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Per- and Polyfluoroalkyl Substance (PFAS) Degradation in Water and Soil Using Cold Atmospheric Plasma (CAP): A Review

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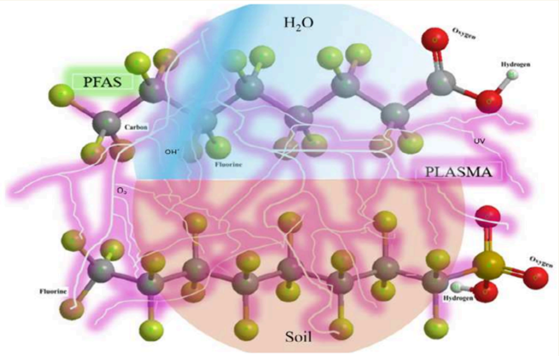
ABSTRACT: Per- and polyfluoroalkyl substances (PFASs) are persistent organic chemicals found in numerous industrial applications and everyday products. The excessive amounts of PFASs in water and soil, together with their link to severe health issues, have prompted substantial public concerns, making their removal from the environment a necessity. Existing degradation techniques are frequently lacking due to their low efficiency, cost-effectiveness, and potential for secondary contamination. Cold Atmospheric Plasma (CAP) technology has emerged as a promising alternative, utilizing energized reactive species to break down PFASs under ambient conditions. Therefore, this review examines the efficacy and effectiveness of CAP in degrading PFASs by reviewing various CAP setups and examining the key factors involved. This review also aims to further the development of CAP as a viable solution for PFAS degradation by addressing outstanding challenges and future directions in soil and water treatment.



KEYWORDS: *Per- and Polyfluoroalkyl Substance, PFAS, Cold Atmospheric Plasma, CAP, Reactive species, Defluorination, Degradation, Water treatment, Soil treatment, Efficacy*

1. INTRODUCTION

Per- and polyfluoroalkyl substances (PFASs), commonly known as forever chemicals, represent a large and diverse group of over 4000 compounds. These substances are characterized by hydrophobic alkyl chains that are either fully fluorinated (perfluorinated) or partially fluorinated (polyfluorinated), contributing to their remarkable stability and persistence in the environment.[1] Among these, perfluorooctanoic acid (PFOA) and perfluorooctanesulfonate (PFOS) have garnered significant attention due to their extensive use across a range of industrial and commercial applications, including fire-fighting foams.[2,3] The disposal and discharge of these PFAS-containing products have contributed to widespread contamination of surface water, soil, and groundwater, affecting both animal and plant systems.[4,5]

Additionally, PFASs can be released into the environment through material shedding during everyday use, washing, and disposal. In landfills, the anaerobic breakdown of PFAS-treated textiles and carpets can release these substances, which may leach into surrounding areas via rainwater if landfill leachate is inadequately contained.[6] Furthermore, human exposure to PFASs primarily occurs through the consumption of contaminated food or the migration of PFASs from food packaging and cookware.[7,8] Exposure to PFASs has been associated with a wide range of acute and chronic health conditions in addition to its impacts on the environment. In humans, studies have linked these substances to conditions such as thyroid disorders, respiratory issues such as asthma, mental health challenges, including anxiety, and metabolic disturbances such as obesity and hyperuricemia. Additionally, PFAS exposure is connected to immune system toxicity, kidney and liver damage, cardiovascular diseases, and pediatric allergies. Beyond human health, PFASs also adversely affect ecosystems, inhibiting plant growth and reducing biomass in agricultural and natural settings.[9−13]

The Environmental Protection Agency (EPA) established new health advisory levels for drinking water in response to these associated health and environmental risks, setting limits at 0.02 parts per trillion (ppt) for PFOS and 0.004 ppt for PFOA.[14−16] However, many water bodies near airports, military installations, and industrial sites continue to exceed these thresholds.[17] In May 2022, the U.S. EPA expanded its list of regional screening levels (RSLs) and remedial management

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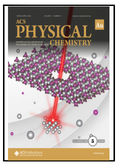

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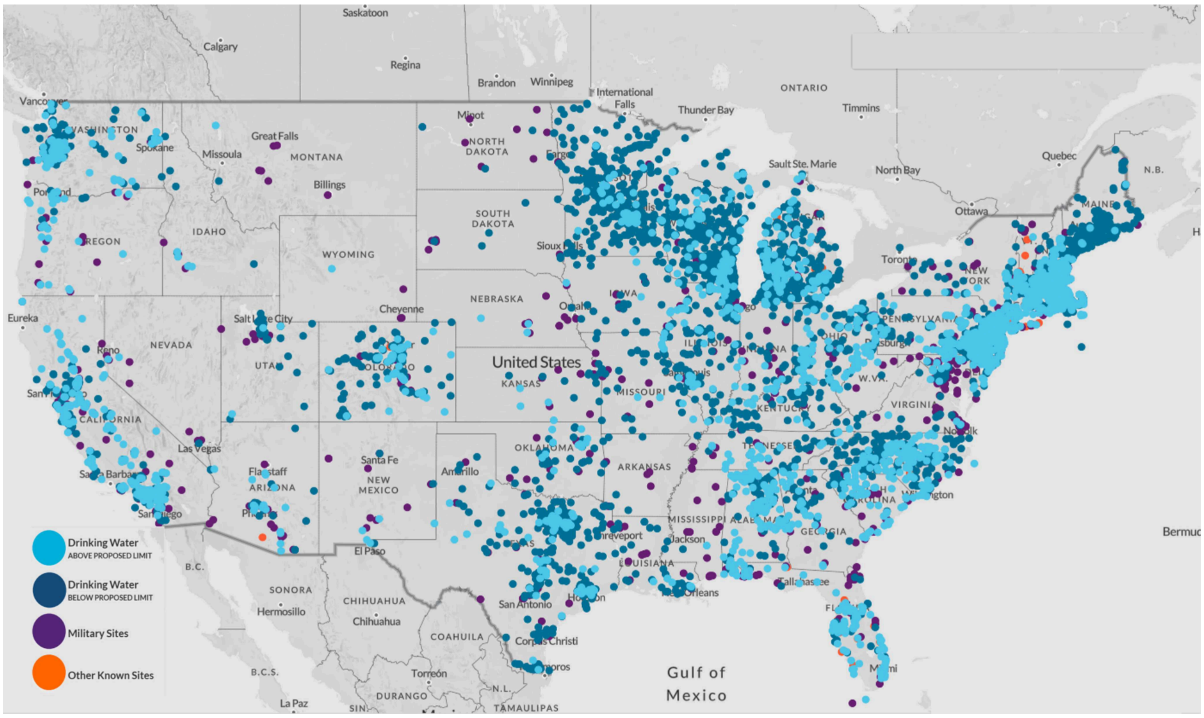

Figure 1. Map showing PFAS contaminated sites in the US from EWG on August 9, 2024. Copyright 2024 Environmental Working Group, www.ewg.org. Reproduced with permission.

levels (RMLs) to include five additional PFAS compounds: PFOA, PFOS, GenX chemicals, perfluorononanoic acid (PFNA), and perfluorohexanesulfonic acid (PFHxS), along with related compounds such as perfluorononanoate, perfluorooctanoate, and perfluorohexanesulfonate.[18−20] Reports from the Environmental Working Group (EWG) on May 21, 2024, showed 6,189 PFAS contaminated sites, but as of August 9, 2024, recent data revealed the presence of PFASs at 7,457 sites across all 50 states, the District of Columbia, and four U.S. territories. in the US indicating an alarming increase in PFAS release into the environment.[21−23] As illustrated in Figure 1, PFAS contamination is widespread, affecting drinking water sources, military sites, and other regions across the U.S.

These findings highlight the urgent need for effective mitigation strategies to address this growing environmental and public health concern.[7,24−29] Various conventional water and soil remediation methods have been applied so far for PFAS degradation in water and soil, including activated carbon adsorption, biodegradation, thermal methods, membrane separation, photocatalysis, and ball milling.[30−42] Researchers have utilized these conventional techniques to explore PFAS occurrence, fate, transport, and treatment through both in situ and ex situ processes.[27−29] However, these traditional methods face several challenges and limitations, including inadequate removal of PFOA, high energy demands, prolonged treatment durations, and the potential for secondary pollution.

In recent years, Cold Atmospheric Plasma (CAP) has gained attention as a promising technology for PFAS degradation due to its ability to generate a variety of reactive species, including ozone, hydroxyl radicals, and atomic oxygen, under near-ambient conditions.[43−46] Despite its potential, most research to date has focused on small-scale, ex situ applications. To better understand the mechanisms driving PFAS degradation using CAP and to identify the factors influencing field deployment and the effectiveness of this process across different PFAS types on a larger scale, further research is needed. The aim of this work is to review different experimental results for the degradation of PFAS in water and soil treatment using different plasma reactors. The optimal operational conditions were identified by reviewing parameters like treatment time, gas type, and plasma reactors.

2. BACKGROUND

2.1. Cold Atmospheric Plasma (CAP)

Plasma, often referred to as the fourth state of matter, is a neutral, ionized gas consisting of free electrons, ions, and photons generated under the influence of a strong electric field. The density of free charges in plasma can vary significantly depending on its properties. Broadly, plasma is categorized into two main types: Thermal Plasma (TP) and Nonthermal Plasma (NTP).[47,48] Thermal Plasma is formed at high gas pressures where frequent collisions between high-energy electrons and neutral particles occur, leading to thermal equilibrium between the gas molecules and electrons ($T_g = T_e$, where T_g and T_e are the temperatures of the gas molecules and electrons, respectively). In this state, electron densities typically range from 10^{16} to 10^{19} cm^{-3} with temperatures for both gas molecules and electrons ranging from 10 to 100 eV. An example of TP is the plasma torch used in Inductively Coupled Plasma − Optical Emission Spectrophotometers (ICP-OES). TP also has a variety of industrial applications, including metal cutting, material processing, waste treatment, and scientific research such as nuclear fusion.[47,49,50]

Nonthermal Plasma (NTP), also known as cold atmospheric plasma, comprises ions, electrons, neutral atoms, and reactive species generated under atmospheric or low-pressure conditions near room temperature.[51,52] In NTP, collisions between free electrons and neutral particles are insufficient to establish thermal equilibrium, resulting in electron temper-

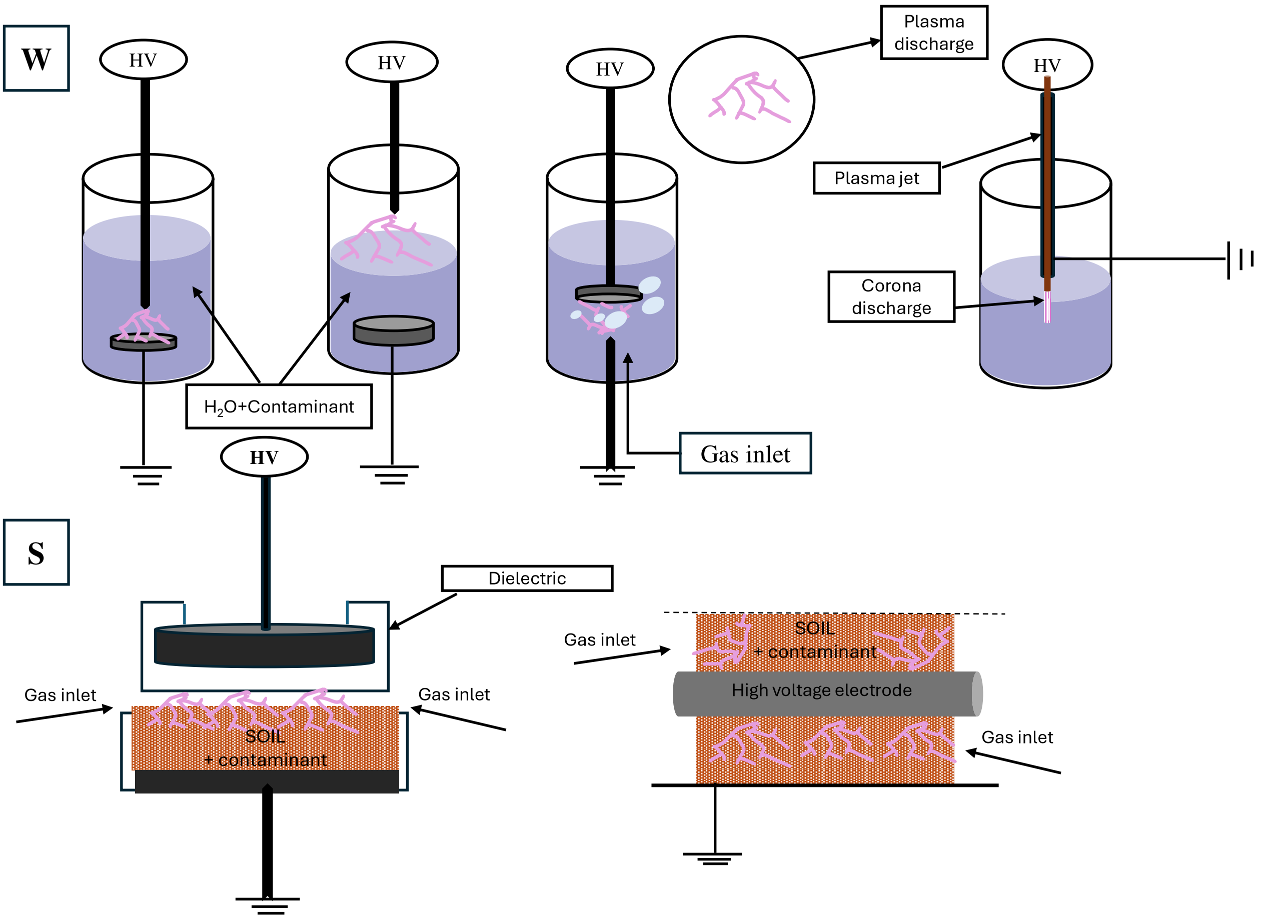

Figure 2. CAP electrode configurations for water (W) and soil (S) treatment. Adapted with permission from ref 53. Copyright 2022 Elsevier.

atures T_e in the range of a few electronvolts, while the gas temperature T_g remains close to room temperature. The electron density in NTP ranges from 10^8 to 10^{14} cm^{-3}.[49,51] Various plasma discharges, such as glow discharge, pulsed corona discharge, radiofrequency discharge, gliding arc discharge, and dielectric barrier discharge (DBD), as well as different operating gases (e.g., air, argon, oxygen, nitrogen), can produce plasma with distinct properties, enabling a wide range of applications.[53−57]

NTP or CAP has several advantages over thermal plasma for pollutant degradation, including lower energy consumption and operational costs, due to its ability to generate reactive species at ambient temperatures rather than requiring the high temperatures of thermal plasma. NTP also minimizes the risk of secondary pollution by avoiding the formation of harmful byproducts that can occur at the high temperatures of thermal plasma. Additionally, the flexibility and scalability of NTP systems make them suitable for in situ applications and easier integration into existing water and soil treatment processes. CAP has also been widely studied for various applications, including surface modification, sterilization, and medical treatments, due to its nonthermal nature and ability to generate a diverse array of reactive species.[58−60] One of the most significant advantages of CAP is its ability to operate at low temperatures, thereby reducing energy dissipation.

CAP has recently demonstrated significant potential for the removal of PFAS from water and soil. Among the commonly used CAP generators for PFAS degradation are pulsed dielectric discharge and corona discharge systems. CAP operates by applying high voltage to ionize nitrogen (N_2) and oxygen (O_2) in the air between the electrodes, producing an array of reactive species, such as free radicals, ultraviolet radiation, ultrasonic waves, electrons, and other active ions.[61−64] These reactive components work to oxidize or reduce contaminants effectively. As an advanced technology for environmental remediation, CAP has shown the ability to degrade pollutants to levels below regulatory limits, achieving degradation rates that surpass those of leading alternative technologies.[65−67] Figure 2 shows the different CAP reactor configurations that have been employed for soil and water treatment, including the use of a dielectric barrier and plasma jet corona discharge.

In soil treatment, the primary reactive species involved in PFOA degradation during plasma treatment were identified by introducing various gases and incorporating electron scavengers during corona discharge experiments. By analyzing the byproducts formed, a degradation pathway for PFOA in the plasma discharge process was proposed.[68] Furthermore, the effects of CAP treatment on soil were evaluated by planting lettuce in various soil matrices, including those treated with CAP. The results indicated an accelerated nitrogen cycle process in the soil, along with enhanced soil fertility. This was confirmed through an analysis of bacterial community distributions at the phylum level, conducted before and after the treatment.[68,69]

3. PER- AND POLYFLUOROALKYL SUBSTANCES (PFASS)

PFASs, commonly called *forever chemicals*, have several applications and can contaminate the soil and water supply via various means. Common sources of PFAS found in some everyday products include pesticides, fire-fighting foams, fast food packaging, clothing, nonstick cookware, washing fluids, paints, and stain resistant products as seen in Figure 3 and include other industrial materials not listed.[70−72]

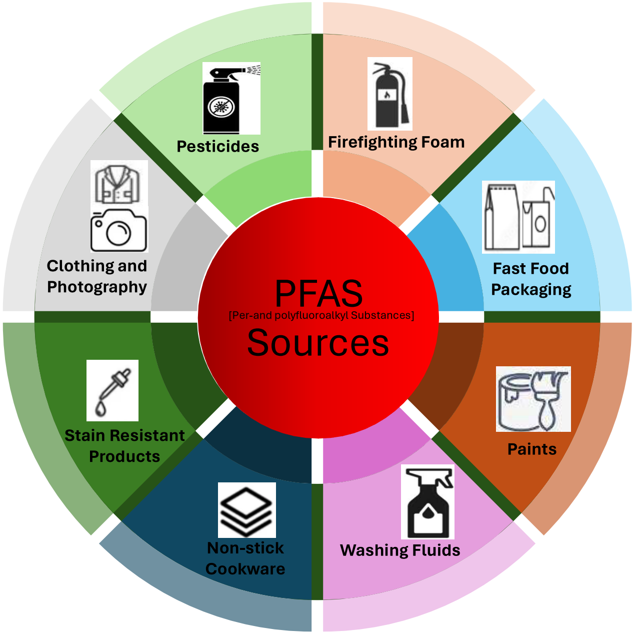

Figure 3. Common sources of PFAS in our environment.

These forever chemicals belong to a class of numerous chemical compounds. However, the commonly encountered PFASs are the perfluorocarboxylic acids (PFCAs) and perfluorosulfonic acids (PFSAs). The PFCAs and PFSAs contain the PFOAs and PFOSs (Figure 4), having vast applications in our everyday lives.[73−77]

PFCAs are characterized by a perfluorinated carbon chain with a carboxylate functional group.[78,79] Examples of PFCAs include PFOA, PFBA, PFDA, and PFHxA (Figure 4). This group of compounds have been used in various industrial processes, consumer products, and fire-fighting foams due to their water and oil repellency, as well as their ability to lower surface tension.[80] PFSAs consist of a perfluorinated carbon chain with a sulfonate functional group. Examples of PFSAs include PFOS, PFHxS, and PFBS (Figure 4). PFSAs have been commonly used in stain-resistant coatings and fire-fighting foams and as surfactants in various consumer products.[81−83]

4. PFAS DEGRADATION USING CAP

Numerous studies have explored the application of CAP to the degradation of PFASs in water and soil matrices. These studies have demonstrated the effectiveness of CAP in PFAS degradation, with high efficiencies and relatively low energy consumption compared to other traditional methods.[30,56,84]

4.1. Water Treatment

The methods employed in CAP-based water treatment can be broadly categorized into three types: direct, indirect, and bubbling methods. In the direct method, electrodes are submerged in the water solution, generating sparks and streamers directly within the bulk of the solution being treated.[85] In contrast, the indirect method positions the electrodes above the water surface, allowing reactive species generated by the plasma to diffuse into the water across the plasma−water interface.[56,86] Finally, in the bubbling method, plasma is generated within bubbles injected into the solution. Reactive species are formed inside these bubbles and are readily dispersed throughout the water.[56,87]

Papalexopoulou et al. compared the degradation efficiency of a plane-to-plane gas−liquid DBD (G-LDBD) and DBD-based underwater plasma microbubbles (PMBs) using a HV nanosecond (ns) and microsecond (μs) pulsed HV supply for the removal of PFOA from water Figure 5).[47] For electrical

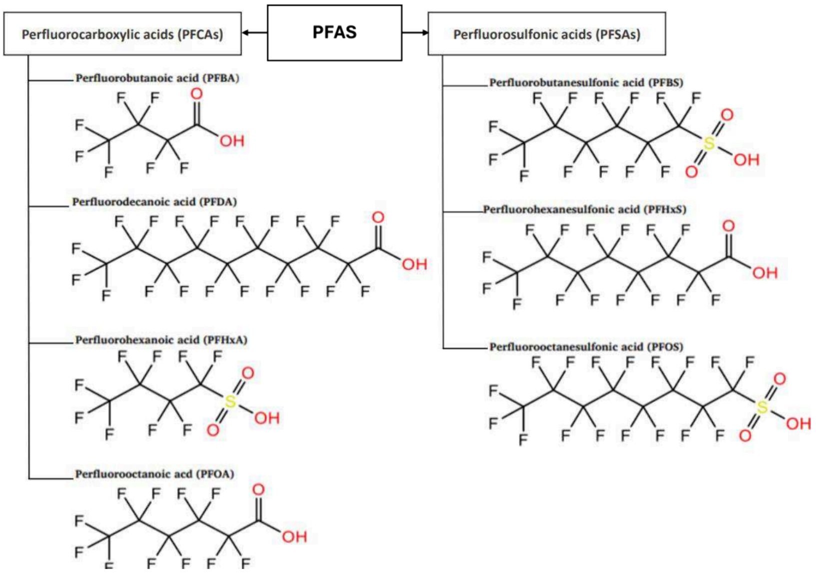

Figure 4. Structure of some PFCA and PFSA compounds. Adapted with permission under a Creative Commons [CC-BY 4.0] license from ref 55. Copyright 2023 Elsevier.

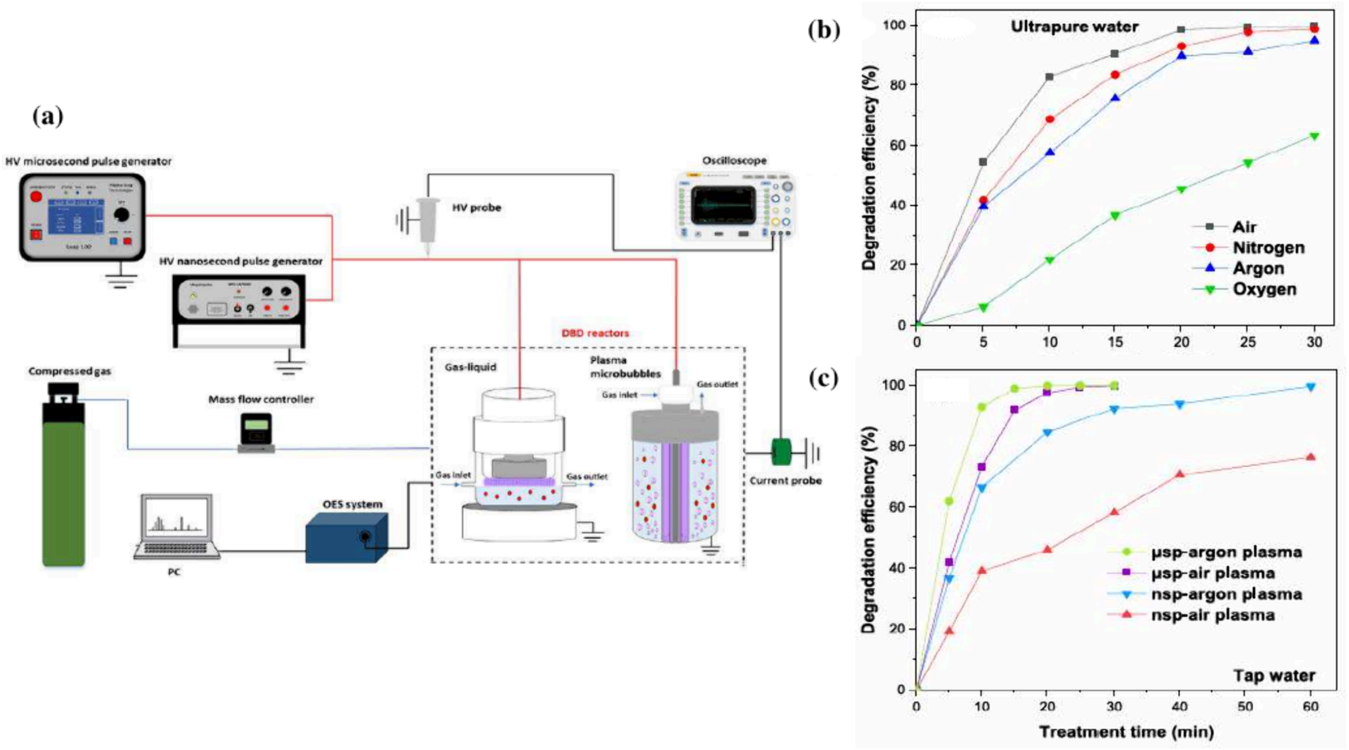

Figure 5. (a) Experimental setup utilized for the treatment of PFOA-contaminated water samples using CAP, (b) PFOA degradation efficiency in different plasma gases in ultrapure water, and (c) comparison of high-voltage microsecond and nanosecond pulses for PFOA degradation in tap water using air- and argon-based plasma. Reproduced with permission from ref 47. Copyright 2024 Elsevier.

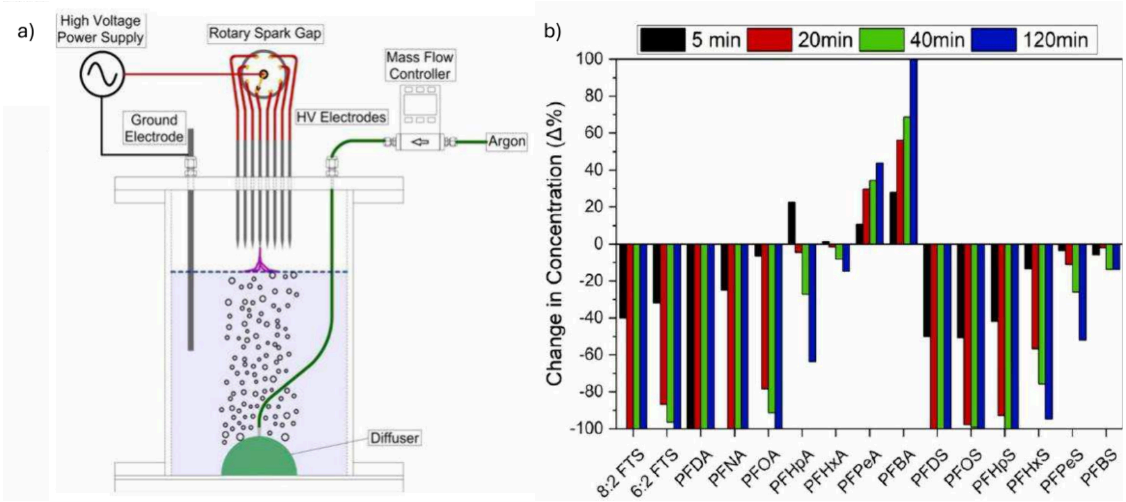

Figure 6. (a) Bubble column cold plasma discharge for water treatment using argon gas and (b) change in PFAS concentration as a function of time during CAP treatment. Reproduced with permission under a Creative Commons [CC-BY 4.0] license from ref 55. Copyright 2023 Elsevier.

characterization, a digital oscilloscope was utilized in conjunction with current and voltage probes. These experiments were carried out under air, Ar, N_2, and O_2 atmospheres. The plasma treated PFOA samples were analyzed by liquid chromatography−mass spectroscopy (LCMS). The experiment was conducted to compare the degradation efficiency of air and argon generated plasma. PFOA degradation was more effective with air−LDBD compared to DBD-based underwater air-PMB. This aligns with the surfactant properties of PFOA and the stronger plasma−liquid interactions and reactive species generation observed during gas−liquid treatment compared to underwater PMB. The results showed higher degradation efficiency (>99.9%) for the removal of PFOA from ultrapure water using the microsecond pulsed air−LDBD after 30 min of treatment (Figure 5). However, a higher degradation efficiency (>99.9%) of PFOA was observed after using the microsecond argon−LDBD for tap water treatment after 20 min (Figure 5).

Alam et al. used a bubble column with cold plasma discharges for PFAS degradation, using argon as the working gas because of its low breakdown voltage of 2.7 kV/cm compared with air at 32 kV/cm.[55] The setup as seen in Figure 6 used 8 tungsten carbide tipped electrodes inside the plasma generator and an argon gas inlet for PFAS degradation. The

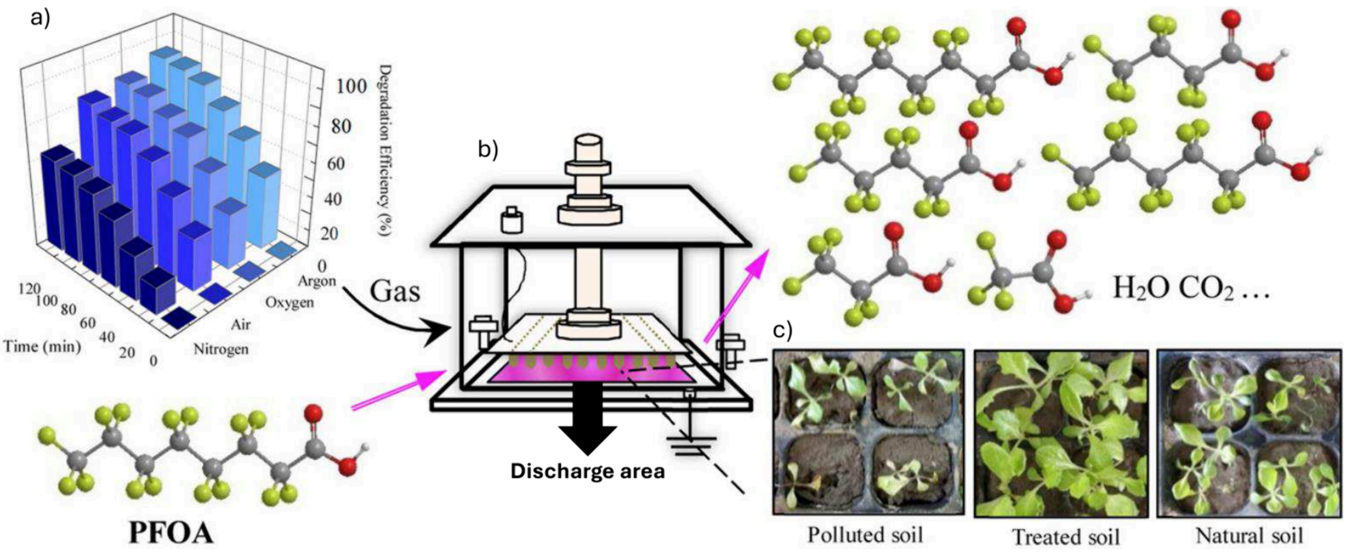

Figure 7. (a) Effects of different atmosphere (Ar, O_2, air, nitrogen) on degradation efficiency as a function of time, (b) plasma based experimental setup for soil treatment, and (c) lettuce grown on polluted soil, CAP treated soil, and natural soil. Reproduced with permission from ref 68. Copyright 2020 Elsevier.

results (Figure 6) showed 99% PFOS degradation after 40 min of treatment. Particularly, long-chain (>6 carbon) PFSA and FTS demonstrated the highest degradation efficiency under plasma treatment, achieving reductions of 99.9%, 94.7%, and 100% for PFOS, PFHxS, and 6:2 FTS (fluorotelomer sulfonate), respectively, after 120 min of treatment.

4.2. Soil Treatment

CAP has been employed for both in situ and ex situ degradation of contaminants in the soil.[53,88−91] Aggelopoulos et al.[88] conducted ex situ soil treatment using a dielectric barrier discharge (DBD) plasma reactor to remove non-aqueous phase liquid (NAPL), assessing the remediation efficiency in relation to soil heterogeneity, NAPL concentration, and treatment duration. The experimental setup included a DBD reactor connected to a high-voltage power supply capable of generating an alternating voltage between 12 and 14 kV (peak-to-peak) at a frequency of 40 kHz, enclosed within a Plexiglas box. Results indicated that the removal efficiency ranged from 97% to 99% at an energy density of 100,000 J g_{soil}^{-1}.[88]

Zhan et al. studied the degradation of PFAS (PFOA) in soil using CAP, examining the influence of various parameters such as discharge power, electrode spacing, air flow rate, soil pH, pollutant concentration, and soil moisture. The experiments utilized pulsed plasma corona discharge, as depicted in Figure 7.[68] The experiments established optimum degradation efficiency (71%, 75%, and 77%) at discharge powers of 38, 47, and 62 W, respectively, with an optimum electrode gap of 10 mm within 120 min of treatment. The results also showed that at soil moisture of 1%, 10%, and 15%, the removal efficiency is 71%, 75%, and 60%. Within the high-voltage plasma discharge, water molecules ionize to form hydrogen ions and electrons. PFOA degradation reduced as soil pH increased from 2.9, 6.3, and 10.9 with degradation efficiencies of 79%, 71%, and 56% favorable for acidic soil conditions. The results (Figure 7) show that higher degradation efficiency was observed using argon (Ar) gas followed by oxygen (O_2). Additionally, the CAP treatment seemingly helped to increase the concentration of ammonia nitrogen (10−462 ppm) and an increase in nitrogen fixing bacteria was observed, proving that the treatment improved soil fertility, as seen in Figure 7.

Table 1 gives an overview of different treatment technologies used for PFAS degradation and removal in soil and water. The studies indicate that CAP technologies have mostly been used for water treatment and only pulsed corona discharge has been used for PFAS degradation in soil.[68] Table 2 shows other PFAS degradation methods employed for water and soil treatment. These methods include biodegradation and bioaccumulation which are biological methods of PFAS degradation, adsorption using activated carbon, thermal treatment methods involving heating of contaminated samples, photocatalysis which involves bombarding contaminated media with photons to improve catalytic degradation, and ball milling which is a more mechanical approach involving rotating the PFAS media among metallic balls (steel).

4.3. Degradation Mechanisms and Pathways

The fundamental degradation mechanism of PFAS includes the generation of reactive oxygen and nitrogen species (RONS), initial decomposition, formation of short chain C−F bonds, byproduct production, and mineralization. The generation of reactive species in Cold Atmospheric Plasma (CAP) treatment for substance degradation occurs through a series of physical and chemical processes, including electron impact dissociation, electron attachment, and ionization of gas molecules.[98−101] These reactive species, such as ozone (O_3), hydroxyl radicals ($OH^\bullet$), and atomic oxygen (O), play a crucial role in the degradation of pollutants, including PFAS, through oxidation and/or fragmentation reactions.[102−106] The breakdown of PFAS directly depends on the efficiency of PFAS degradation and defluorination (eqs 1 and 2).

$$\text{Defluorination efficiency [\%]} = \frac{F^-}{\text{PFAS}_0 \times N_F} \times 100 \tag{1}$$

$$\text{Degradation efficiency [\%]} = \frac{\text{PFAS}_{\text{degradation}}}{\text{PFAS}_0} \times 100 \tag{2}$$

where F^- represents fluoride ions, N_F denotes the quantity of fluorine atoms, PFAS_0 is the initial concentration of PFAS, and

Table 1. Overview of CAP Technology Tested for PFAS Degradation in Water and Soil

reactor type (atmosphere)	PFAS	media	treatment time	process	degradation efficiency	limitations	refs
Corona discharge (Ar)	(a) PFDA and 8:2 FTS, (b) PFOS, (c) PFOA and PFHpS	Water	(a) 20 min, (b) 40 min, (c) 120 min	Reactive oxygen and nitrogen species degrade PFAS. The diffuser produces bubbles which interact with the liquid and corona discharge to degrade the PFAS.	>99%	Due to increased dissipation in the corona discharge for effective treatment it has low energy efficiency and requires further modifications for large-scale and long-term applications.	55, 56
Pulsed streamer (Air)	PFOA	Milli-Q and tap water	30 min	Production of reactive species as in the case of corona discharge aid in PFAS degradation.	>84%	Faster degradation was observed in tap water compared to Milli-Q water. Difficulty in scaling-up for field deployment.	92
Pulsed corona (O_2)	PFOS	Water	180 min	The use of oxygen directly promotes the production of reactive oxide species for PFAS degradation.	80%	Possible byproduct formation and accumulation due to incomplete degradation of PFAS compounds. Difficulty in scaling up.	84
Pulsed corona discharge (Ar)	PFOA	Soil	120 min	Electrons and the use of argon gas proved to be efficient in the degradation of PFAS.	71%	Further modifications needed for in situ applications. Limited research on PFAS degradation in soil using CAP. Nitrogen gas showed the lowest affinity for PFAS degradation.	68

Table 2. Other Remediation Techniques for PFAS

treatment	PFAS	media	treatment time	process	degradation efficiency	limitations	refs
Biodegradation (*Acidimicrobium* sp.)	PFOS and PFOA	Water	100 days (anaerobic conditions)	Microorganisms break down the PFAS long chains in water, using the pollutants as energy.	67%	Requires very long treatment time because PFAS does not readily biodegrade.	32, 33
Bioaccumulation (*Pseudomonas* sp.)	PFOS and PFOA	Soil	10 days (aerobic conditions)	Microorganisms breakdown the PFAS long chains in soil, using the pollutants as energy.	32.00%	PFAS is converted into stable medium chains which requires long treatment time.	34, 35
Adsorption (Granular activated carbon)	PFOS	Water	33.5 h	Activated carbon functions as a granular filter medium for removing the PFAS pollutant.	>80%	It is not very efficient in removing dissolved particles hence not all PFAS is removed. Routine maintenance is required and might be costly due to filter change.	36−38, 93, 94
Thermal treatment	PFOS and PFOA	Soil	10−14 days	Typically, high temperature (600−700 °C) is required to vaporize the PFAS by incineration.	99.99%	Energy-intensive and costly treatment method. There is risk of further environmental pollution due to the release of toxic gas which is challenging to capture.	39
Photocatalysis	PFOA	Water	180 min	Photogenerated radicals form potent oxidizing agents for breaking down the pollutants.	23%	PFAS is not completely destroyed but left in contaminated media especially in large media volume which may require longer treatment time.	40, 95−97
Ball milling	PFOS and PFOA	Soil	16 h	Rotation of the soil particles among steel balls causing reduction reaction.	99.98%	Increased treatment time. In-situ application is limited so there is a need for excavation and off-site treatment.	41, 42

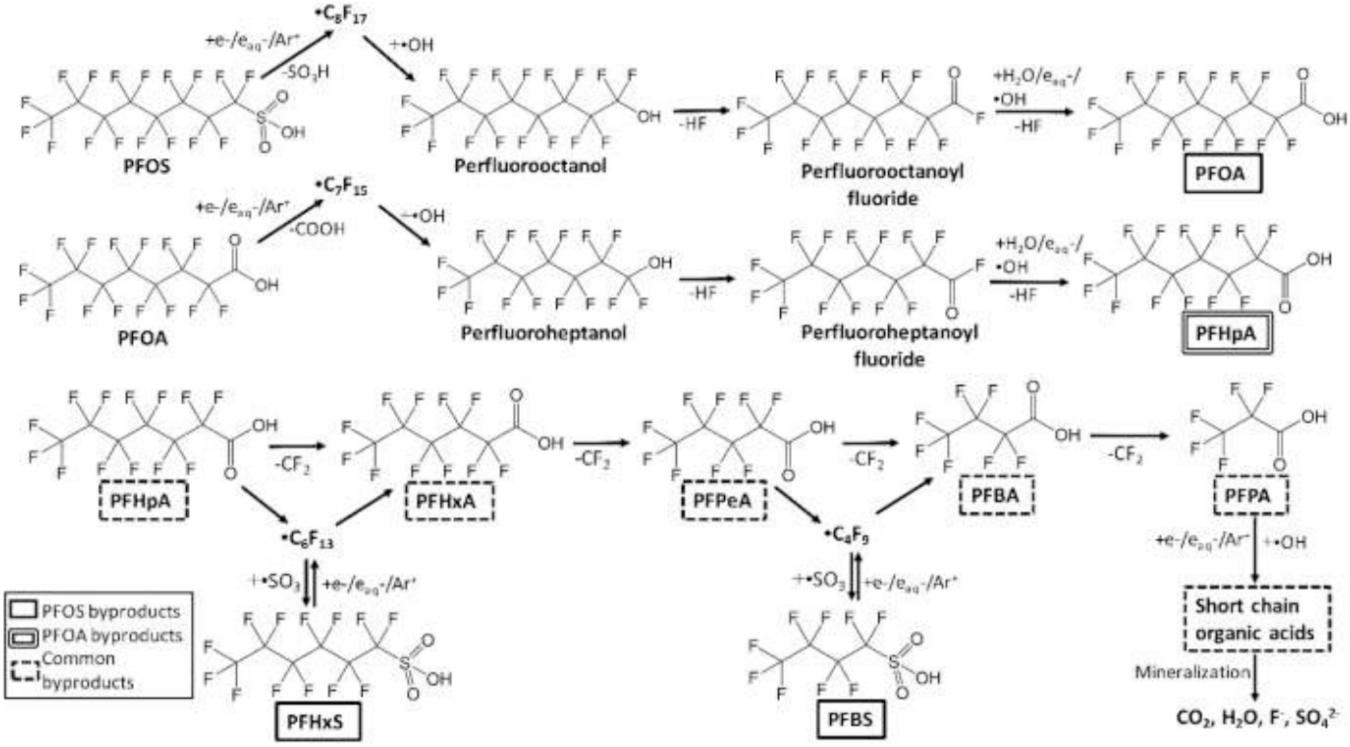

Figure 8. Reaction pathways for PFOS and PFOA degradation with CAP treatment. Reproduced from ref 4. Copyright 2019 American Chemical Society.

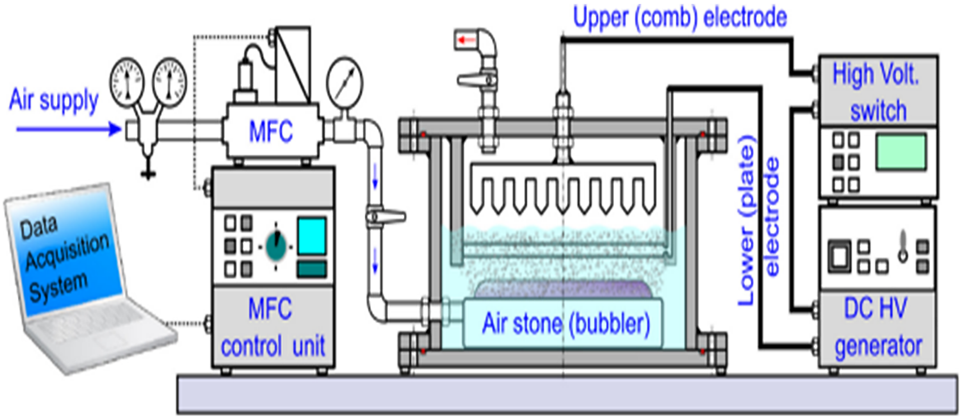

Figure 9. Experimental setup developed for the CAP treatment of PFAS compounds in water. Adapted with permission under a Creative Commons [CC-BY 4.0] license from ref 114. Copyright 2022 MDPI.

$\mathrm{PFAS}_{\mathrm{degradation}}$ refers to the concentration of degraded PFAS.[4,102] CAP is also capable of generating reactive species for advanced oxidation processes (AOPs). Combining ozone, ultraviolet radiation, peroxide, and catalytic reaction, AOPs employ highly reactive oxidant species with relatively low selectivity, such as hydroxyl radicals ($^{\bullet}$OH) and sulfate radicals ($SO_4^{\bullet-}$), to cleave C−F bonds and disrupt head groups along with C−C bonds.[89,92,107,108]

In addition, PFAS can be defluorinated through the breaking and substitution of the strong H−F bonds, reducing their toxicity and facilitating subsequent detoxification steps. Currently, an effective mechanism involves irradiating water solutions containing PFAS components with high-energy UV light, which generates hydrated electrons (e_{aq}^{-}), promoting the cleavage of H−F bonds.[4,109] As this degradation mechanism becomes more elucidated, numerous studies on enhancing the production efficiency of hydrated electrons are gaining attention. Some mechanisms are very similar to Advanced Oxidation Processes (AOPs), involving the use of UV as the primary energy source combined with the addition of anions (including OH^-, Cl^-, Br^-, and I^-). This will induce a substantial production of hydrated electrons in water, significantly improving degradation efficiency compared to direct UV irradiation.[110,111] Moreover, literature also suggests that reducing the concentration of competitive scavengers in the aqueous solution can maintain the effectiveness of anion action or improving the surface contact between the treatment apparatus and the sample can enhance degradation efficiency.[112] These aspects constitute recent advancements in research on PFAS degradation mechanisms.

During CAP treatment, PFASs undergo bond cleavage, whereby the reactive species react with the PFASs to yield a

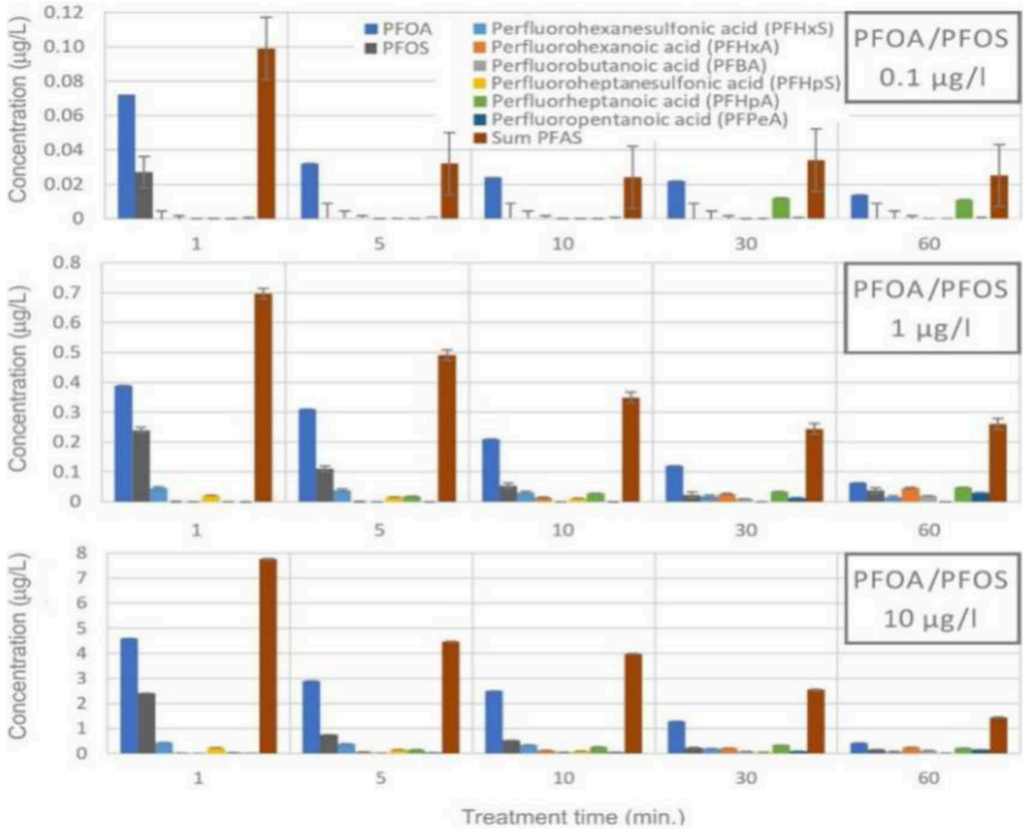

Figure 10. Transformation dynamics of PFOA and PFOS into short-chain PFAS compounds at varying initial concentrations during CAP treatment in distilled water. Reproduced with permission under a Creative Commons [CC-BY 4.0] license from ref 114. Copyright 2022 MDPI.

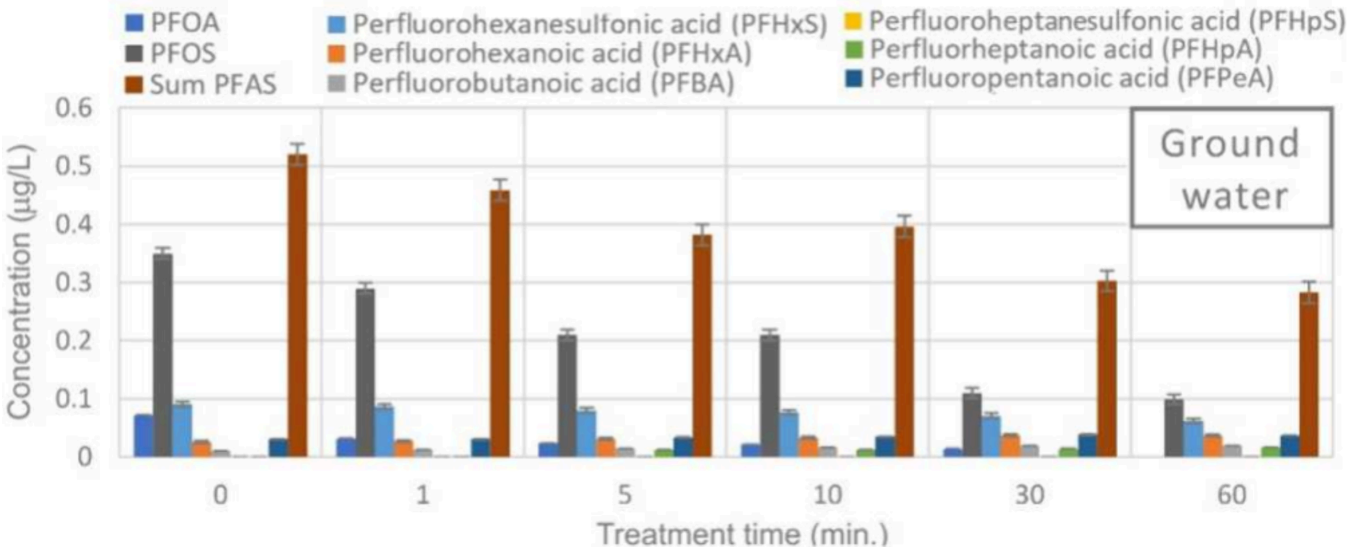

Figure 11. Degradation and transformation dynamics of initial PFAS compounds in groundwater samples into short-chain PFAS species. Reproduced with permission under a Creative Commons [CC-BY 4.0] license from ref 114. Copyright 2022 MDPI.

variety of less toxic and less bioaccumulative short chain PFASs.[4,30,56,87] The reaction pathways for the degradation of PFOS and PFOA shows that perfluoroheptanoic acid (PFHpA), perfluorohexanoic acid (PFHxA), perfluoropentanoic acid (PFPeA), and perfluorobutanoic acid (PFBA) are common byproducts of both PFOA and PFOS, as seen in Figure 8.[4,113] Typically CO_2, H_2O, F^-, and other small molecules are produced in the byproduct formation during degradation before the breakdown to less-toxic end products.[43]

Khan et al. investigated the dynamics of transforming long-chain PFOA and PFOS compounds into short-chain PFAS species in distilled water and groundwater.[114] The experiment utilized a batch reactor, designed as a cubical vessel with dimensions of 300 mm × 100 mm × 100 mm, constructed from poly(methyl methacrylate) (PMMA). (Figure 9).[114] The reactor was filled with 750 mL of PFAS-contaminated water for the experiments. The high-voltage (HV) electrode, constructed from stainless steel, featured a comb-like design with 11 pins, each 30 mm high, positioned toward the water surface. The distance between the HV electrode pins and the steady water surface was 14 mm. A rectangular stainless steel plate, measuring 200 × 50 × 2 mm^3, served as the ground electrode and was located 12 mm below the water surface, creating a total gap of 26 mm between the HV and ground electrodes. To maximize plasma exposure at the water surface, dry air was continuously introduced below the plate electrode, generating a dense flow of air bubbles. This was facilitated by two 100 mm long aquarium air stones (diffusers) placed at the bottom of the reactor. Solutions of PFOA and PFOS were prepared at concentrations of 0.1, 1, and 10 μg/L in distilled water and treated using a DC-plasma reactor.

The analysis of distilled water samples (Figure 10) demonstrated that over 90% of PFOA and PFOS were eliminated within 60 min of CAP treatment, regardless of the starting concentrations. Notably, more than half of the degradation occurred within the initial 5−10 min of the process. Figure 10 further depicts the degradation pathway, which follows a stepwise reduction from PFOA (C7) and PFOS (C8) to shorter-chain compounds such as PFHpA (C7), PFHxA (C6), PFPeA (C5), and PFBA (C4). Throughout the treatment, the total concentration of PFAS compounds in the samples continuously decreased, highlighting CAP's efficiency in breaking down these pollutants. This evidence indicates that CAP technology not only facilitates the conversion of long-chain PFOA and PFOS into shorter-chain PFAS compounds but also achieves a significant reduction in the overall PFAS burden within the treated samples. In contrast, groundwater treatment presented a more complex challenge. Samples obtained from soil remediation sites contained a heterogeneous mixture of long- and short-chain PFAS species, along with a diverse range of organic and inorganic contaminants. The analysis of the groundwater samples (Figure 11) revealed the presence of short-chain PFAS compounds after CAP treatment, along with varying dynamics in the transformation of long-chain to short-chain PFAS compounds at different treatment durations. The results also indicated that prolonged CAP treatment led to lower overall degradation levels. Additionally, a significantly greater variety of short-chain PFAS was observed in the groundwater samples compared to the PFOA/PFOS solutions in distilled water.

Typically, the degradation of PFSAs and PFCAs proceeds via the removal of carboxyl or sulfur trioxide functional groups, resulting in decarboxylation (CO_2 release) or the production of sulfur trioxide (SO_3). The hydroxyl radical ($^\bullet$OH), a highly reactive transient species, is predominantly formed in situ during ozone (O_3)- and ultraviolet (UV)-driven processes through the activation of stable precursors like hydrogen peroxide (H_2O_2).[115−117] While the second-order reaction rate constants of diverse radicals with PFAS remain inconclusive, their estimated values for PFOS and PFOA are markedly lower than those observed for the majority of organic pollutants.[117]

5. METHODS OF ASSESSING CAP EFFICACY FOR PFAS DEGRADATION

Different methods exist for assessing CAP efficacy in PFAS degradation. They include analytical chemistry techniques, treatment optimization, monitoring reactive species generation, and long-term stability studies.

5.1. Analytical Chemistry Techniques

Analytical chemistry techniques such as liquid chromatography−mass spectrometry (LC-MS) or gas chromatography−mass spectrometry (GC-MS) can be used to assess PFAS concentration before and after treatment, allowing assessment of the degradation efficiency.[118] GC-MS is suitable for analyzing volatile chemicals as degradation products and byproducts. High performance liquid chromatography (HPLC) often coupled with mass spectrometry is another method for separating PFAS compounds. It is particularly useful for distinguishing between various PFAS isomers and degradation products.[119,157] Fourier Transform Infrared Spectroscopy (FTIR) can be used to detect changes in functional groups during PFAS degradation.[120] It is useful for understanding the chemical structures of degradation products. Nuclear Magnetic Resonance (NMR) Spectroscopy can be used to provide detailed information on the molecular structure of PFAS degradation products.[119] It is particularly useful for identifying structural changes and elucidating degradation pathways. Ion Chromatography (IC) is used to detect ionic degradation products, such as fluoride ions, released during the breakdown of PFAS. It is often used in combination with other techniques to provide a comprehensive analysis of degradation products. These methods can be used for soil and water treatment.[54,121]

5.2. Treatment Optimization

Treatment parameters such as treatment time, power, gas composition, and electrode spacing can be systematically varied to optimize treatment efficiency. This method includes implementing different treatment configurations to identify optimal conditions for PFAS degradation. Different configurations of electrodes and choice of dielectric material (e.g., parallel plate, coaxial, needle-plate) can impact the distribution and intensity of the plasma field, affecting the uniformity and efficacy of PFAS degradation.[122,123] Adjusting the voltage and frequency of the plasma discharge can influence the generation of reactive species and the overall energy efficiency of the degradation process. Higher voltages typically increase the intensity of the plasma, potentially enhancing PFAS breakdown but also increasing energy consumption. Input power can be varied to calculate the energy consumption per unit of degraded PFAS or compare the energy efficiency of CAP treatment to other methods of soil and water remediation to determine the cost effectiveness of CAP treatment. The type of gas (e.g., argon, helium, and air) and varying its flow rate can significantly affect the nature and concentration of reactive species produced by CAP devices. For instance, using air or oxygen can introduce additional oxidative radicals that may enhance degradation rates.[77,124,125] As a result, RONS concentration can be greatly influenced by the volume of ionized gas which forms when the primary plasma species (ions, electrons, radicals, and dissociated molecules) interact with the liquid phase.[126]

5.3. Monitoring Reactive Species Generation

CAP reactors generate RONS including ozone, hydroxyl radicals, nitrogen ions, and atomic oxygen which are essential for PFAS or contaminant degradation.[127] Monitoring the generation of these reactive species using techniques such as optical emission spectroscopy (OES), mass spectrometry, and electron paramagnetic resonance (EPR) can help determine the efficacy of CAP treatment. The noninvasive OES diagnostic technique is widely used for real-time monitoring of reactive species in plasma systems. By analyzing the light emitted during electronic transitions, the OES detects transient species such as atomic oxygen (O), nitrogen (N), and hydroxyl radicals ($^{\bullet}$OH) through their characteristic spectral lines. This makes it particularly effective for understanding the chemical dynamics of plasma discharges.[128,129] Mass spectrometry, more so, provides detailed information about the composition and quantity of reactive species by ionizing the molecules and analyzing their mass-to-charge ratios. This technique is particularly useful for detecting and quantifying both stable and transient species, offering a complementary approach to OES in understanding the efficacy of CAP treatments.[130] Meanwhile, EPR specializes in the detection and characterization of short-lived free radicals such as superoxide and hydroxyl radicals through spin-trapping techniques. By reacting spin traps with these radicals, EPR generates stable adducts with unique spectral signatures, enabling species-specific detection in both plasma and biological systems. Electron paramagnetic resonance (EPR) spectroscopy is a technique utilized to investigate materials containing unpaired electrons. The fundamental principles of EPR are similar to those of nuclear magnetic resonance (NMR); however, EPR involves the excitation of electron spins rather than the spins of atomic nuclei. Instead of monitoring nuclear transitions within the sample, EPR measures the transitions of unpaired electrons under the influence of an external magnetic field.[131] Together, these complementary techniques provide a comprehensive toolkit for characterizing reactive oxygen and nitrogen species (RONS), advancing the understanding of plasma-induced processes and their applications.[132] In general quantification of reactive species, (ozone, hydroxyl radicals, hydrogen peroxide) can help assess the oxidative capacity of the plasma, correlating with the degradation efficiency of PFAS.[127,133,134] It can also help elucidate the degradation pathways of PFAS. For example, the identification of specific intermediates or byproducts formed during treatment can indicate which reactive species are involved in the degradation process and how they interact with PFAS molecules. Additionally, real-time monitoring allows for dynamic adjustments to the CAP system ensuring optimal conditions are maintained throughout the treatment process.[135,136]

5.4. Long-Term Stability Studies

Long-term stability studies involving the monitoring of PFAS concentrations, degradation intermediates, and byproducts in CAP-treated samples over time help to determine PFAS degradation efficiency and potential. These studies can be carried out on the consistency of degradation performance, system durability, energy efficiency and cost effectiveness, degradation byproducts and pathways, and environmental and health safety. This will involve toxicity assessment and regulatory compliance.[136]

6. FACTORS AFFECTING CAP EFFICACY FOR PFAS DEGRADATION

General PFAS degradation procedures using CAP would include building the CAP reactor, preparing the polluted sample, pretreatment analysis of PFAS concentration treatment of the sample, and post-treatment and data analysis. Initial and final PFAS concentration assessment is typically done using analytical techniques like liquid chromatography and mass spectroscopy (LC-MS) followed by optimizing treatment conditions to investigate the effects of certain factors. The methods of accessing CAP efficacy can be useful to determine how certain factors tend to affect CAP efficacy for degradation. They include plasma parameters, treatment time and temperature, reactor design configuration, contaminant type and concentration, presence of co-contaminants, and sample pH.

6.1. Plasma Parameters

The effectiveness of CAP systems for the degradation of PFAS can be greatly impacted by a number of plasma parameters, which include gas composition and flow rate, generation of reactive species, and input power.[73,137,138] Optimizing these plasma generation conditions can enhance the production of reactive species that are responsible for degradation. The use of different gases in plasma generation has been shown to impact degradation efficiency for a given treatment time as seen in the reviews. Argon gas has been shown to have a higher degradation efficiency for PFAS removal from soil and tap water, while air or oxygen is best suited for distilled water treatment. The generation of reactive species, like ozone, hydroxyl radicals, nitrogen ions, atomic oxygen, and other reactive species produced by the CAP system, and their concentration are important factors in the breakdown of per- and polyfluoroalkyl substances (PFASs). Higher voltages increase the electric field strength, enhancing the ionization of gas molecules and the production of reactive species such as hydroxyl radicals, ozone, and other reactive oxygen species.[139,140,156] However, excessively high voltages can lead to greater energy consumption and potential damage to the plasma reactor components. The frequency of applied voltage affects the stability and intensity of the plasma discharge.[47] The gas flow rate influences the residence time of reactive species in the plasma region and their interaction with the contaminated media.[124] Optimal flow rates ensure sufficient treatment time between reactive species and PFAS without diluting the reactive species concentration. The power input to the plasma system determines the energy density, influencing the production and concentration of the reactive species. Higher energy densities can enhance degradation but may also lead to higher operational costs.[73,141]

6.2. Treatment Time and Temperature

Extended treatment time can lead to longer PFAS exposure and interaction with reactive species that aids in degradation. Prolonged treatment times can result in the formation and subsequent degradation of intermediate byproducts. This can either improve overall degradation efficacy or, in some cases, lead to the accumulation of less degradable intermediates.[142] Determining the optimal treatment time is crucial to balance the efficacy of PFAS degradation and energy consumption of the CAP system. Excessive treatment times may not significantly improve degradation efficiency but will increase the operational costs. Higher temperatures can enhance the generation of reactive species such as hydroxyl radicals, ozone, and other RONS.[4] This increase in reactive species concentration can accelerate the degradation of PFAS. In soil treatment, the temperature can affect the desorption of PFAS from soil particles, making them more accessible to reactive species. In water treatment, temperature influences the solubility and mobility of PFAS, affecting their interaction with reactive species. Temperature also affects the stability and uniformity of the plasma discharge. Higher temperatures may lead to more stable plasma, which consistently generates reactive species. However, excessively high temperatures might destabilize the plasma and reduce efficiency.[143] In summary, longer treatment times and higher temperatures generally enhance the degradation efficiency by allowing more exposure to reactive species. However, prolonged treatment times and high temperatures can easily lead to high power consumption and formation of volatilized toxic compounds.[144−147]

6.3. CAP Reactor Configuration and Design

The plasma reactor configuration and design can affect the plasma generation and interaction between the generated species and the contaminated samples. This may include electrode spacing, configuration, and reactor type which may affect the homogeneity of treatment.[140,147,148] The dielectric barrier and corona plasma discharge are the most commonly used setup for degradation of PFAS or other chemicals, surface sterilization, and material decontamination/treatment.[144,148,149] This particular factor also depends on the plasma discharge mode, which can be either continuous or pulsed.[150]

6.4. Contaminant Type and Concentration and Presence of Co-contaminants

Contaminant type, functional groups, physicochemical properties, and chemical structure of the PFAS or any other contaminant can affect their susceptibility to CAP application together with the initial concentration of contaminants.[139] Research has shown that some PFAS types are more susceptible to degradation even after shorter treatment times, having higher degradation efficiency than others as seen in the reviews using different plasma reactors.[55,114,151] The presence of co-contaminants can affect the CAP efficacy for degradation because of the possible formation or production of secondary toxic byproducts after treatment, especially during PFAS removal, potentially becoming more persistent. For groundwater treated samples, noticeable decrease in the degradation efficiency is due to increased concentration of short chain PFAS and presence of organic and inorganic co-contaminants.[114] Hence this effect can also alter reaction pathways and additionally cause plasma quenching leading to a decrease in reactive species generation.[139,152] Therefore, it is important to consider and mitigate the effects of co-contaminants when designing and implementing plasma based treatment processes for environmental remediation.[153]

6.5. Sample pH

Sample pH can significantly influence CAP efficacy by affecting the chemistry of treatment and optimization.[135,154] pH levels tend to be influenced by the generation of reactive species during water treatment. Therefore, it is important to consider the sample pH as a critical parameter during the operation. Study has shown that CAP treatments are more favorable under relatively acidic conditions for the sample being treated or sterilized especially for soil and water treatment. However, excessively low pH levels can lead to the generation of unwanted byproducts.[68,114,155]

7. CONCLUDING REMARKS AND FUTURE OUTLOOK

CAP technologies represent an innovative and promising approach for the degradation of PFAS, offering unique capabilities for generating highly reactive species, i.e., RONS, under ambient conditions. These reactive species have been shown to effectively break down the robust carbon−fluorine bonds in PFAS, leading to significant reductions in contamination levels. CAP has demonstrated success in applications such as the treatment of contaminated water and soil as well as in controlled laboratory setups targeting concentrated PFAS solutions. The potential for rapid degradation within relatively short treatment times highlights CAP's utility in addressing environmental pollution.

Although the rich literature on PFAS degradation using CAP shows promising results, there are a few outstanding challenges and considerations that could hinder scaling-up and practical field deployment, especially for complex applications like PFAS degradation in soil. Scaling up CAP systems for field use requires the development of reactors capable of delivering uniform plasma over large treatment areas while maintaining operational efficiency. Addressing variability in environmental conditions and PFAS concentrations will require robust and adaptable system designs. Furthermore, the generation of intermediate byproducts during PFAS degradation underscores the need for effective monitoring and control mechanisms to ensure environmental safety. Energy efficiency is another critical factor for practical implementation. Portable, low-energy plasma systems must be developed to enable field deployment, particularly in remote or resource-constrained areas. In addition, hybrid systems combining CAP with other treatment methods, such as chemical oxidation or advanced filtration, could enhance the degradation efficiency and broaden the range of applicable scenarios.

In conclusion, CAP offers a transformative opportunity for PFAS remediation, combining effectiveness with flexibility across various contamination settings. Overcoming challenges related to scalability, energy optimization, and byproduct management will be key to unlocking its full potential as a sustainable solution for addressing PFAS pollution in real-world environments.

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Notes

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