is below its pH_{PZC} value, the minerals become positively charged due to surface protonation. Therefore, at lower pH, there is a strong electrostatic attraction between positively charged mineral surfaces and negatively charged PFCs, resulting in higher adsorption affinity. With the gradual increase of the pH value of the solution, the surface of the mineral gradually changes from positive to neutral, or even to negative, and the electrostatic attraction disappears. The interaction between adsorbent and adsorbate (including electrostatic repulsion) dominates, resulting in weakened mineral adsorption to PFCs [72].

Chen et al. [67] studied the adsorption of PFOS by materials such as maize straw-origin ash (MA). The pseudo-second-order model fits the sorption kinetics well. According to the sorption isotherms, MA had high sorption capacities (over 700 mg g^{-1}). Due to the positively charged nature of the MA surface, hydrophobic interactions and attractive forces are present, and the development of hemimicelles will enhance adsorption. This shows that MA is very good at removing PFOS from water. MA is the best way to remove PFOS because it is less expensive and it is generally used.

3.1.5. Chitosan

Chitosan is one of the most widely used natural biopolymers and is often produced by alkaline deacetylation of chitin. It is a unique alkaline polysaccharide in nature. Furthermore, chitosan has excellent properties, such as biocompatibility, non-toxicity, and high hydrophilicity [73]. Chitosan has a high content of amino, acetamido, primary, and secondary hydroxyl groups, and these groups are highly active and easily become good chelating sites for target pollutants. These groups also make chitosan extremely easy to chemically modify, so chitosan has been widely studied as a potential adsorbent for the removal of pollutants in water [74]. To overcome the rigidity of chitosan due to its slow adsorption kinetics and solubility under acidic conditions, Elanchezhiyan et al. [75] incorporated materials into the chitosan polymer matrix by cross-linking or metal doping. The insolubility at low acidic pH and the adsorption capacity of chitosan will be enhanced. They combined the composite with chitosan to synthesize a magnetic bead-type material (rGO-ZF@CB), which was immobilized on chitosan beads by reduced graphene oxide (rGO)-modified zinc ferrite. This material can be easily separated and effectively used to remove PFOA and PFOS molecules with maximum adsorption capacities of 16.07 and 21.64 mg/g, respectively. In addition, it is believed that both electrostatic attraction and hydrophobic interaction drive the adsorbent to remove PFOA and PFOS from water. He et al. [76] developed a novel fluoro-functionalized covalent organic framework (F-COF). A layer of chitosan was modified on the surface of F-COF by cross-linking polymerization, and the obtained chitosan/F-COF had desirable hydrophilicity and high specific surface area and surface wettability. Chitosan/F-COF showed high adsorption capacity for PFCs such as PFOS, PFOA, and GenX, and it is believed that electrostatic interaction, hydrogen-bonding interaction, fluorine–fluorine interaction and channel size selection played a major role in the adsorption mechanism. The maximum amount of PFOS and PFOA that could be absorbed (q_{max}) was 8307.1 mg/g and 6177.1 mg/g, respectively. This shows that chitosan and its composites could be used to remove PFCs.

Table 3. Comparison of the adsorption performances of different adsorbents on PFCs.

Adsorbents		Adsorbates	pH	C_0 (mg/L)	Equilibrium Time (h)	Kinetics		Refs
						q_m (mg/g)	k_2 (g/mg/h)	
Activated carbons (AC)	Powder AC	PFOA	3.01~7.20	50	2~3	183.12~344.44	1.18×10^{-2} ~3.19×10^{-2}	[62]
		PFOS	3.00~7.18	50	3~4	322.80~371.22	9.09×10^{-3} ~1.01×10^{-2}	[62]
	Granular AC	PFOA	3.10~7.28	50	90~168	130.80~165.68	1.61×10^{-4} ~8.49×10^{-4}	[62]
		PFOS	3.08~7.20	50	~168	220.58~274.38	1.12×10^{-4} ~1.30×10^{-4}	[62]
	Ordered mesoporous carbon	PFOA	6	10	1	82.5	1.51	[77]
	Reed straw-derived biochar	PFOA	-	0.1	20	17.91	9.61×10^{-4}	[78]
		PFOS	-		20	34.62	8.38×10^{-4}	[78]
	Vitis vinifera leaf litter AC	PFOA	4	0.125	1	119.12	0.74	[79]
		PFOS	4	0.125	1.5	66.01	2.08	[79]
Resins	Anion-exchange resins	PFOA	3.02~7.09	50	168	789.16~1478.04	2.29×10^{-5} ~4.59×10^{-5}	[62]
		PFOS	3.10~7.10	50	48	182.92~193.68	2.23×10^{-4}~2.97×10^{-4}	[62]
	Anion-exchange resins	PFOS	3	200	48~168	210~2575	1.3×10^{-5}~3.2×10^{-4}	[80]
Carbonaceous nanomaterials	Graphene oxide	PFOA	3	10	48	10.17	11.81×10^{-2}	[81]
			5	1000	48	424.1	5×10^{-3}	[81]
	Carbon nanotubes	PFOA	7	0.218	5	0.8284	-	[68]
		PFOS	7	0.269	5	1.0491	-	[68]
	Carbon nanotubes	PFOS	7	100	2	237	1.83×10^{-2}	[67]
Mineral materials	Hydrotalcite	PFOA	-	1000	1	1033	31.77	[82]
		PFOS	-	1000	1	998	6.39	[82]
	Zeolites	PFOA	6.8	300	3	37	7.92	[83]
		PFOS	6.8	300	3	126	3.69	[83]
	Montmorillonite	PFOS	3~6.3	50~500	24	83~99	5×10^{-3}	[84]
	Metal-organic frameworks	PFOS	7	5~100	3.5	154.56	2.51×10^{-3}	[85]

Table 3. *Cont.*

Adsorbents		Adsorbates	pH	C_0 (mg/L)	Equilibrium Time (h)	Kinetics		Refs
						q_m (mg/g)	k_2 (g/mg/h)	
Chitosan	Magnetic bead-type chitosan material	PFOA	3	20	120	16.150	8.22×10^{-1}	[75]
		PFOS	3	20	120	19.545	4.74×10^{-41}	[75]
	Chitosan/covalent organic framework	PFOA	-	0.2	4	99.2	1.38×10^{-1}	[76]
		PFOS	-	0.2	4	86.8	7.8×10^{-2}	[76]
		GenX	-	0.2	4	82.3	3.84×10^{-2}	[76]
	Chitosan-based molecularly imprinted polymer	PFOS	5	0.05	32	275.27	6.00×10^{-4}	[86]
	Non-imprinted polymer	PFOS	5	0.05	32	122.98	7.00×10^{-3}	[86]

3.2. Flocculation

The flocculation process removes PFCs pollution in the water environment, mainly through the adsorption of the flocculant itself and the flocs produced by the flocculant and the suspended solids. Deng et al. [87] used polyaluminum chloride (PAC) as a flocculant to treat PFOA in surface water. The effects of solution pH, PAC dosage, turbidity, and temperature on removal efficiency were investigated using the change in PFOA and suspended solids removal as indicators. The results showed that the removal rate of PFOA increased with the increase of PAC dosage. When the dosage of PAC reaches 10mg/L, the removal rate exceeds 90%. The removal principle is that the PAC cation as a flocculant attracts the anion of PFOA in water through electrostatic action. In addition, the hydrophobic properties of PFOA are extremely beneficial to the adsorption of PFOA by suspended solids. Therefore, by removing the suspended solids, most of the PFOA can also be removed. The adsorption capacity of PFOA by suspended solids increases due to low pH and low temperature energy. At the same time, PFOA (mass concentration of 0.5–3.0 mg/L) can be reduced to below 1 μg/L when 1–16 mg/L powdered activated carbon is added to the wastewater containing PFOA before flocculation. It shows that adding powdered activated carbon before flocculation may help remove PFOA from the water much more quickly. Yu et al. [88] observed that residual PFOS concentrations remained high in water after flocculation treatment. Therefore, the combined process of nanofiltration on the basis of flocculation can significantly improve the removal rate of PFOS in water.

Xiao et al. [89] used polymerization ferric chloride (PFC) and polyaluminum sulfate (PAS) as flocculants to treat wastewater containing PFOA by traditional and enhanced flocculation methods, respectively. Under the conditions of a flocculant dosage of 60 mg/L and a pH value of 4.5 to 6.5, the removal rate of PFOA in wastewater was relatively high. The principle is to form fine aluminum hydroxide flocs in the initial stage of flocculation, and these fine aluminum hydroxide flocs can adsorb PFOA in water. However, with the prolongation of flocculation time, the removal rate of PFOA did not change significantly. Furthermore, the removal rate of PFOA has no relationship with the removal rate of turbidity particles in the wastewater.

Bao et al. [27] found that coagulation resulted in higher removal ratios of PFCs in acidic conditions, suggesting the favorable sorption process of PFOS and PFOA on flocs. The distinctly different degrees of adsorption capacity for PFOS and PFOA were presumably due to their different functional groups and molecular sizes. Under the conditions of 10 mg/L kaolinite clay for turbidity, 1 mmol/L $NaHCO_3$ for alkalinity, and 1 mmol/L NaCl as ionic strength, the removal ratio of PFOS (~32%) was higher than that of PFOA (~12%) at a $FeCl_3 \cdot 6H_2O$ dose of 50 mg/L. The removal of PFOS is better than that of PFOA, even though they have the same $-CF_2-$ chain length, due to the difference in their functional groups. Since the flocs made by the flocculant are oxides, it is easier for strong bases such as sulfonate groups to stick to the oxides than to carboxylate groups.

Electroflocculation (EC) is a green and efficient electrochemical technology that requires metal ions generated by the continuous dissolution of anode electrodes to act as flocculants. The iron anode is oxidized to metal ions, and under the action of oxygen, the Fe ions will further form insoluble ferric hydroxide so that PFOS is adsorbed on its surface.

Meanwhile, Li et al. [90] further activated peroxymonosulfate (PMS) with Fe ions, and the generated OH would degrade PFOS.

The disadvantage of flocculation in water treatment is that the removal rate of PFCs is relatively low. At the same time, the flocculation efficiency is greatly affected by factors such as pH value, flocculant dosage, and temperature in the water body. Similar to the adsorption method, the PFCs are not fundamentally degraded, while there may be problems such as secondary pollution in the experiment. The follow-up treatment of flocculated sludge is also a problem worthy of further study.

4. Advanced Oxidative Degradation Process

Physical techniques are very effective in separating and removing PFCs from water, and the operation is relatively simple. However, whether it is adsorption or flocculation, it can only transfer pollutants from one phase to another and cannot effectively destroy the molecular structure to achieve harmlessness. Furthermore, chemical technology, especially advanced oxidation technology, can effectively degrade most organic pollutants through highly oxidative free radicals, reducing their toxicity and even completely mineralizing them. Compared with long-chain PFCs, short-chain PFCAs tend to exhibit lower biotoxicity and bioaccumulation. Pi et al. [91] investigated the bioaccumulation behavior of PFCs in aquatic macrophytes, including one submerged species, *Echinodorus horemanii* (*E. horemanii*), and one free-floating species, *Eichhornia crassipes* (*E. crassipes*). To represent the plant availability of PFCs in plant uptake characteristics, bioconcentration factors (BCFs) could be employed. Table 4 shows apparent steady-state bioconcentration factors (BCFss, L/kg) of PFCs at the end of the uptake phase in whole-plant of *E. horemanii* (submerged macrophyte) and *E. crassipes* (free-floating macrophyte). The study showed that BCFs increase as perfluoroalkyl chain length increases and that PFCs are easily accumulated in aquatic macrophytes. This means that short-chain PFCs are used less by plants, which is good for environmental security.

Table 4. BCFss (L/kg) of PFCs in leaf, roots, and whole-plant of *E. horemanii* and *E. crassipes*. Reprinted with permission from Ref. [91]. 2022, Pi, N.; Ng, J.Z.; Kelly, B.C.

PFCs	Compounds	Perfluoroalkyl Chain Length	*E. horemanii*	*E. crassipes*
PFTeDA	$C_{13}F_{27}COOH$	13	29.5 ± 2.0	18.8 ± 1.3
PFTrDA	$C_{12}F_{25}COOH$	12	13.7 ± 1.1	26.0 ± 2.2
PFDoDA	$C_{11}F_{23}COOH$	11	19.2 ± 1.3	24.9 ± 1.8
PFUnDA	$C_{10}F_{21}COOH$	10	42.0 ± 2.9	27.9 ± 2.0
PFDA	$C_9F_{19}COOH$	9	24.5 ± 1.8	25.2 ± 1.5
PFNA	$C_8F_{17}COOH$	8	43.0 ± 2.7	27.5 ± 2.9
PFOS	$C_8F_{17}SO_3H$	8	86.8 ± 6.5	89.8 ± 6.9
PFOA	$C_7F_{15}COOH$	7	43.0 ± 2.7	27.5 ± 2.9
PFHpA	$C_6F_{13}COOH$	6	24.5 ± 1.8	25.2 ± 1.5
PFHxS	$C_6F_{13}SO_3H$	6	44.7 ± 3.8	27.9 ± 2.0
PFHxA	$C_5F_{11}COOH$	5	29.4 ± 1.9	24.9 ± 1.8
PFPeA	C_4F_9COOH	4	13.7 ± 1.1	26.0 ± 2.2
PFBS	C_3F_7COOH	3	29.5 ± 2.0	18.8 ± 1.3
PFPrA	C_2F_5COOH	2	-	-
TFA	CF_3COOH	1	-	-

However, due to the abnormally stable physical and chemical properties of PFCs, traditional technologies such as hydrogen peroxide, ozone, and Fenton have poor degradation effects on PFCs [92]. Table 5 summarizes common advanced oxidation techniques for PFOA removal and their second-order rate constants for radical degradation. Recent research has shown that new oxidation methods such as electrochemical approaches and photocatalytic processes are more effective at breaking down PFCs.

Table 5. The second-order rate constant for the reaction of PFOA with the main radical.

Oxidant Species	Formula	Oxidation Potential (V)	Rate constant ($M^{-1}s^{-1}$)	Refs
Hydroxyl free radical	$\cdot OH$	2.8	$\leq 3 \times 10^7$	[93]
Sulfate radical	$\cdot SO_4{}^{2-}$	2.5~3.1	$2.59 (\pm 0.16) \times 10^5$ $\leq 5.0 \times 10^4$	[94] [95]
Ozone	O_3	2.07	1.1×10^4	[96]
Chlorine free radicals	$\cdot Cl$	2.41	/	[89]
Hypochlorous acid	HClO	1.49	/	[89]

4.1. Fenton Oxidation

Because of the strong stability of the C-F bonds of PFCs, it is generally believed that hydroxyl free radicals ($\cdot OH$) alone cannot effectively decompose PFCs. However, $\cdot OH$ still plays a significant role in Fenton's oxidation of PFCs. Mitchell et al. [97] researched the removal of PFOA by the Fenton method. The Fenton system, composed of 1 mol H_2O_2 and 0.5 mmol Fe^{3+}, degraded 89% of PFOA within 150 min. Only 24% of the PFOA was eliminated within 150 min after the OH scavenger 2-propanol was added. This shows that $\cdot OH$ was needed for the PFOA to break down in the Fenton system.

Light radiation (such as ultraviolet light and visible light) combined with Fenton can improve the oxidation activity of the system, which can reduce the amount of Fenton reagent and the pollution of residual Fe^{2+}. Tang et al. [98] researched the degradation of PFOA by the UV-Fenton system and found that the removal rate of PFOA in the UV-Fenton system was over 95%, with a defluorination rate of 53.2%. The reaction is divided into two stages: in the first stage, a large amount of $\cdot OH$ is produced and the removal and defluorination rate of PFOA is faster in this stage; in the second stage, H_2O_2 is depleted and only a small amount of $\cdot OH$ is produced. At this stage, the degradation and defluorination of PFOA are controlled by electron transfer between Fe^{3+} and PFOA (and degradation intermediates), and the reaction rate is relatively slow.

The high-strength C-F chemical bonds in PFCs are extremely difficult to degrade, so they cannot be oxidized by AOPs under normal temperature and pressure conditions. Santos et al. [99] found that PFOA (0.1 mM) had been completely defluorinated after 18 h by thermal activation (T = 343.15 K) catalyzed by persulfate (100 mM). The general use of heat treatment is not desirable from an economic and low carbon standpoint. Therefore, Santos proposed adding humic acid HA (600 mg L^{-1}) and Fenton's reagent (165 mM in H_2O_2, 3 mM in Fe^{3+}) to the PFOA solution. Humic acid is oxidized by Fenton's reagent, while PFOA is quantitatively and irreversibly captured, which makes PFOA unavailable for the aqueous phase. At neutral pH, the remaining Fe(III) acts as a coagulant to solid-phase separate the precipitated oxide HA (containing PFOA). This degradation mechanism is the step-by-step breakdown of PFOA into various intermediates including perfluoroheptanoic acid (PFHpA), perfluorohexanoic acid (PFHxA), perfluoropentanoic acid (PFPA), and perfluorobutanoic acid (PFBA).

Tang et al. [98] studied the process of photo-Fenton UV-Fenton degradation of PFOA. It is divided into two stages: degradation and defluorination. In the first stage of degradation (<1 h), PFOA is rapidly degraded, which is mainly caused by the $\cdot OH$ generated from the decomposition of H_2O_2 by the simultaneous action of Fe^{2+} and UV irradiation. At this stage, the PFOA removal rate is about 90%, while the defluorination rate is only about 35.8%. In the second stage of defluorination (>1 h), when H_2O_2 is almost completely decomposed, Fe^{3+} (mainly generated by the first stage) and UV continue to generate $\cdot OH$ to degrade the residual PFOA. The final removal rate is close to 100%, with the defluorination rate further improved to 53.2%. The degradation of PFOA in the first stage proceeds by Equations (1)–(5).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \cdot OH + OH^- \quad (1)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2\cdot + H^+ \quad (2)$$

$$C_7F_{15}COO^- + \cdot OH \rightarrow C_7F_{15}COO\cdot + OH^- \tag{3}$$

$$C_7F_{15}COO\cdot \rightarrow CO_2 + C_7F_{15} \tag{4}$$

$$C_7F_{15}\cdot + 2H_2O \rightarrow C_6F_{13}COOH + 2F^- + 2H^+ + H \tag{5}$$

The intermediate $C_6F_{13}COOH$ is gradually degraded to short-chain perfluorocarboxylic acids through reactions similar to Equations (6) and (7). In the second stage, when H_2O_2 is completely consumed, the degradation of PFOA may involve the following reactions.

$$C_7F_{15}COO^- + Fe^{3+} \rightarrow [C_7F_{15}COO - Fe]^{2+} \tag{6}$$

$$[C_7F_{15}COO - Fe]^{2+} + hv \rightarrow C_7F_{15}COO\cdot + \ Fe^{2+} \tag{7}$$

4.2. Ozone Oxidation

Ozone oxidation is an efficient, low-residue water treatment method that has been widely used for decades. Several articles and studies [96] have reported on the application and effects of ozone. Ozone oxidation of organic pollutants in water includes two categories:

1. Organic pollutants react directly with O_3 molecules;
2. Organic pollutants are indirectly decomposed by ozone, resulting in the generation of hydroxyl radicals.

The process involves major reactions, as shown in Equation (8):

$$O_3 + H_2O \rightarrow 2\cdot OH + O_2 \left(k = 1.1 \times 10^4\ M^{-1}s^{-1}\right) \tag{8}$$

Second-order rate constants for ozone oxidation are typically between <0.1 $M^{-1}\ s^{-1}$ to $7 \times 10^9\ M^{-1}\ s^{-1}$ [100]. Ozone often reacts as an electrophile or nucleophile due to its electron configuration and structure. In addition, due to its high oxidative activity, ozone is very unstable in aqueous environments, which are highly dependent on the pH of the water. However, at the same time, ozone is also one of the strongest oxidants, with its reaction equation as follows:

$$O_3 + 2H^+ + 2e^- \rightarrow H_2O + O_2\ (E_0 = 2.07\ V) \tag{9}$$

Therefore, ozonation is one of the most common and efficient AOPs. It is widely used in drinking water disinfection, oxidative degradation of organic micro-pollutants, and the removal of inorganic substances. The main catalytic mechanism of ozone is based on promoting the decomposition of organic matter through the formation of OH, or the substance is broken down and forms a complex with the catalyst to speed up the process.

However, there are data showing that ozonation also affects the removal of PFCs in the water treatment process, with related research undertaken. For example, several PFCs used in Australian water reclamation plants [101] showed that the concentrations of PFOS and PFOA in the influent (WWTP effluent) were as high as 3.7 and 16 ng/L, respectively; in the final effluent after ozonation, their concentrations were reduced to 0.7 and 12 ng/L, respectively. Additionally, all of these water quality values were substantially lower than those that are recommended for PFOS and PFOA according to interim drinking water guidelines for health. During the removal of PFOS and PFOA from surface water in drinking water treatment plants, only 9% of PFOA and 2% of PFOS were reported in the ozonation step [102].

Lin et al. [103] reported that PFOA could finally be effectively degraded by adjusting the amount of ozone and the pH value of the reaction process, with the degradation rate reaching 99% within 4 h of reaction time. However, the ozone degradation experiments in the laboratory were performed under relatively high ozone doses (8.7 g h^{-1}) and strongly alkaline conditions (pH 11). This will produce a lot of hydroxyl radicals but will also require a lot of energy, cost a lot of money, pollute the environment in the process, and also generate secondary pollution.

Huang et al. [104] proposed the efficient degradation of PFOA through a combined process of photocatalysis and ozone. PFOA was hardly degraded by ozone alone, with only 0.5% of F defluorinated by limited hydroxyl radicals (generated by autolysis at low levels of ozone) within a 4 h reaction time. However, when ozone was added to the photocatalytic reaction, the degradation efficiency was significantly increased to 99.1% after 4 h of reaction, and almost 44.3% of the C-F bonds were cleaved. In addition, the research proposed photocatalytic ozonation as a method of PFOA degradation. It was discovered that this method resembled the Photo-Kolbe reaction. The PFOA anion is degraded by direct electron transfer from its carboxyl group to photogenerated holes (h^+) to generate the PFOA radical. The PFOA radical is then decarboxylated to form a perfluoroheptyl radical, which is then oxidized to form $C_7F_{15}OH$ by a hydroxyl radical (radical dotOH) generated by photogenerated electrons captured by ozone. Due to its thermal instability, $C_7F_{15}OH$ will be converted to $C_6F_{13}COF$, which is further hydrolyzed to $C_6F_{13}COO^-$. The addition of ozone has been reported to improve the quantum efficiency of photocatalysis [105], resulting in a large number of photogenerated holes participating in the reaction with PFOA instead of recombining with photogenerated electrons. Therefore, photocatalytic ozonation is an efficient process to degrade PFOA.

4.3. Photochemical Oxidation

Sunlight or ultraviolet light (higher energy in the spectrum) can directly act on pollutants to decompose them, which is called photolysis. This method is efficient and environmentally friendly. The degradation of stable contaminants such as PFOA and PFOS by direct photolysis is often slow and inefficient. Therefore, some chemical reagents, catalysts, or combinations with other techniques can be added to enhance the degradation, such as the synergistic effect of light radiation and Fenton in Section 4.1. The degradation efficiency of PFOA for direct photolysis is generally poor. It is reported that the decomposition efficiency reaches 17.5% and the defluorination rate reaches 8.3% within 4 h under 254 nm UV light irradiation [104]. The photon energy value for 254 nm UV irradiation is 471.1 kJ mol^{-1} [106], while the single F bond energy value is 530 kJ mol^{-1}. Therefore, single F bonds are difficult to cleave by 254 nm UV irradiation [107]. Thus, direct photolysis under 254 nm UV light irradiation cannot effectively degrade PFOA.

Hori et al. [108] found that the photolysis of persulfate radical ($S_2O_8{}^{2-}$) would produce strong oxidizing substances such as sulfate radical ($\cdot SO_4{}^-$). The decomposition rate of PFOA was found to be 11 times higher than under direct illumination. The degradation rate of PFOA (1.35 mmol/L) was almost 100% after 4 h of irradiation with a 200 W xenon mercury lamp. Under 254 nm UV light, the combined action of carbonate and $\cdot OH$ can promote the degradation of PFOA, with the degradation rate reaching 95.7% after 8 h and complete degradation after 12 h. This is mainly due to the generation of a strong oxidizing substance, $\cdot CO_3{}^-$, which greatly improves the degradation rate of PFOA [38], as shown in Equations (10) and (11).

$$\cdot OH + CO_3^{2-} \rightarrow OH^- + \cdot CO_3^- \tag{10}$$

$$\cdot OH + HCO_3^- \rightarrow H_2O + \cdot CO_3^- \tag{11}$$

Photocatalytic oxidation has been developed based on photochemical oxidation. Compared with photochemical oxidation, the photocatalytic method mainly realizes the degradation of pollutants through photocatalysts. Cho et al. [109] researched the degradation of PFOA under three systems of TiO_2, VUV, and VUV/TiO_2 at different pH. It was found that the degradation rate of each system at 6 h was TiO_2/VUV (pH 4) > TiO_2/VUV (pH 10) > VUV (pH 4) > VUV (pH 10) > TiO_2 (pH 4) > TiO_2 (pH 10). The degradation rate and defluorination rate of PFOA in the TiO_2/VUV (pH 4) system reached 98.3% and 52.4%, respectively. Chen et al. [110] researched the degradation of PFOA by the Pb–TiO_2/UV system and found that the degradation rate of this system was 32.5 times that of the TiO_2/UV system. This is mostly because the width of TiO_2's forbidden band has been changed. This changes the electron-hole recombination and makes the photocatalytic effect better.

Figure 2 shows the mechanistic pathways of PFOA decomposition. UV irradiation excites Pb–TiO_2 to form perfluorinated alkyl radicals, ·PFOA. The C-C bond between the -C_7F_{15} and -COOH group is cleaved, and the formed perfluoroalkyl radical might then react with water to undergo HF elimination to form $C_6F_{13}COF$. The produced shorter-chain PFCAs follow the hydrolysis pathway to form PFHpA, PFHeA, PFPeA, PFBA, PFPrA, and TFA, and eventually mineralize to CO_2 and F^-.

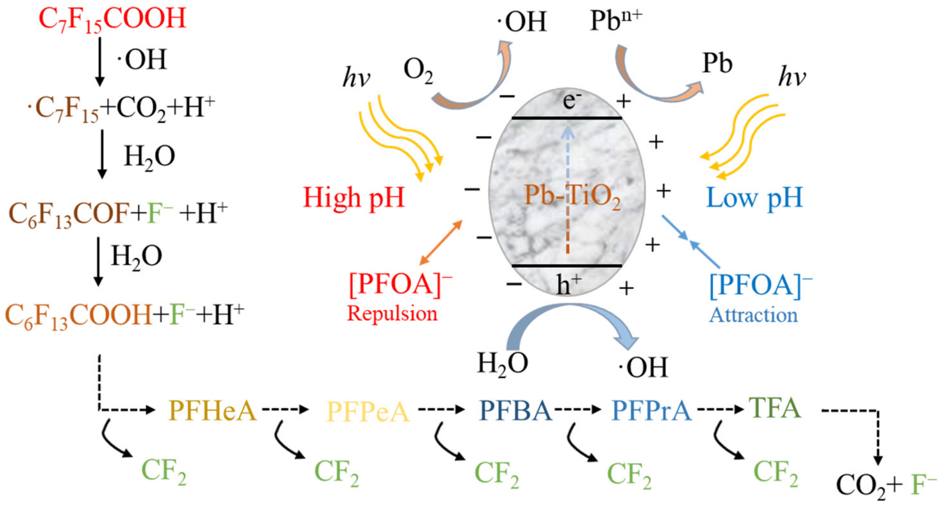


Figure 2. Photocatalytic pathways of PFOA decomposition. Reprinted with permission from Ref. [110]. 2016, Chen, M.-J.; Lo, S.-L.; Lee, Y.-C.; Kuo, J.; Wu, C.-H.

Qian et al. [111] proposed a photochemical degradation mechanism, as shown in Figure 3, which utilizes zeolite-bound iron as the catalytic site for carboxylate−to−metal charge transfer. They used a UV−A irradiated photochemical system to enable photochemical degradation of PFOA adsorbed on Fe-doped zeolite. In a slightly acidic environment ($pH \leq 5.5$), PFOA achieved a relatively complete decomposition (>99%) in a PFOA ($1\ g\ L^{-1}$)-loaded Fe zeolite system within 24 h. PFOA degradation does not occur when there are no iron-containing zeolites or dissolved iron ions in the system. A degradation mechanism with molecular oxygen (O_2) as the final oxidant is proposed. Molecular oxygen participates in the reoxidation of Fe^{2+} to Fe^{3+}, a process that generates reactive species and promotes further mineralization of PFOA. In the presence of Fe-doped zeolite, up to 90% of the initial PFOA was decomposed after 6 h of UV-A irradiation. In addition, complete degradation (>99%) was achieved within 24 h, and the defluorination rate also reached 38%. Compared to the control experiments (non-iron-doped, no zeolite, iron ore), only the combination of UV-A irradiation and Fe-doped zeolite produced significant PFOA degradation, with the Fe species in the zeolite also playing an important role.

4.4. Ultrasonic Oxidation

The principle of ultrasonic treatment is mainly attributed to the cavitation bubbles generated by the liquid. Cavitation bubbles release a lot of energy during the collapse process, creating a local high temperature and high-pressure environment (4000 K and 1800 atm) in the tiny space near the bubble. Therefore, when water enters the cavitation bubbles, the water molecules will decompose to generate hydroxyl radicals and active hydrogen. Moriwaki et al. [112] confirmed that after sonication, PFOA and PFOS were first degraded into fluorine intermediates with fewer carbon atoms. It is then degraded to produce inorganic substances such as fluoride ions and sulfate ions. The degradation of PFOS can be described by Equation (12).

$$C_8F_{17}SO_3^- + \mathrm{Ultrasound} \rightarrow 17F^- + SO_4^{2-} + \sum C_{(8-n)}(n = 0 \sim 7) \tag{12}$$

In the formula, $\sum C_{(8-n)}(n = 0 \sim 7)$ represents the formation of lower molecular weight organic compounds.

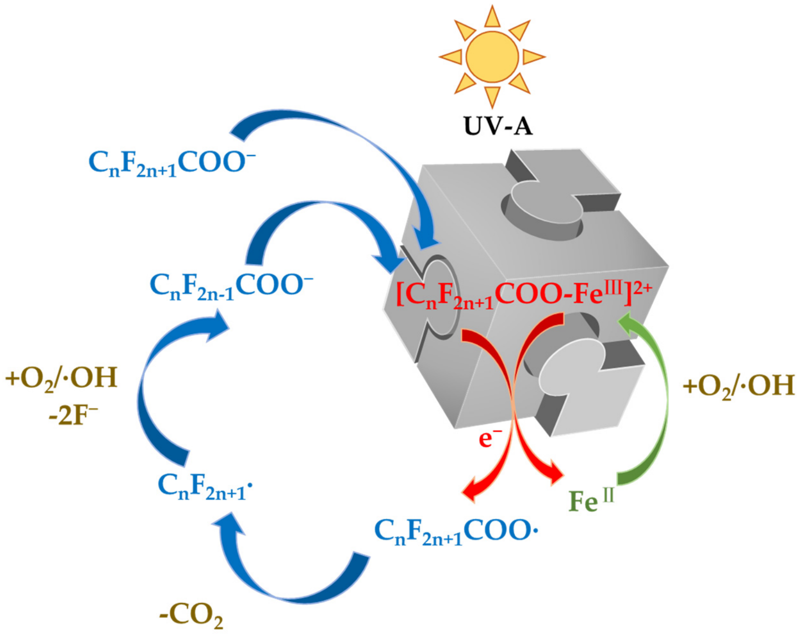


Figure 3. Photochemical mechanism of degradation of PFOA by UV−A irradiation on iron−doped zeolite. Reprinted with permission from Ref. [111]. 2020, Qian, L.; Georgi, A.; Gonzalez-Olmos, R.; Kopinke, F.-D.

Since PFCs are aqueous anionic surfactants, they can be easily adsorbed at the bubble–water interface. The PFCs were then mineralized and degraded by ultrasonic cavitation. Lin et al. [113] studied the enhancement of ultrasonic degradation of PFOA in solution by adding surfactants. Cationic (cetyltrimethylammonium bromide, CTAB) surfactants, anionic surfactants (sodium dodecyl sulfate, SDS), and non-ionic surfactants (octylphenol ethoxylate) were investigated separately (TritonX-100). The sequence was CTAB > TritonX-100 > SDS based on the size of the effect on the rate of degradation. In an ultrasonic system, the reaction proceeds through cavitation bubbles, including the creation and destruction of bubbles. Since PFOA is an anionic surfactant, CTAB is a cationic surfactant. They migrate to the bubble–water interface and reduce the interfacial tension, which in turn enables better PFOA adsorption. Therefore, with the generation of microplasma, PFOA and other surfactants are easily adsorbed on the surface of the bubbles and then destroyed when the bubbles collapse. The removal rate of PFOA was studied with increasing CTAB concentration, reaching 79% after 120 min of reaction at 25 °C and CTAB (additive dose of 0.12 mM).

Rodriguez-Freire et al. [114] investigated the effects of acoustic frequency and initial concentration on the sonochemical degradation of PFOS. It was found that when the acoustic frequency was 500 KHz, the degradation of PFOS (concentration 10–460 µM) conformed to pseudo-first-order kinetics, and the degradation rate increased with the increase of PFOS concentration. When the audio frequency was 1 MHz, the degradation reaction of PFOS (concentration 10–100 µM) also conformed to pseudo-first-order kinetics, and PFOS was completely decomposed in this concentration range. The degradation reaction of PFOS (concentrations greater than 100 µM) followed zero-order kinetics, that is, the rate was independent of the initial PFOS concentration. In addition, when the PFOS concentration was increased to 460 µM, the degradation rate at 500 kHz audio frequency was similar to that at 1 MHz. The reason may be that when the ultrasonic frequency is higher, the time for bubble generation and collapse is shorter, and the probability and intensity of cavitation bubbles are smaller. This directly leads to the deterioration of the degradation effect. Overall, sonication is an effective method for treating aqueous solutions containing low or high concentrations of PFCs. However, the research on ultrasonic processing still remains based on laboratory experiments.

4.5. Electrochemical Oxidation

The electrochemical oxidation method is a promising technology for the degradation of PFCs [115]. In the electrochemical oxidation process of PFCs, the anode material is the main factor affecting the oxidation process. Anode electrode materials with high oxygen evolution potential and good stability are extremely necessary. Otherwise, the current efficiency is low due to a large volume of precipitation of O_2 and the effective electrochemical reaction will not be performed [116]. Typical anode materials that can effectively oxidize PFCs are generally boron-doped diamond (BDD) and some undoped or doped oxides (PbO_2, SnO_2, RuO_2, IrO_2, TiO_2). The degradation rate and defluorination rate of PFOA for some anode electrodes under their optimal conditions are shown in Figure 4 [117].

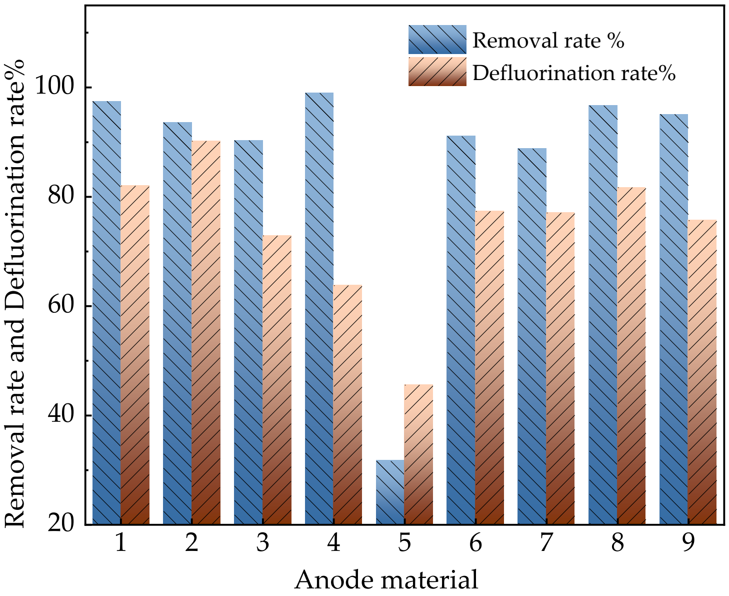


Figure 4. Removal rate of PFOA by different anodes (1-BDD, 2-Si/BDD, 3-Ti/SnO2-Sb, 4-Ti/SnO2-Sb-Bi, 5-Ti/SnO2-Sb/MnO2, 6-Ti/SnO2-Sb/PbO2, 7-Ti/SnO2-Sb/PbO2, 8-Ti/SnO2-Sb/PbO2-Ce, 9-Ti/SnO2-Sb/Yb-PbO2) [116,117].

In addition to the electrode material, other factors can also impact the oxidation effect, such as electrolyte type, current density, and the initial pH of the solution. For example, when the electrolyte is $NaClO_4$, active chlorine species with strong oxidizing properties such as $\cdot Cl$, $\cdot Cl_2^-$, HClO, and ClO_3^- will be generated during the electrolysis process, which promotes the electrochemical degradation reaction [118]. The current density affects the degradation and defluorination rate of PFOA, which mainly affects the electron transfer ability and hydroxyl radical generation in the electrolysis system. In general, it was found that the removal rate increased as the current density increased. In addition, it is more suitable for the degradation of PFOA under acidic conditions [117]. The oxygen evolution potential is higher under acidic conditions, and it is difficult for the oxygen evolution side reaction to occur, which improves the ·OH yield. Under alkaline conditions, OH^- will be adsorbed on the electrode surface and lose electrons due to the electrostatic force. Furthermore, OH^- competes with the $CF_3(CF_2)_6COO^-$ anion for the active site, inhibiting the decarboxylation reaction [116].

Tang et al. [119] performed electrochemical degradation of PFCs in municipal wastewater treatment plant effluent via silver-coated titanium (Ti/Ag) anodes. Compared with the pristine Ti electrode (−0.9707 V), the electrode obtained a higher corrosion potential (−0.3086 V). The results found that the maximum removal efficiencies of short-chain PFCs (C-F < 7), long-chain PFCs (C-F ≥ 7), and chemical oxygen demand (COD) were approximately 70.8%, 91.5%, and 92.0%, respectively. The optimal conditions obtained in the experiment are: the current density is 20.0 mA cm^{-2}, the pH value is 6, the electrolysis time is within 100 min, and the electrode distance is 1.6 cm. Simultaneous degradation kinetic analysis showed that the pseudo-first-order degradation kinetics of COD and PFCs were consistent, with COD degradation occurring before PFCs degradation. The concentrations of long-chain PFCAs including PFDA and PFOA decreased significantly with increasing electrolysis time. On the other hand, the removal efficiency of short-chain PFCA, especially PFHxA, was poor, and it was found that the removal efficiency did not increase until after 45 min. In addition, the major intermediates of PFC were detected as $C_7F_{15}COO^-$, $C_6F_{13}COO^-$, $C_5F_{11}COO^-$, $C_4F_9COO^-$, and $C_3F_7COO^-$. This means that short-chain PFCA is a byproduct of long-chain PFCA during the electrolysis process. The longer the electrolysis time, the more long-chain PFCA is broken down.

The electrochemical oxidation process of the Ti/Ag anode to decompose PFCA (PFC mineralization process) follows a stepwise CF_2 exfoliation reaction, as shown in Figure 5. The PFCs will be gradually mineralized and removed by repeated CF_2 decompression cycles [119]. The carboxylate or sulfonate ions are oxidized to carboxylate and sulfonate radicals, respectively (Equations (13) and (14)) [120]. The highly labile PFCs radicals undergo Kolbe decarboxylation or desulfurization reactions to form perfluoroalkyl radicals (Equations (15) and (16)) [18,116]. Under the action of ·OH (Equation (17)), it will be further converted to perfluoroalcohol, and the subsequent reaction product is perfluorocarbonyl fluoride (Equation (18)). The final intermediate is hydrolyzed to give smaller perfluorocarbons and HF (Equation (19)) [121–123].

$$\mathrm{C_nF_{2n+1}COO^- + Ti/Ag \rightarrow C_nF_{2n+1}COO\cdot + Ti/Ag + e^-} \tag{13}$$

$$\mathrm{C_nF_{2n+1}SO_3^- + Ti/Ag \rightarrow C_nF_{2n+1}SO_3\cdot + Ti/Ag + e^-} \tag{14}$$

$$\mathrm{C_nF_{2n+1}COO\cdot \rightarrow C_nF_{2n+1}\cdot + CO_2} \tag{15}$$

$$\mathrm{C_nF_{2n+1}SO_3^-\cdot + H_2O \rightarrow C_nF_{2n+1}\cdot + SO_4^{2-} + 2H^+} \tag{16}$$

$$\mathrm{C_nF_{2n+1}\cdot + \cdot OH \rightarrow C_nF_{2n+1}OH} \tag{17}$$

$$\mathrm{C_nF_{2n+1}OH \rightarrow C_{n-1}F_{2n-1}COO + HF} \tag{18}$$

$$\mathrm{C_{n-1}F_{2n-1}COF + H_2O \rightarrow C_{n-1}F_{2n-1}COO^- + HF + H^+} \tag{19}$$

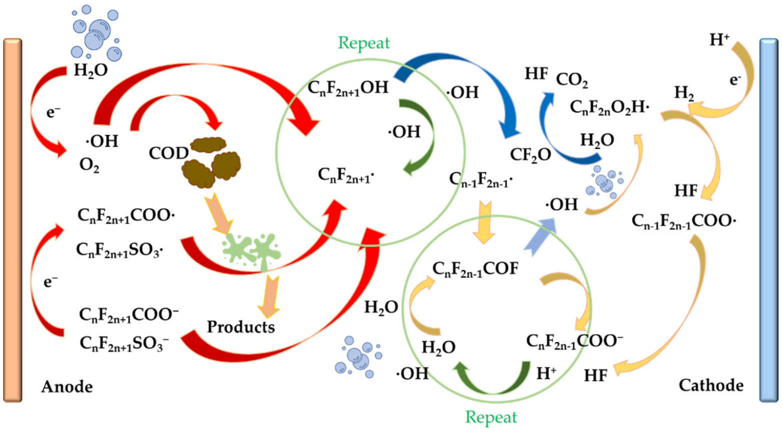


Figure 5. Electrochemical oxidation process of Ti/Ag anodic decomposition of PFCA (stepwise exfoliation of CF_2) Reprinted/adapted with permission from Ref. [119]. 2021, c Tang, J.; Liu, Z.; Lu, W.; Wang, L.; Zhang, C.; Su, P.

5. Conclusions

Perfluorinated compounds (PFCs) have received extensive attention in recent years due to their persistent environmental pollution, bioaccumulation, and biotoxicity. Therefore, this paper firstly summarizes the pollution and distribution of PFCs in the water environment. At the same time, this review focuses on the research status of physical and chemical removal methods for PFCs, including adsorption and flocculation technologies such as activated carbon, ionic resins, carbon nanomaterials, mineral materials, advanced oxidation technologies such as photocatalysis, electrochemical processes, ozone, Dayton, and ultrasound. Several methods for removing PFCs from water have been studied in the laboratory.

PFCs can be degraded by generating ·OH, $SO_4{\cdot}^-$, and other free radicals, with the degradation rate ranging from 24% to 100%. Researchers have discovered technologies that combine electro-oxidation, photocatalysis, or combined with PMS are better based on the rates of degradation and defluorination. Oxidation processes such as Fenton and ozone are generally ineffective when used alone, limited by free radical production conditions. However, the degrading impact may be significantly enhanced by photochemical or electrochemically supported catalysis. From an economic and low-carbon perspective, heat treatment is not frequently used. Most of the water treatment technologies on PFCs work well at the laboratory scale, but large-scale practical field applications have not been evaluated to verify their practical treatment capacity and feasibility, especially for degradable treatment technologies such as AOPs. Although PFCs can gradually remove CF_2 through a series of reactions through decarboxylation, the actual water composition is too complicated and more influencing factors need to be considered. For example, inorganic ions and organics can reduce the removal efficiency of PFC.

Adsorption (activated carbon, ion resin) technology is relatively mature, while PFC removal in the factory application can also be convenient and may fast forward adsorption. However, the treated waste also has the risk of secondary pollution due to the durability of PFOS. Most of the existing research focuses on water and organisms, and there are relatively few studies on other environmental media (soil, atmosphere). Therefore, it is still necessary to explore new technologies to elucidate the reaction mechanism. In a long-term perspective, it is possible to use a combination of technologies to increase the amount of PFCs (through adsorption or concentration) and then use other techniques to eliminate them. PFCs through targeted processes of degradation or reduction is a direction that in-depth research should consider.

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