



Supplementary Materials

A Review on Removal and Destruction of Per- and Polyfluoroalkyl Substances (PFAS) by Novel Membranes

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Table S1. Recently reported adsorption techniques to remove PFAS from wastewater.

| Adsorbate ($\mu\text{g/L}$) | Adsorbent Material | Experimental Conditions | Water Matrix | Efficiency/Adsorption Capacity | Ref. |
|--|--|--|---|-----------------------------------|------|
| PFOA and PFOS: $(2.00 \times 10^4\text{--}2.50 \times 10^5)$ | Activated carbon (10 mg, $2450 \text{ m}^2/\text{g}$) | pH 3–10 175 rpm 48 h 25 °C | DI Water and actual wastewater (100 mL) | 98% | [1] |
| PFOA and PFOS: $1.25 \times 10^2\text{--}1.00 \times 10^3$ | Granular activated carbon (20–50 mg, $295.49 \text{ m}^2/\text{g}$) | pH 4–9 120 rpm 24 h 25 °C pH 3–9 | DI Water | 100% | [2] |
| PFOS: $500\text{--}3.25 \times 10^5$ | Biochar (0.57–1 g/L, 0.5–2 mm) | 120 rpm 24 h 25 °C | DI Water | 178–194 mg/g | [3] |
| PFAS: (20 different types) | Cyclodextrin polymer (10–60 mg/L) | pH ~6.7 40 rpm 48 h 23 °C | Milli-Q water and groundwater (10 mL) | ~100% | [4] |
| PFOA: $5000\text{--}20 \times 10^5$ | Carbon-polysulfide (2–40 g) | pH 2–11 250 rpm 1 h 25 °C | DI water and surface water(5–10000 mL) | 500 $\mu\text{g/g}$ | [5] |
| Flow Rate 200mL/min | | | | | |
| PFOA: $2 \times 10^5\text{--}4.5 \times 10^5$ | Fe ₃ O ₄ @GAC (0.05 g, 2.27 nm, $687 \text{ m}^2/\text{g}$) | pH 2–11 200 rpm 500 h 25 °C | DI water (100 mL) | 91% | [6] |
| PFOA and PFOS: 1×10^5 | All-silica Beta-Zeolite (5 mg) high-silica zeolite | 24 h 25 °C | DI water (5 mL) | PFOA ~ 45 mg/g, PFOS ~ 52 mg/g | [7] |
| PFOS: $(15 \times 10^3\text{--}150 \times 10^3)$ | NaY (0.1 g, 700–780 m^2/g) | pH 7.2 150 rpm 30 °C | DI water (100 mL) | 120 mg/g | [8] |
| PFOA and PFOS: 5×10^5 | UiO-66 and UiO-66-(F4) MOFs (1 mg/mL) | pH 3–10 1 h 150 rpm 25 °C | DI water | 470 mg/g | [9] |

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|-------------------------------|--|--|------------------|-------------|------|
| PFOA | MIL-101(Cr)-MOFs (4 mg) | pH 3–10 12 h 150 rpm 23 °C | DI water (40 mL) | 1.89 mmol/g | [10] |
| PFAS: 200–5 × 10 ⁴ | [NH ₂]-COF (100 mg/L, 1900 ± 1% m ² /g) | pH 3–9 0.5 h 500 rpm 23 °C | DI Water | >90% | [11] |
| PFAS: 8.33–133 | Ferric nanoparticles (40 µg/L) | pH 3–7.5, Ozone bubbling times 15–45 s | DI Water | 44% | [12] |

Table S2. Recently reported studies on ion exchange resins for PFAS removal.

| Adsorbate (µg/L) | Adsorbent Material | Experimental Conditions | Water Matrix | Efficiency/ Adsorption Capacity | Ref. |
|---|---|-------------------------------------|---------------------------------------|------------------------------------|------|
| PFOA and PFOS: 0.1–50 | Purolite A860 (capacity 0.8 meq /mL, a median pore diameter of 7.2 nm and average resin diameter of 750 µm, dosage: 50 mg/L) | pH 3–10 24 h 150 rpm 23 °C | Milli-Q water, and natural wastewater | 50 ng/mg | [13] |
| PFAS: 25 × 10 ³ –4 × 10 ⁵ | Polyacrylic-DVB gel IRA67 (Exchange capacity = 1.6 meq/ mL; Effective size = 0.3–1.2 mm, dosage: 50 mg/L) | pH 3 48 h 150 rpm 25 °C | Actual wastewater | 2397 mg/g | [14] |
| PFOA: 1.2 × 10 ⁵ | Polyacrylic-DVB gel IRA67 (Exchange capacity = 1.6 meq/ mL; Effective size = 0.3–1.2 mm, dosage: 100mg/L) | pH 4 48 h 170 rpm 25 °C | Actual wastewater | 1167 mg/g | [15] |
| PFOA: 5 × 10 ⁴ –4 × 10 ⁵ | Polystyrene-DVB macroporous material IRA910 (Exchange capacity = 1.0 meq/ mL, dosage: 100mg/L) | pH 6 240 h 160 rpm 25 °C | DI water | 1437 mg/g | [16] |
| PFHxS: 5 × 10 ⁴ –4 × 10 ⁵ | Polystyrene-DVB macroporous material IRA910 (Exchange capacity = 1.0 meq/ mL, dosage: 100mg/L) | pH 6 240 h 160 rpm 25 °C | DI water | 1364 mg/g | [16] |
| PFOS: 25 × 10 ³ –4 × 10 ⁵ | Polyacrylic-DVB gel IRA67 (Exchange capacity = 1.6, dosage: 0.05 mg/L) | pH 3 48 h 150 rpm 25 °C | Actual wastewater | 2750 mg/g | [14] |
| PFOS: 4 × 10 ⁵ | Polyacrylic macroporous material | pH 3 48 h | DI water | 2000 mg/g | [17] |

| | | | | | |
|--|---------------------------|----------|------|------|--|
| IRA958 (Exchange capacity = 3.7 meq/g; Effective size = 0.3–1.2 mm, dosage: 35–500 mg/L) | 150 rpm 25 °C | | | | |
| Polystyrene-DVB macroporous material | pH 6 | | | | |
| IRA910 (Exchange capacity = 1.0 meq/mL, dosage: 100 mg/L) | 240 h 160 rpm 25 °C | DI water | 1395 | [16] | |

SI 1. Destruction Techniques:

1.1. Biological Treatment

The biological treatment technologies for PFAS are sustainable and eco-friendly technology, which does not produce any secondary pollutants. Biological degradation of PFAS by bio-catalysts (Eg. enzymes) that directly eliminate fluorine atoms from PFAS either (a) by oxidation (addition of oxygen across the C-F bond) or (b) reduction (addition of electrons across the C-F bond). The strong C-F bond in PFAS requires a significant amount of energy to catalyze the reaction. The reported study shows that the biological treatment of PFAS can be done by microbial remediation (bacteria: *Gordonia*, *Acidimicrobium*, etc.), mycoremediation (fungi: *Aspergillus niger*, *P. chrysosporium*, etc.), and phytoremediation (Plant: *Betula pendula*, *Picea abies*, etc.). There are a few known microbes that can break down the C-F bond in aerobic (with oxygen) or anaerobic (without oxygen) conditions, but the degradation process is either very slow (it takes a few days to 180 days depending on the process) or less efficient compared to adsorption or membrane separation.

In the future, the defluorinating microorganisms in anaerobic digestion that can use generated methane and hydrogen as an electron donor, while PFAS act as an electron acceptor could intensify the breakdown of the C-F bond in the biodegradation of PFAS compounds. The combination of bioremediation techniques also has the potential to enhance the efficacy to degrade PFAS [18–20].

1.2. Advanced Oxidation or Reduction Processes (AOPs and ARPs)

AOPs and ARPs are capable of mineralization of PFAS in an eco-friendly way from the environment, which is one of the biggest advantages of this process. In an AOP or ARP, the efficiency of the process depends on the amount of OH or O₂⁻ radical present in the system. Whereas the generation of OH or O₂⁻ radicals depends on the process implemented and the operating parameters. Several treatment technologies come under AOPs and ARPs, such as ozonation [21], UV irradiation, H₂O₂ treatment, fenton, photo-fenton, photolysis, photocatalysis, and reduction with elemental iron sonolysis, electrocatalysis, and/or a combination of the above-mentioned methods. Even though these above-mentioned methods are capable of destroying PFAS efficiently, most of the processes are not suitable for the long run (reusable) or expensive, or both. Few of the most recently published AOPs and ARPs for the decomposition of PFAS shows that the time required for ~90% decomposition is a few minutes to days, which is subjected to the treatment process and operating conditions (Photocatalysis: light source, catalyst, initial pollutant concentration, etc.; Ozonation: ozone dosage, initial pollutant concentration, etc.; Fenton: catalyst dosage, pH, temperature, initial pollutant concentration, etc.; Zero valent iron: pH, initial pollutant concentration, etc.) [21–23].

Since the AOPs and ARPs are mostly costly technique, it is often used in combination with other separation techniques. In such cases, first, the PFAS contaminated water is

treated by the most popularly known efficient techniques (adsorption or membrane separation process) followed by AOPs or ARPs. This type of operation can efficiently remove and mineralize PFAS from the contaminated water, but this area of research has been poorly studied till now, which needs more attention [24].

1.3. Electrochemical Oxidation Treatment

Electrochemical oxidation is a wastewater treatment technology in which electrical current is passed through the solution to oxidize the contaminants. Electrochemical oxidation treatment of strong organic pollutants (such as PFAS) has been illustrated in several recently reported bench and pilot-scale operations [25–27].

Advantages of this process include simple, operable in the ambient environment, and does not require any chemical oxidants as additives. Though, it has a few drawbacks, which include the potential of generating toxic by-products, partial or incomplete destruction of PFAS in some cases, reduction in efficiency due to minerals build up on the anode, expensive electrodes, and possible volatilization of pollutants. In spite of these limitations, electrochemical oxidation can be a promising technology for the destruction of PFAS in certain cases because of its proven ability to mineralize PFAS with comparatively lower energy demands than thermal incineration [26,28,29].

The future work on the electrochemical oxidation of PFAS should be focused on (a) inexpensive electrodes; (b) coupling the process with another water treatment operation to destroy by-products; (c) consideration of long-term operation; (d) detailed analysis of potential by-products to understand the degradation pathway of PFAS; and (e) scale-up consideration and process parameters optimization [30–32].

1.4. Sonochemical Treatment

The sonochemical treatment process has been widely studied for environmental applications, especially for pollutant removal from wastewater due to its significant superiority, such as simple, cleanliness, energy-saving, complete destruction of pollutants without generating secondary contaminants. Numerous lab-scale reported studies on sonochemical destruction of PFAS showed outstanding potential. The ultrasound (>16 kHz) used during the sonochemical treatment generates significantly high pressure and temperature, which is sufficient to pyrolyze the PFAS present in wastewater. In the course of this process, it also generates highly reactive OH radical, which actively participates in pollutant degradation as an oxidizing agent [33].

The sonochemical treatment can be affected by various operating parameters, namely ultrasound frequencies, power densities, temperature, additives, atmospheric conditions, initial concentration of PFAS, chemical properties of PFAS, etc. To improve the sonochemical technology for the degradation of PFAS, the future research work can be focused on (a) reduction of the energy consumption; (b) combining this technology with other potential methods (photocatalysis, ozonation, Fenton, etc.) to increase the effectiveness and lower the energy usage; and (c) scale-up of the process [34–42].

1.5. Photocatalytic Treatment

Photocatalytic destruction of pollutants from wastewater has gained popularity for the last few decades because of its several advantages, such as eco-friendliness, simple process, complete destruction is possible, efficient, and economical. In a photocatalytic system, there are two main components involved: catalyst (Heterogeneous: TiO₂, graphene oxide, ZnO, etc.) and the light source (UV, visible, solar, LEDs, etc.). The basic working principle of this process is the generation of electron-hole pairs into the system under light irradiation on the catalyst surface. This electron-hole pair helps to produce OH and/or O₂[·] radicals, which react with the pollutants present in the wastewater. The photocatalytic process can effectively mineralize very small amounts of pollutants

from contaminated water, but it depends on several operating parameters (catalyst loading, initial pollutant concentration, light source, catalyst surface area, reactor geometry, pH, temperature, etc.). Photocatalysis is one of the most explored techniques for the degradation of PFAS, but the research work is mainly based on a lab-scale operation. Thus researchers are still working on the implementation of large-scale operations by overcoming the limitations (utilization of sunlight instead of UV light and large scale reactor design) of the photocatalytic process.

A distinctive and important property of the photocatalytic process is the recovery of the catalyst after the operation, which makes the catalyst reusable (without regeneration in most cases), and the process becomes economically viable. For the further improvement of PFAS removal efficiency, the combination of membrane or adsorption technology with photocatalytic processes has been reported in a few recent studies. These combined techniques have a good potential for the successful mineralization of PFAS, but this area is poorly studied which needs immediate attention [43–49].

1.6. Incineration

Incineration or thermal treatment for the destruction of toxic chemicals (PFAS) is a simple, old and effective technique, but it requires an ample amount of heat energy. This process is well known for the regeneration of contaminated adsorbents and ion exchange resins. Heat is directly applied to the PFAS-contaminated liquids (wastewater) or solids (soil, adsorbent) and vaporizes the combustion products. The vaporized products can be captured and/or subjected to further oxidation at elevated temperatures ($\geq 1000^{\circ}\text{C}$). The main parameters affecting the incineration process are temperature, time, gas supply (H_2), etc. Furthermore, there are things to be careful about during the incineration process: (a) degree of destruction, (b) generation of by-products, and (c) transport of contaminants to the incineration point.

Incineration of PFAS waste may not be a cost-effective disposal method, but it ranks as the best among available technologies in the current scenario. Also, the treatment of large volumes of liquid or solid by this method is problematic and prohibitively expensive. That is why the common practice is to combine this technology with membrane technology. At first, the PFAS are separated by a membrane, and then the concentrated PFAS solution is treated by incineration [50–55].

1.7. High Energy Electron Beam

The high-energy electron beam is a nonthermal, highly efficient, chemical-free technology that uses electron accelerators to produce a huge number of highly energetic electron-mass from electricity. These highly energetic electrons participate in ionization reactions with the materials that they interact with and generate a variety of short-lived radical species which are involved in the oxidative and reductive destruction of PFAS. The efficiency of this process depends on several parameters, such as the type and thickness of the material used during the interaction, exposed time of the material to the electron beam, and beam power. This process is a promising mineralization technique to break down the short-chain PFAS, but the process needs further attention to overcome the high operating cost [56–60].

Abbreviations:

- PFAS: Per and Poly-fluoroalkyl Substances.
- PFHxS: Perfluorohexane sulfonate
- PFOS: Perfluorooctanesulfonic acid
- PFHpA: Perfluoroheptanoic acid
- PFOA: Perfluorooctanoic acid
- PFNA: Perfluorononanoic acid
- PFDA: Perfluorodecanoic acid

PFUnDA: Perfluoroundecanoic acid
PFDoDA: Perfluorododecanoic acid
PFTA: Perfluorotetradecanoic acid
PPeA: Perfluoropentanoate
PFHxA: Perfluorohexanoate
PFPrS: Perfluoropropane sulfonate
PFBS: Perfluorobutane sulfonate
PFPeS: Perfluoropentane sulfonate
PFHpS: Perfluoroheptane sulfonate
PFDS: Perfluorodecane sulfonate
GO: Graphene Oxide
PMPA: Perfluoro-2-(perfluoromethoxy) propanoic acid
PEI: Polyethyleneimine
PVDF: Polyvinylidene fluoride
AFFF: Aqueous Film-Forming Foam
UF: Ultrafiltration
MF: Microfiltration
RO: Reverse Osmosis
NF: Nanofiltration
MD: Membrane Distillation
MOF: Metal Organic Frameworks
COF: Covalent Organic Frameworks
AOPs: Advanced Oxidation Processes
ARPs: Advanced Reduction Processes
UV: Ultraviolet

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