



Innovations in PFAS remediation: a review on the growing role of cold plasma technology

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are a class of highly persistent and bioaccumulative environmental contaminants that have raised global concern due to their mobility, toxicity, and resistance to degradation. Conventional treatment technologies often fall short in achieving sufficiently high levels of destruction, particularly in complex matrices such as groundwater, wastewater, or soil. In this context, cold plasma technology has emerged as a promising, chemical-free approach for the effective and energy-efficient degradation of PFAS. Cold plasma produces a rich mixture of oxidative and reductive species, electrons, UV photons, and electric fields, capable of breaking down the strong carbon-fluorine bonds characteristic of PFAS compounds. This critical review provides a comprehensive assessment of recent research efforts on the application of cold plasma for PFAS remediation from aqueous and solid-phase environments. It systematically examines the influence of plasma types and reactor configurations, along with working gases, water matrices, plasma electrical parameters, and treatment conditions on degradation efficiency. Key factors such as plasma chemistry, energy consumption, pH, treatment duration, and PFAS structure are analyzed in detail. The review also addresses mechanistic insights, degradation pathways, and the main challenges for scaling cold plasma systems for real-world applications, including energy demand and integration with existing infrastructure. By critically synthesizing current findings, this review highlights the growing role of cold plasma in PFAS destruction and identifies research gaps and technological directions necessary to advance its practical deployment in environmental remediation.

1. Introduction

Water and soil pollution pose growing threats to ecosystems and human health worldwide. Among the primary contributors are persistent organic pollutants (POPs) such as pesticides, pharmaceuticals, and industrial chemicals, which are often resistant to natural degradation due to their stable molecular structures [1–5]. These substances can accumulate in environmental matrices, causing long-term contamination of essential resources. Owing to their toxicity, carcinogenicity, and endocrine-disrupting potential, they are a major concern for water safety and environmental quality.

Within this class of pollutants, *per-* and polyfluoroalkyl substances (PFAS) have garnered heightened attention. These synthetic compounds, used since the 1950s in a range of industrial and consumer products, are characterized by their exceptional chemical and thermal stability. Their persistence, bioaccumulation potential, and mobility in environmental matrices have resulted in widespread contamination and public health concern (e.g., cancer, liver damage, and immune system

disruption) [6–13]. PFAS are structurally defined by a fluorinated carbon chain (the “tail”), classified as short-chain or long-chain, terminated by a polar head group such as a carboxylic or sulfonic acid. This distinctive structure imparts PFAS with unique physicochemical properties, including high thermal and chemical stability, as well as repellency to water which justifies their extensive use and existence in environment [14,15]. Furthermore, the carbon-fluorine (C–F) bond, among the strongest in organic chemistry (bond dissociation energy: 480–560 kJ/mol), renders these compounds highly resistant to conventional oxidation, hydrolysis and biological breakdown.

PFAS are now referred to as “forever chemicals” due to their extreme persistence. While over 4000 PFAS have been identified, their environmental and toxicological profiles are still under investigation. Their detection in remote ecosystems has raised global concern about their long-range transport and ecological impact. In response, regulatory bodies, such as the European Commission, have introduced stringent guidelines, lowering acceptable concentrations in water to ng/L levels.

Conventional treatment strategies, including adsorption (e.g.,

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activated carbon) and ion exchange, offer only temporary removal without degradation, leading to secondary pollution and disposal concerns [16–23]. Among advanced oxidation processes (AOPs), only a few, such as electrochemical oxidation, sonolysis, and photocatalysis, have shown partial success in breaking PFAS structures [24–34].

More recently, plasma-based treatment technologies have emerged as promising alternatives. In particular, cold atmospheric plasma (CAP) has shown strong potential due to its chemical-free operation and the ability to generate a suite of highly reactive oxygen and nitrogen species (RONS), including hydroxyl radicals ($\bullet\text{OH}$), singlet oxygen ($^1\text{O}_2$), atomic oxygen ($\text{O}\bullet$), ozone (O_3), superoxide anion radicals ($\bullet\text{O}_2^-$), hydrogen peroxide (H_2O_2), nitrites, nitrates, as well as UV photons and electromagnetic radiation [35–48]. Notably, plasma systems can be powered by various gases (air, argon, nitrogen, helium, or mixtures), without requiring added chemicals, improving sustainability.

However, PFAS degradation presents unique challenges. Unlike conventional organic contaminants that can be effectively broken down by hydroxyl radical-based AOPs, PFAS require more specialized mechanisms capable of cleaving the exceptionally strong C–F bond. Cold plasma has shown promise in this regard, owing to the combined action of oxidative species, such as $\bullet\text{OH}$, H_2O_2 and O_3 , and plasma-induced reductive species, most notably hydrated electrons (e_{aq}^-), which are increasingly recognized as critical agents in disrupting C–F bonds. The coexistence of oxidative and reductive species is believed to enable parallel degradation pathways, increasing the likelihood of defluorination and mineralization of PFAS compounds. Nevertheless, the overall degradation efficiency depends on a complex interplay of factors, including plasma generation method, reactor configuration, discharge mode (e.g., dielectric barrier or corona), energy input, gas composition, environmental matrix, and treatment duration [49–56]. Additionally, partial degradation often leads to transformation products (TPs) that may retain fluorine atoms or toxicity, necessitating complete mineralization strategies. Understanding the complex interplay between plasma chemistry and PFAS degradation pathways is therefore critical.

Cold plasma has only recently emerged as a remediation strategy for PFAS. Systematic studies have appeared mainly over the past decade, with the number of publications on PFAS treatment growing rapidly, reflecting increasing interest in its potential as a sustainable alternative to conventional approaches. Most work to date has focused on lab-scale investigations aimed at elucidating the mechanisms of PFAS degradation by plasma, while a first field demonstration was only reported recently [57].

This review consolidates and critically examines nearly the complete body of available studies (emphasizing to those of the last 6–8 years), with the objective of identifying key operational parameters, contrasting divergent findings, and extracting mechanistic insights that can guide future reactor design and real-world application. The review aims to (i) summarize advances in plasma reactor configurations and discharge types and identify operational parameters but also physicochemical properties of under treatment medium, and matrix effects that influence treatment outcomes (Section 3), (ii) explore PFAS degradation pathways and the role of plasma species (Section 4), (iii) identify the defluorination efficiency and total organic removal of treatment methods (Section 4), (iv) provide a comparative analysis of PFAS destruction methods in terms of energy efficiency and effectiveness, and (v) assess current limitations and challenges for upscaling and field implementation (Section 5). This scientific, critical, and in-depth evaluation of existing studies integrates mechanistic insights with practical design considerations, providing a strategic foundation for advancing cold plasma technologies from experimental research to scalable, sustainable solutions for PFAS remediation. The review highlights cold plasma as a versatile and increasingly mature approach, while outlining key research priorities such as optimizing energy efficiency to enable the effective degradation of both parent PFAS molecules and their transformation intermediates.

2. Cold atmospheric plasma as a pollutant degradation method

2.1. Key characteristics and principles

Cold plasma is increasingly recognized as a powerful tool for environmental remediation, especially for addressing persistent organic pollutants in water and soil [35]. Its effectiveness stems from the generation of a diverse array of reactive agents, including radicals, ions, UV photons, and charged particles, that can attack and degrade a broad spectrum of contaminants. Applications demonstrated to date range from wastewater and soil treatment to pathogen inactivation and the breakdown of complex organic molecules.

Optimizing the efficacy of cold plasma requires a comprehensive understanding of the multiple factors influencing its performance. These factors include the type of plasma discharge, reactor design, and the specific interaction mode between plasma and the contaminated medium. Plasma systems are commonly based on dielectric barrier discharge (DBD), corona discharge, or plasma jets, each with distinct modes of species generation and pollutant interaction, offering unique advantages and limitations [39,58–62].

The breadth of research activity in this field is significant, with more than 1200 published studies available in academic databases addressing cold plasma applications for water and soil remediation [63]. This scientific momentum underscores the strong interest and potential of cold plasma as a sustainable and scalable treatment technology.

Plasma systems can be broadly classified according to their mode of interaction with the contaminated medium: direct, indirect, and bubbling configurations [35,39]. In direct discharge systems, plasma is generated directly within the medium, either in solution or in soil through electrodes immersed in the solid matrix [64–67]. This setup promotes the formation of reactive species directly at the contaminated medium. In contrast, indirect discharge systems generate plasma in the gas phase above the liquid/solid surface, with reactive species subsequently diffusing into the water or soil. While this setup minimizes electrode degradation, its efficiency may be limited by the shallow penetration depth of short-lived reactive species [68,69], which primarily interact with pollutants at or near the water/soil surface, reducing treatment uniformity throughout the bulk liquid/solid. The bubbling mode, wherein plasma is generated within gas bubbles injected into the liquid, has emerged as a particularly promising strategy. This approach enables efficient mass transfer, extended residence time, and uniform distribution of reactive species, resulting in rapid and energy-efficient degradation of various contaminants [39,70–72].

It is also critical to recognize that the structural characteristics of the target pollutant play a decisive role in treatment outcomes. Cold plasma systems primarily function through the generation of a complex and dynamic blend of reactive species as outlined in Section 1. These include short-lived radicals and long-lived oxidants that collectively initiate a cascade of degradation reactions. Numerous studies have validated plasma's effectiveness against a wide range of pollutants, including volatile organic compounds (VOCs), pesticides, pharmaceuticals, and other persistent contaminants found in water and soil.

A fundamental strength of cold plasma lies in the breadth of reactive species it produces, which engage in a series of interconnected chain reactions. These involve not only interactions between the parent pollutants and plasma species but also secondary reactions involving degradation intermediates and newly formed species [73]. This intricate reaction web significantly contributes to the system's high efficiency. However, it also introduces complexity in elucidating the underlying mechanisms.

Therefore, while cold plasma is recognized as a broadly effective tool for pollutant degradation, its mechanisms are highly sensitive to system conditions. These include reactor geometry, gas composition, discharge type (e.g., DBD vs corona), treatment mode (e.g., in-liquid vs gas-liquid interface), water/soil matrix composition, and pollutant type. Understanding and controlling these variables is essential for optimizing

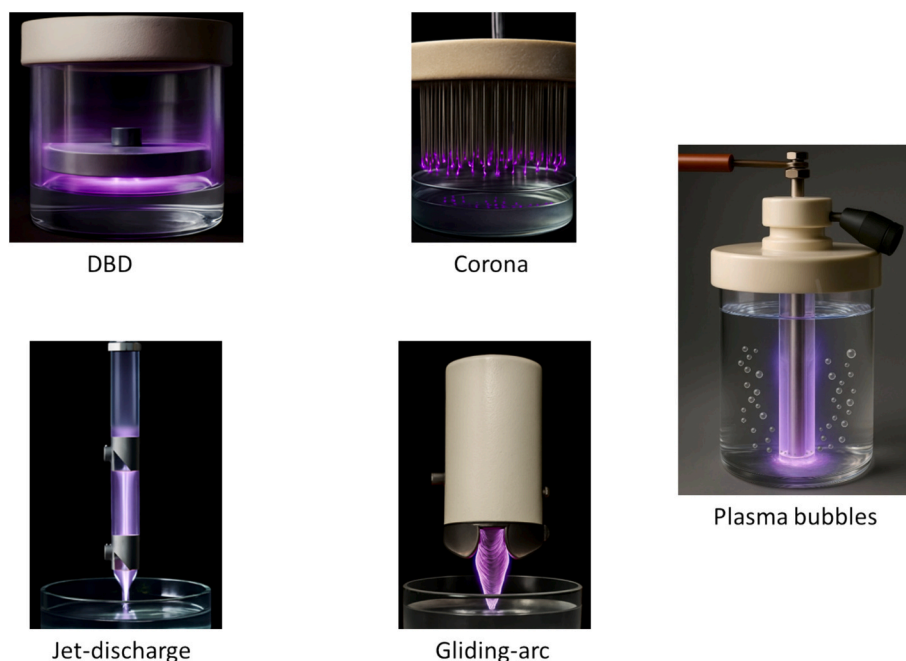


Fig. 1. Schematic representation of various cold atmospheric plasma reactor configurations applied in the degradation of *per*- and polyfluoroalkyl substances (PFAS).

treatment efficiency and enabling the transition of cold plasma from laboratory research to real-world environmental applications.

2.2. Influence of plasma conditions on species formation and pollutant degradation

The composition and reactivity of plasma-generated species are strongly influenced by plasma gas, reactor configuration, and operating parameters (e.g., HV waveform), while degradation efficiency and pathways depend on the specific pollutant.

The plasma reactive species include a diverse mix of oxidizing species, such as $^1\text{O}_2$, H_2O_2 , O_3 , $\bullet\text{OH}$ and nitrogen-based compounds (e.g., ONOO^- , $\text{NO}\bullet$, NO_2^-), as well as highly reductive species such as e_{aq}^- . The dominant plasma-generated species are strongly dependent on the discharge gas: air plasmas yield mixed reactive oxygen and nitrogen species (ROS/RNS) enabling multiple oxidative pathways, argon plasmas favor e_{aq}^- , oxygen plasmas produce strong ROS, and nitrogen plasmas generate RNS. Consequently, the plasma gas directly governs the balance between oxidative and reductive pathways, shaping pollutant degradation efficiency and mechanisms [74,75]. Notably, for highly recalcitrant pollutants like PFAS, hydrated electrons have been identified as particularly critical [76,77], favoring the use of argon-based plasmas or configurations that promote reductive species generation.

This gas-dependence is inherently pollutant-specific. For instance, in a study on cephalixin degradation, oxygen and air plasmas achieved >99.9 % and 96.1 % degradation after 20 min of treatment, while nitrogen plasma achieved only 69.8 % under identical conditions [78]. Conversely, in the degradation of PFOA, air, argon, and nitrogen plasmas exhibited significantly higher degradation efficiencies compared to oxygen plasma, with degradation rates of approximately >99.9 %, 95.1 %, and 98.8 %, respectively, after 30 min, whereas oxygen plasma showed a substantially lower efficiency of ~63 % under similar conditions [79]. These contrasting results underscore that no plasma gas provides a universal solution; rather, degradation pathways depend on the interplay between pollutant structure and the reactive species generated.

Reactor configuration also plays a decisive role in shaping plasma chemistry and efficiency. A comparative study of plasma bubble and

gas-liquid DBD systems for the degradation of dyes, including methylene blue (MB), methyl violet (MV), methyl orange (MO), and sunset yellow (SY), highlighted that degradation performance depends both on the reactor configuration and the molecular structure of the pollutants. Plasma bubbles yielded faster and more energy-efficient degradation for MB, MV, and MO, while SY responded better to gas-liquid DBD [39,80]. This was attributed to the differences in plasma-liquid interactions between the two systems resulting in different plasma species formation: plasma bubbles predominantly generated short-lived species like $\bullet\text{OH}$, effective for rapid degradation of some dyes, whereas gas-liquid DBD produced higher concentrations of certain long-lived species which were more effective for SY.

Thus, pollutant degradation via plasma is dictated by a multifactorial interplay of plasma gas, discharge mode, reactor design, and pollutant structure. Factors such as coexisting salts or organics in real matrices may further enhance or inhibit degradation. This complexity makes mechanistic generalizations difficult and highlights the need for case-specific optimization and mechanistic investigation. Given these challenges, it is essential to recognize that a complete understanding of plasma mechanisms is inherently complex and pollutant-dependent.

While cold plasma offers broad applicability for the degradation of organic pollutants, the treatment of PFAS presents unique demands. The highly stable C—F bond in PFAS necessitates specific reactive environments that differ from those effective for conventional organics. The following sections critically evaluate PFAS degradation using cold plasma, with a focus on the influence of plasma gas composition, reactor configuration, water matrix, physicochemical properties, and plasma operating parameters.

3. Cold atmospheric plasma for PFAS degradation

3.1. The effect of reactor configuration and discharge type

The configuration of a plasma reactor is pivotal in shaping both the efficiency and energy demand of PFAS degradation. Because of the exceptional chemical stability of PFAS, reactor designs must ensure not only the robust generation of reactive species but also their efficient transfer to the target contaminants. Over the past decade, diverse plasma reactor configurations have been developed and tested for PFAS

treatment in water, each characterized by distinct plasma-liquid interaction dynamics, reactive species profiles, and energy requirements (Fig. 1).

Among the earliest and most extensively studied designs are DBD reactors operating at the gas-liquid interface, valued for their simplicity of operation and versatility. A recent study demonstrated that a falling film water DBD system could effectively degrade perfluorocarboxylic acids (PFCAs) of varying chain lengths, achieving a maximum of 99.1 % degradation and 60.9 % defluorination efficiency [81]. These results suggest that pollutant accumulation at the gas-liquid interface enhances the contact between plasma generated reactive species and PFAS molecules. However, the energy efficiency of gas-liquid DBD systems is generally limited due to the restricted penetration of short-lived species into the bulk liquid phase, confining treatment efficacy primarily to the upper water layers.

To overcome this limitation, bubbling reactors coupled with above-liquid plasma have been developed [77]. In these configurations the discharge is produced above the liquid surface, while gas is introduced into the liquid through fine or medium bubble diffusers. The use of bubbles increases the gas-liquid interfacial area and provide a transport pathway for reactive species generated at the gas-liquid boundary to enter the bulk liquid. This enhanced interfacial transfer and the ensuing interfacial chemistry improve the exposure of PFAS molecules to oxidizing/reductive species and can therefore enhance PFAS degradation efficiency. In a PFOS related study, >90 % degradation was achieved at an electrical energy per order (E_{EO}) of 1.6 ± 0.1 kWh/m³ using fine bubbles, compared to 8.4 ± 2.5 kWh/m³ with medium bubbles, emphasizing the importance of bubble size and interfacial area [77]. One study for PFOA degradation using a needle-plate pulsed discharge reactor integrated with a water jet (NPDW), in which microbubbles with different carrier gases were introduced to enhance interfacial reactions, reported an E_{EO} of approximately 241 kWh/m³ [82].

A comparative study of gas-liquid DBD versus plasma bubble reactor (where plasma is generated directly inside the gas bubbles injected into the liquid) under air provided further insights [79]. Under air plasma, gas-liquid DBD systems achieved ~99 % PFOA degradation in 20 min, whereas plasma bubble reactor achieved lower degradation efficiency. The superior performance of the air-liquid DBD system was attributed to PFOA's surfactant nature, which promotes its accumulation at the plasma-liquid interface. However, future studies should focus on an extensive comparison between gas-liquid DBD and plasma bubble systems under argon atmospheres to further elucidate the influence of reactive species and discharge localization.

Corona discharge systems have also been explored for PFAS degradation, where performance is largely governed by plasma-liquid interfacial dynamics. For instance, a nanopulsed corona discharge achieved degradation of PFOS in water with an energy yield at 50 % conversion (G_{50}) of 22 mg/kWh under argon or oxygen, and 27 mg/kWh under helium [49]. Plasma jets, consisting of non-thermal plasma plumes directed at water surfaces, have also demonstrated promise in lab-scale PFAS degradation studies [83–85]. High removal efficiencies (>90 %) were reported in deionized water for long-chain PFAS, including PFOS (99.9 %), PFHxA (94.6 %), and ADONA (94.8 %). However, in more complex matrices such as tap water or synthetic effluents, degradation efficiencies decreased markedly, especially for short-chain PFAS: in tap water, PFOS and GenX reached only 50 % and 32 % degradation, respectively, while short-chain compounds were degraded by ~10 % [84].

Gliding arc plasma (GAP) discharges offer a promising alternative, with the advantage of generating larger plasma volumes at lower power densities. In one study, GAP reactors achieved >90 % degradation and > 25 % defluorination of long-chain PFAS in just one hour using air plasma, with E_{EO} values of 23.2 kWh/m³ for PFOS and 213.4 kWh/m³ for PFOA [86].

In a recent study that radial plasma discharge was used, a ≥ 99 % PFOA conversion was noticed in less than 2.5 min and 30 min in

solutions with initial concentrations of 41 μ g/L and 41 mg/L, respectively, with corresponding E_{EO} values 1.02–13.8 kWh/m³ depending on the conditions [87].

In terms of lifetime and penetration depth, the behavior of reactive plasma species varies markedly across reactor configurations and aqueous versus solid matrices. Plasma bubbles are particularly advantageous for delivering short-lived species such as \bullet OH and hydrated electrons (e_{aq}^-), since these are generated directly inside the bulk liquid. Before and upon bubble collapse, the radicals are released rapidly into the surrounding solution, making them immediately accessible to pollutants before decaying. This mechanism circumvents the diffusion barrier that limits gas-liquid systems, where short-lived species must first traverse the plasma-liquid interface and therefore remain confined to the upper nanometer layers of the water column, with penetration depths typically only 100–200 nm. By contrast, long-lived species such as ozone, hydrogen peroxide, and nitrate dominate in gas-liquid systems. Their much higher lifetimes allow them to diffuse deeper into solution and persist long enough to interact with contaminants.

In summary, plasma reactor configurations that promote intimate plasma-liquid contact and enhance the generation of short-lived reactive species, particularly hydrated electrons, tend to perform better in terms of PFAS degradation and energy efficiency. The distinct discharge characteristics of plasma types strongly influence the degradation pathways of PFAS. For instance, DBD systems typically generate diffuse plasmas with high ROS and RNS densities at the gas-liquid interface, favoring interfacial attack. Corona discharges, in contrast, produce filamentary plasmas with intense local electric fields, which can enhance direct electron-driven C–F cleavage. Plasma jets provide directed plumes with abundant short-lived species (\bullet OH, $O\bullet$, e_{aq}^-) but limited penetration into the bulk liquid, leading to high surface activity yet constrained volume treatment. On the contrary, gas-liquid reactors that introduce bubbles (e.g., air or inert gas sparging) promotes bulk-phase contact between PFAS and reacted species, improving degradation efficiency [82]. On the other hand, plasma-bubble reactors, in which plasma discharges occur inside the liquid phase, enhance the transport of reactive plasma-derived species through the large interfacial area and direct delivery via bubble collapse and interfacial mixing [79]; in particular, the potential of argon-based plasma bubbles for PFAS degradation remains to be thoroughly explored. These differences highlight that the physical nature of the discharge (uniform vs filamentary, localized vs volumetric) and the dominant plasma chemistry (oxidative and reductive) in each reactor type are key determinants of the mechanistic routes observed in plasma-driven PFAS degradation.

Nevertheless, reactor design alone does not dictate performance. As explored in the following sections, additional variables play crucial roles in defining cold plasma treatment outcomes for PFAS degradation.

3.2. The effect of plasma gas

The composition of the plasma feed gas is pivotal in shaping the reactive environment (oxidative, reductive, or mixed) thereby determining the dominant species formed and the mechanisms driving PFAS degradation. Crucially, treatment success depends not only on the abundance of reactive species but also on their specific ability to attack the exceptionally stable C–F bonds in PFAS. Plasma-based degradation typically employs discharge atmospheres such as air, oxygen, nitrogen, or argon, either individually or in combination.

Air plasmas generate a rich mixture of reactive oxygen species (ROS) and reactive nitrogen species (RNS). Ionization and dissociation of oxygen (O_2), nitrogen (N_2), and water vapor by energetic electrons produce key species such as $O\bullet$, \bullet OH, and $NO\bullet$, as shown in reactions below:



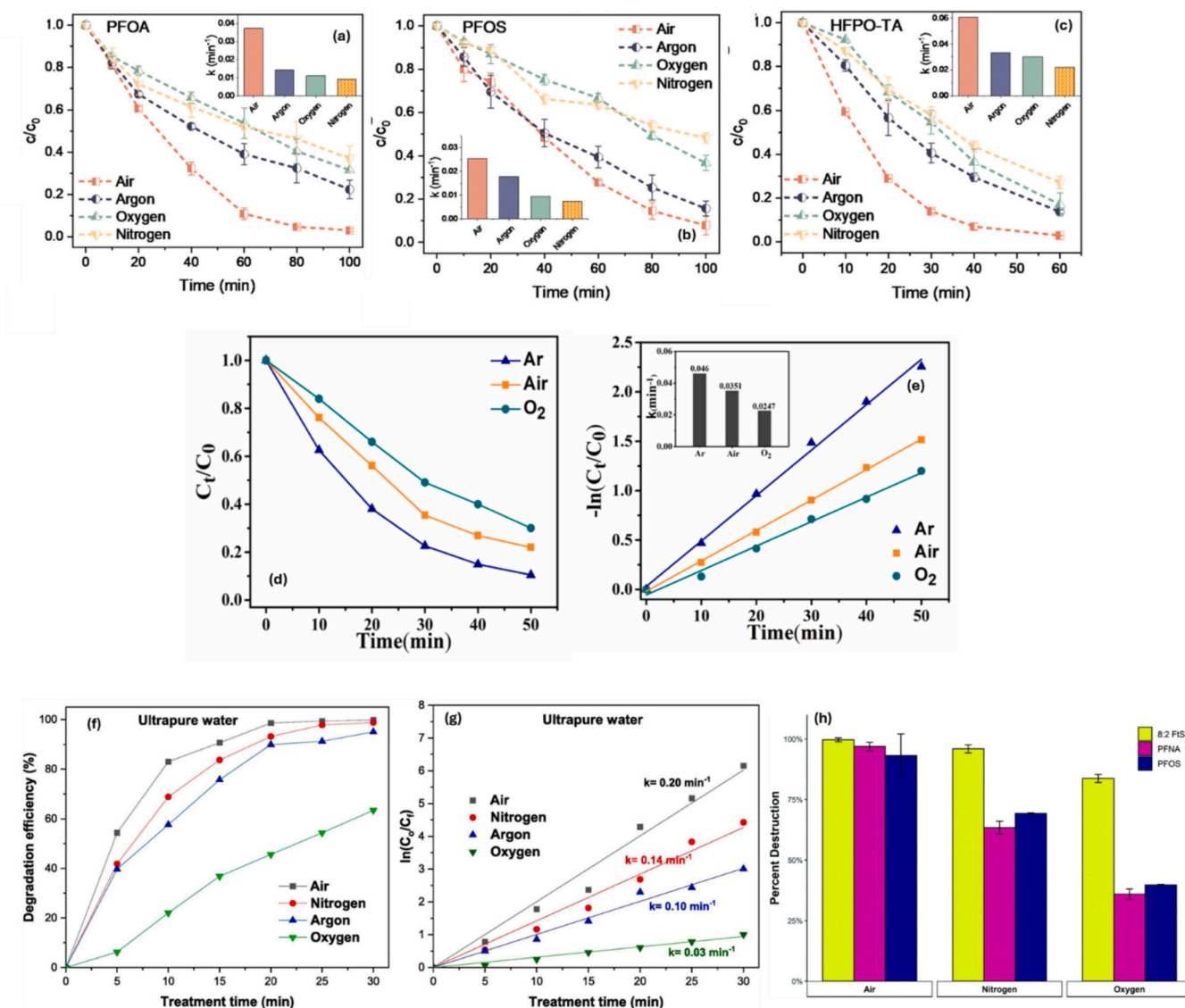


Fig. 2. The effect of plasma gas in two different reactor types (i.e., DBD and gliding arc) on (a) PFOA, (b) PFOS and (c) HFPO-TA degradation. System parameters are: discharge power = 92.3 W, liquid circulation rate = 150 mL/min, gas flow rate = 0.6 L/min, $C = 20 \mu\text{M}$. Figure generated with permission based on data reported in ref 81. (d) The degradation efficiency of PFOA under plasma from three different gases, (e) First-order kinetics and kinetic constants under plasma from different discharge gases. Figure generated with permission based on data reported in ref 88. (f) PFOA degradation efficiency at different plasma gases and (g) pseudo-first-order PFOA degradation kinetics at different plasma gases (HV waveform: micropulses; plasma reactor: gas-liquid DBD; initial PFOA concentration: 1 mg/L; water matrix: ultrapure water). Figure generated with permission based on data reported in ref 79. (h) Effect of gas used for plasma discharge on destruction of PFNA, PFOS, and 8:2 FTS ($C = \sim 100 \text{ mg/L}$, gas flow = 50 L/min, power = 150 W, $t = 1 \text{ h}$). Figure generated with permission based on data reported in ref 86.



These primary radicals undergo secondary reactions to form additional species such as O_3 , ONOO^- , and other nitrogen oxides. Air-fed plasmas thus create a mixed oxidative/reductive environment particularly well suited to initiate multiple PFAS degradation routes.

Several studies report superior degradation performance with air compared to pure O_2 . For instance, one study showed higher degradation efficiencies for PFOA, PFOS, and HFPO-TA in air plasma compared to Ar, N_2 , or O_2 atmospheres [81], with performance following the order: air > Ar > N_2 , O_2 (Fig. 2a-c). While Ar plasma discharge produces higher electron density and gas-phase hydroxyl radicals ($\bullet\text{OH}$) than air plasma, the degradation efficiencies for PFOA and HFPO-TA were slightly lower in Ar than in air, suggesting that ROS and RNS generated uniquely in air plasma work synergistically to destabilize PFAS molecules. Another recent comparison of air and argon in ultrapure water

also found air to be slightly more effective than argon, with oxygen again performing worst (Fig. 2f-g) [79]. In addition, the superior performance of air compared to both N_2 and O_2 is consistent with previous studies (Fig. 2h) [85,86] where air demonstrated the highest effectiveness, followed by nitrogen, while oxygen showed significantly lower performance. However, it has been also reported higher PFAS degradation efficiency in pure argon, followed by air and oxygen (Fig. 2d-e) [88].

Inert gases like argon represent a fundamentally different plasma chemistry. In argon plasmas, ionization primarily produces energetic electrons and Ar^+ ions through:



These energetic electrons interact with water vapor and dissolved gases at the plasma-liquid interface to generate hydrated electrons (e_{aq}^-) and hydrogen atoms ($\text{H}\bullet$) via:

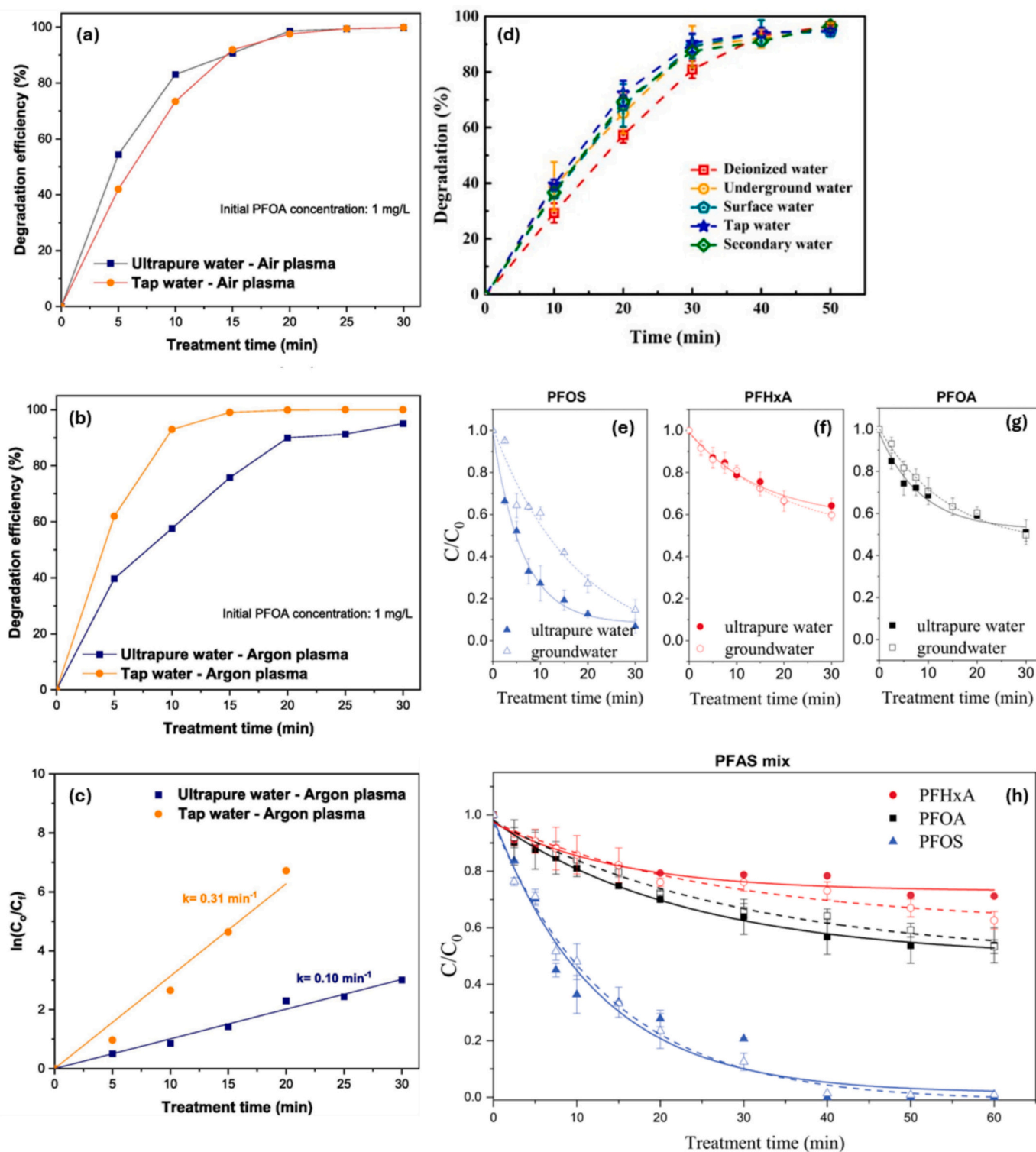
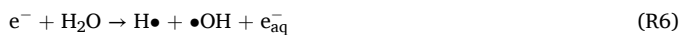


Fig. 3. Comparison of PFOA degradation between ultrapure and tap water under (a) air-plasma; (b) argon-plasma and (c) pseudo-first-order PFOA degradation kinetics under argon-plasma (HV waveform: micropulses; plasma reactor: gas-liquid DBD). *Figure generated with permission based on data reported in ref 79.* (d) Degradation efficiency of PFOA during gas-liquid DBD process in different water matrices (Experimental conditions: power = 30.8 W; liquid flow = 320 mL/min. *Figure generated with permission based on data reported in ref 94.* Degradation of (e) perfluorooctanesulfonic acid (PFOS), (f) perfluorohexanoic acid (PFHxA) and (g) perfluorooctanoic acid (PFOA) treated individually in ultrapure and groundwater matrices. Degradation of perfluoroalkyl substances (PFAS) treated in mixture (h) in ultrapure (solid symbols, solid lines) and groundwater (open symbols, dashed lines). *Figure generated with permission based on data reported in ref 96.*



The formation of hydrated (or solvated) electrons is a critical

advantage of argon plasmas for PFAS degradation [89]. Hydrated electrons are highly reductive, possessing reduction potentials sufficient to cleave C–F bonds directly, bypassing the need for high-energy oxidative pathways. Recent studies, report that argon-fed plasma systems

consistently outperform other plasma gases [88,89].

Critically, the concentration and lifetime of hydrated electrons are maximized under argon plasma conditions, especially when powered by nanopulsed discharges, which enhance plasma uniformity and minimize species recombination [79]. In addition, The application of bubble-mediated transport to convey long-chain PFAS ($\geq C6$) to the gas-liquid interface, where exposure to argon-based plasma occurs, has demonstrated significant efficacy in promoting their degradation in both synthetic matrices and contaminated surface waters [89].

In contrast, oxygen-fed plasmas generate an oxidative environment, dominated by species such as atomic oxygen and ozone:



While ROS are strong oxidants, they are generally ineffective against PFAS. Consequently, oxygen plasma, dominated by oxidative pathways, has consistently shown lower degradation performance in multiple studies [79,86,88]. This limitation arises because typical ROS cannot efficiently attack the highly stable fluorinated chains, leading to incomplete degradation and higher energy demands. As a result, oxidative pathways alone, particularly those driven by O_2 -derived species, are insufficient to achieve effective PFAS degradation.

Nitrogen plasmas create environments rich in RNS and moderate electron densities through:



While RNS can play a role in PFAS degradation, nitrogen plasmas generally produce lower concentrations of hydrated electrons needed for efficient C–F bond cleavage. However, their contribution is evident in some studies under certain experimental conditions [79].

Further comparisons using gas mixtures such as argon-air or argon-oxygen, suggest that combining argon's electron density with oxidative or RNS pathways can improve PFAS degradation. Argon-air plasmas, in particular, provide more uniform discharges and increase species transport into solution [88].

Beyond water, cold plasma applications for soil remediation are emerging. Plasma treatment in soils faces additional challenges due to heterogeneity, moisture, and restricted electron mobility. Studies using corona discharge or gliding arc plasmas have reported partial PFAS removal from soil, with effectiveness depending on soil porosity and moisture [90,91]. In soil matrices, argon also reveals a dominance as feeding gas with a PFOA degradation efficiency of 85 %, compared to 75 %, 77 %, and 53 % under O_2 , air, and N_2 atmospheres, respectively [91]. Nevertheless, air-fed plasmas tend to dominate due to easier scalability, but the reduced mobility of reactive species in solid matrices compared to water can limit degradation rates. The formation of long-lived species such as ozone and nitric oxides within soil pores offers indirect degradation pathways, but complete mineralization remains more challenging compared to liquid systems.

An important distinction in soil treatment is that hydrated electron generation is typically much lower than in aqueous plasmas, due to both the limited availability of free water molecules and electron trapping by soil particles [92]. As a result, oxidative pathways dominated by species such as O_3 , NO_x , and $\bullet OH$ prevail, but these mechanisms are less effective against PFAS and therefore demand longer treatment times and higher energy inputs compared to water-based plasma processes. Continued optimization of plasma parameters, soil pre-conditioning (e. g., moisture adjustment), and reactor designs are necessary to improve the applicability of cold plasma for PFAS-polluted soil remediation.

Along with different species composition deriving from the different gases, the mechanism of reactive species transport in soils is

fundamentally different: plasma discharges can be generated either directly within soil pores or in the overlying gas phase. Unlike in water, where species mobility is largely diffusion-limited, soil systems involve multiphase transport where penetration depends not only on lifetime but also on connectivity of pore networks and water content. Consequently, aqueous PFAS degradation generally benefits most from short-lived reductive species, whereas soil remediation relies more on the mobility and persistence of long-lived oxidants capable of migrating through porous structures.

In conclusion, feed gas composition is undoubtedly a central determinant of the plasma chemistry and directly influences PFAS degradation mechanisms. Reported discrepancies between air and argon plasmas can be reconciled by recognizing that each favors distinct degradation routes: air plasma is often superior when interfacial ROS/RNS reactions dominate, whereas argon excels under conditions that preserve hydrated electrons for bulk reductive pathways. Air plasmas, offering a mix of ROS and RNS, provide versatile reactivity, while argon plasmas excel due to abundant hydrated electron formation. For aqueous PFAS remediation, argon-fed plasma, particularly when operated under nanopulsed regimes and coupled with optimized reactor configurations, currently offers the most effective and energy-efficient approach, whereas soil applications still face challenges due to mass transport limitations and reduced electron mobility. In contrast, oxygen plasmas generate strong ROS but lack the reductive potential needed for efficient C–F bond cleavage, while nitrogen plasmas supply crucial RNS but their performance is restricted by the lower hydrated electron production.

However, definitive conclusions require case-by-case evaluation. An integrated strategy that combines gas selection, reactor design, and treatment optimization is essential for advancing cold plasma-based PFAS remediation technologies.

3.3. The effect of water matrix

The physicochemical properties of the treated matrix, including chemical composition, ionic strength, organic matter content, pH, and conductivity, strongly influence the efficiency of plasma processes for PFAS degradation. While initial studies often employ ultrapure water to isolate plasma-pollutant interactions and establish baseline kinetics, real-world matrices such as tap water, groundwater, and wastewater introduce additional complexities that can significantly affect treatment performance.

In ultrapure water, the absence of background ions and organics allows more direct interaction between plasma-generated reactive species and PFAS molecules. However, in tap water, which typically contains bicarbonates, chlorides, sulfates, and natural organic matter, along with higher ionic strength and slightly alkaline pH, several matrix-induced effects emerge which may act either as prohibiting or promoting factors. For example, a direct comparison between ultrapure and tap water was performed using gas-liquid DBD plasma under both air and argon atmospheres for PFOA degradation [79]. Under air plasma, degradation efficiencies were similar in both matrices (Fig. 3a). Conversely, argon plasma treatment exhibited enhanced PFOA degradation in tap water. A 10-min treatment achieved ~93 % degradation in tap water, compared to ~58 % in ultrapure water. Extending the treatment to 15 and 20 min further improved degradation to 99 % and 99.9 %, respectively, versus 76 % and 90 % in ultrapure water (Fig. 3b-c) [79]. This enhanced performance in tap water under argon plasma was attributed to favorable matrix properties. Specifically, the slightly alkaline pH (~8.7), reduces the concentration of hydronium ions. Since hydrated electrons (e_{aq}^{-}) are key species in PFAS degradation and are quenched by H_3O^{+} ($e_{aq}^{-} + H_3O^{+} \rightarrow H_2O + \bullet H$), the alkaline environment preserves their availability. This observation is supported by earlier γ -irradiation studies that demonstrated improved PFOA degradation at alkaline conditions [93]. In contrast, acidic conditions in ultrapure or air-treated water may limit hydrated electron activity. Overall, the

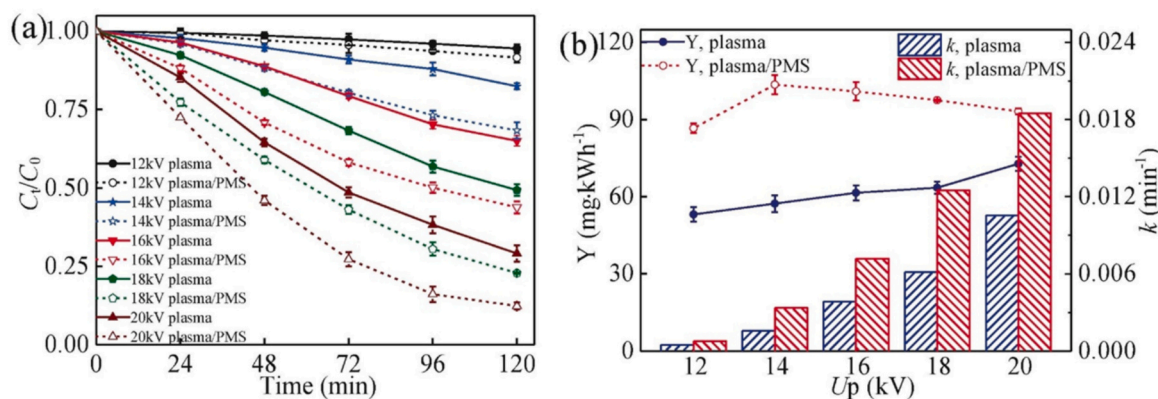


Fig. 4. (a) PFOA degradation, and (b) Y and k under different input voltages. Reaction conditions: [PFOA] = 5 mg/L, [PMS] = 445 mg/L, U_p = 12–20 kV, d = 5 mm, solution circulation rate = 300 mL/min, and P = 0.39–3.65 W, depending on input voltage. Figure generated with permission based on data reported in ref. 95.

observed matrix-dependent behavior underscores the capability of plasma to degrade PFOA effectively, with degradation efficiencies either maintained (air plasma) or further improved (argon plasma) in more complex and realistic water matrices such as tap water [79].

Interestingly, common water constituents, often seen as scavengers, can also act beneficially under specific plasma conditions. For instance, moderate concentrations of bicarbonate (HCO_3^-) can react with hydroxyl radicals to form carbonate radicals ($\text{CO}_3^{\bullet-}$), which possess notable oxidative potential:



Similarly, sulfate ions (SO_4^{2-}), often present in tap or treated water, can be converted to sulfate radicals ($\text{SO}_4^{\bullet-}$) via:

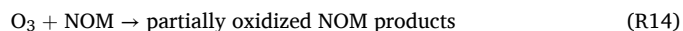


These species are effective in initiating PFAS degradation, including C–F bond cleavage [94]. Notably, deliberate addition of sulfate salts has been shown to increase PFAS solubility, enhance ionic strength, and stabilize plasma-liquid interactions, resulting in higher degradation rates and lower energy consumption [95]. Thus, sulfates can act synergistically by both generating secondary radicals and optimizing the overall plasma conditions for effective PFAS mineralization. For instance, the addition of 445 mg/L of peroxymonosulfate (PMS) increased PFOA degradation from 54.4 % to 81.0 % by enhancing reactive species formation (Fig. 4) [95]. These observations suggest that controlled additions of specific anions like carbonates and sulfates can be strategically used to enhance plasma performance for PFAS degradation.

Groundwater and surface water matrices introduce additional complexity to PFAS degradation. In a recent study, the influence of the water matrix on PFOA degradation by gas-liquid DBD was evaluated using five types of water: groundwater, secondary effluent, surface water, tap water, and deionized water [94]. After 50 min of treatment, PFOA removal exceeded 98.0 % in all matrices (Fig. 3d). Interestingly, removal efficiencies were slightly higher in natural and treated waters compared to deionized water. This was attributed to the structural design of this reactor, which enabled the generation of large amounts of effective radicals. In another study, where PFOS, PFOA, and PFHxA were evaluated as a mixture, their degradation patterns differed from those observed when tested individually [96]. Notably, PFOS showed slightly lower degradation efficiency in groundwater compared to ultrapure water when treated alone. However, in the mixture, similar degradation levels were observed in both matrices. PFOA exhibited consistent degradation (~46 %) across both water types, regardless of whether it was tested alone or as part of a mixture. Interestingly, PFHxA, which showed minimal matrix-dependent variation when assessed individually, degraded more effectively in groundwater when part of the mixture

(38 % vs. 29 % in ultrapure water). These findings suggest that PFAS degradation may be enhanced in complex matrices when compounds are present as mixtures, potentially due to altered reaction dynamics or indirect interactions among co-contaminants (Fig. 3e–h).

Other studies revealed that the presence of natural organic matter (NOM), humic substances, and other dissolved organics in environmental waters poses a challenge by scavenging plasma-generated oxidants [83,84,97]. Reaction such as:



compete with PFAS for reactive species, reduce plasma efficiency, and may introduce additional degradation byproducts. This necessitates longer treatment durations or adjustments in plasma operating parameters (e.g., applied voltage, gas flow rate) for effective PFAS degradation in complex matrices.

Since real wastewater contains a wide range of co-existing organic and inorganic constituents, its treatment is generally more challenging compared to synthetic matrices. In a recent study, the degradation and defluorination of PFAS were evaluated in both synthetic wastewater and real wastewater effluents to assess the performance of the plasma process under realistic conditions [98]. The results showed that although both degradation and defluorination percentages were slightly lower (by less than 10 %) in real wastewater compared to synthetic wastewater, highlighting a very low reduction in efficiency. This demonstrates that the non-thermal plasma reactor maintains high effectiveness even in complex effluents, confirming its potential for practical wastewater treatment applications.

In conclusion, matrix composition impacts the effectiveness of plasma-based PFAS degradation. While ultrapure water provides a clean benchmark, studies have shown that PFAS degradation in tap and groundwater can be comparable to, or even exceed, that in ultrapure water, despite the presence of additional ions and organics. Real-world applications therefore require a nuanced understanding of how ions, pH, and natural organic matter interact with plasma chemistry. Strategic additions (e.g., sulfate, carbonate) may enhance degradation, while NOM can inhibit it. Consequently, matrix engineering (e.g., controlled addition of ions such as sulfate) combined with tailored plasma operating conditions offers a promising path toward robust and scalable PFAS remediation.

3.4. The effect of physicochemical properties

The physicochemical properties of the treatment medium, particularly pH, conductivity, and the presence of surfactants, play a decisive role in determining the efficiency and mechanism of PFAS degradation by plasma. These parameters influence plasma-liquid interface chemistry, the generation and stability of reactive species, and ultimately the

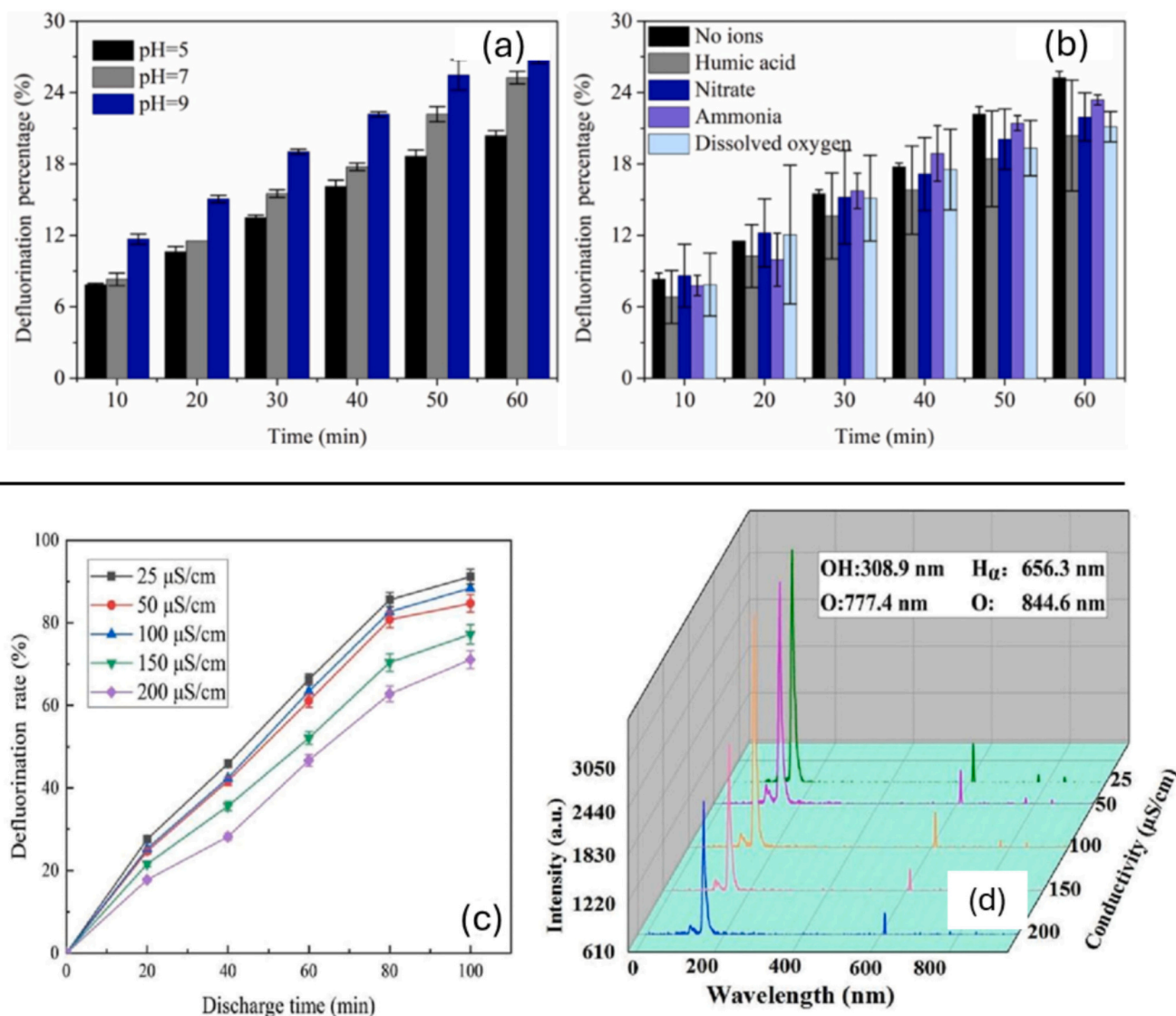


Fig. 5. Effect of (a) the initial pH and (b) coexisting components (initial pH: 7) on the defluorination percentages of PFAS-containing synthetic wastewater. Experimental conditions: (carrier gas: Ar; water flow rate: 100 mL/min; power: 60 W). Figure generated with permission based on data reported in ref [98]. (c) Effect of conductivity on the defluorination of PFOA, and (d) effect of conductivity on the luminescence intensity of free radicals. Figure generated with permission based on data reported in ref [100].

degradation pathways followed.

The influence of pH on plasma-induced degradation is multifaceted. A key consideration is the stability of hydrated electrons, which are widely recognized as highly effective for cleaving the robust C–F bonds in PFAS molecules. Hydrated electrons are more stable under alkaline conditions and are rapidly scavenged under acidic environments through reactions such as:



Thus, lower pH reduces the availability of e_{aq}^- , diminishing reductive degradation pathways. In a recent study, the degradation of PFOA using argon-fed plasma was significantly faster and more energy-efficient in tap water (pH ~8.7) than in ultrapure water (neutral pH), primarily due to enhanced e_{aq}^- stability [79]. The effect of pH on PFAS degradation varies with both the PFAS structure and the plasma system used. In general, alkaline conditions enhance defluorination by preserving e_{aq}^- , with studies reported a decrease in defluorination efficiency as pH dropped from 9 to 5 (Fig. 5a-b) [98]. However, exceptions exist depending on reactor configuration; for example, needle-plate

discharges with microbubbles may favor degradation under acidic conditions due to enhanced PFAS accumulation at the plasma-liquid interface via electrostatic attraction [82].

Conductivity also affects plasma discharge characteristics and degradation efficiency. While increases in conductivity at extended treatment times can promote the formation of certain reactive species, such as NO_3^- [99], moderate conductivity levels are generally more favorable for PFAS degradation. For example, a study examining PFOA degradation at varying conductivities showed that values above 100 µS/cm reduce defluorination efficiency during plasma treatment (Fig. 5c) [100]. High conductivity weakens the electric field, leading to a substantial decrease in the intensity of reactive species such as $\bullet OH$, $H\bullet$, and $O\bullet$ radicals (Fig. 5d). Consequently, higher ionic strength environments can inhibit PFAS degradation by suppressing plasma discharge and radical generation.

In addition to intrinsic matrix properties, the co-presence of surfactants like cetyltrimethylammonium bromide (CTAB) has been shown to significantly enhance PFAS degradation. CTAB forms complexes with negatively charged PFAS headgroups, promoting their migration to the

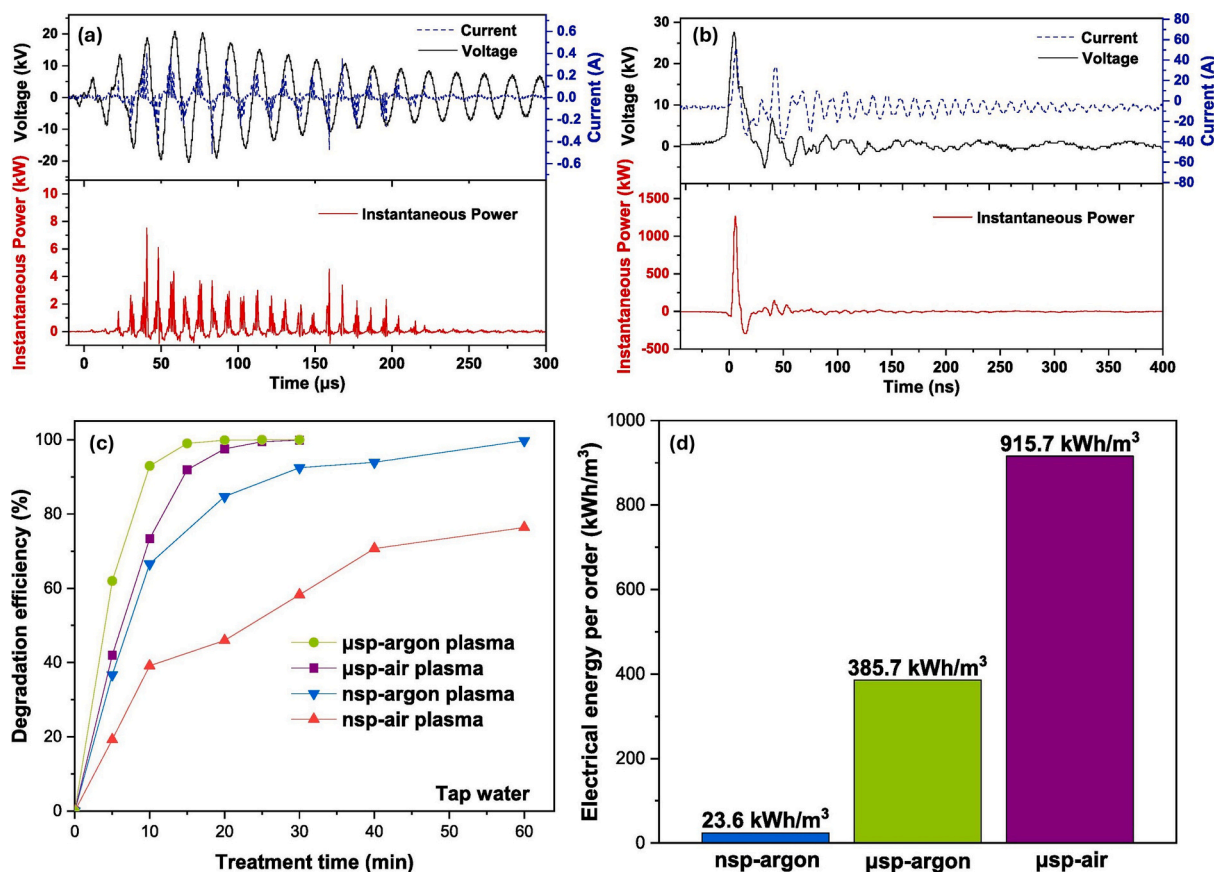


Fig. 6. Instantaneous voltage, current and power waveforms of the (a) micropulsed gas-liquid DBD and (b) nanopulsed gas-liquid DBD reactor during PFOA treatment (nanopulse voltage: 27.6 kV; nanopulse frequency: 200 Hz; micropulse voltage: 20.0 kV; micropulse frequency: 1000 Hz), (c) Comparison between HV microsecond pulses (μ sp) and HV nanosecond pulses (nsp) for PFOA degradation in tap water under air- and argon-plasma and (d) electrical energy per order (plasma reactor: gas-liquid DBD; initial PFOA concentration: 1 mg/L). Figure generated with permission based on data reported in ref. 79.

plasma-liquid interface. In one study, in the presence of CTAB, $\sim 99\%$ of the short-chain PFBS was removed from the bulk liquid and concentrated at the interface, where 67% of the concentrate was degraded and 43% of that amount was defluorinated within one hour [101], highlighting the importance of improved interfacial mass transfer for plasma-driven degradation [76,90,101].

In summary, the interplay of pH, conductivity, and surfactants critically shapes the reactive environment during plasma treatment, influencing the stability of key species such as hydrated electrons and the accessibility of PFAS molecules at the interface. Changes in pH and conductivity due to acidification, oxidation, and ion release create dynamic feedback loops that further affect plasma chemistry. Careful control of these evolving physicochemical conditions is therefore essential for maximizing the efficiency and selectivity of cold plasma systems in practical PFAS remediation.

3.5. The effect of plasma electrical characteristics

The effectiveness of plasma-based PFAS degradation is strongly influenced by the electrical parameters that govern plasma generation. Among these, the waveform of the high-voltage (HV) signal, the amplitude of the applied voltage, and the discharge frequency are particularly critical, as they dictate the nature and concentration of plasma-generated reactive species, and ultimately determine the energy efficiency and degradation kinetics of the process.

The temporal characteristics of the voltage waveform (especially pulse duration) have a significant impact on plasma behavior. Recent studies comparing HV nanosecond (ns) and microsecond (μ s) pulses have shown that while μ s-pulsed plasmas can produce faster PFAS

degradation, ns-pulsed systems offer markedly superior energy efficiency (Fig. 6a–d) [79]. For instance, under argon plasma conditions, μ s-pulsed discharge achieved rapid PFOA degradation ($\sim 93\%$ in 10 min, $>99.9\%$ in 20 min), but with a high electrical energy per order (E_{EO}) of ~ 385 kWh/m³. In contrast, the ns-pulsed system achieved slower degradation kinetics ($\sim 67\%$ at 10 min, complete only after 60 min), yet with a much lower E_{EO} of ~ 23.6 kWh/m³ demonstrating significantly improved energy efficiency. Nanopulsed air plasma, however, failed to achieve complete degradation. These findings highlight a clear trade-off between degradation rate and energy consumption: μ s pulses produce more reactive species due to higher energy input, while ns pulses minimize thermal losses and enhance the generation of high-energy electrons that selectively cleave strong C–F bonds. This makes ns-pulsed systems particularly attractive for sustainable and cost-effective PFAS remediation.

The amplitude of the applied voltage strongly influences PFAS degradation by increasing the electric field intensity within the reactor, which enhances electron-impact reactions and promotes the generation of reactive species [88]. Importantly, the effect of voltage on degradation efficiency is pollutant-dependent. For instance, in a study using a reverse vortex gliding arc plasma reactor [86], higher power improved degradation, but energy efficiency varied markedly between compounds. PFOS reached 90% degradation at 180 W with an E_{EO} of 23.2 kWh/m³, whereas PFOA required 255 W to achieve 75% degradation, with an energy consumption of 213.4 kWh/m³. This difference reflects both general power-related effects and the inherent structural differences between compounds, with sulfonate PFOS degrading more readily than carboxylate PFOA under plasma treatment [86].

Finally, the pulse frequency affects the concentration and continuity

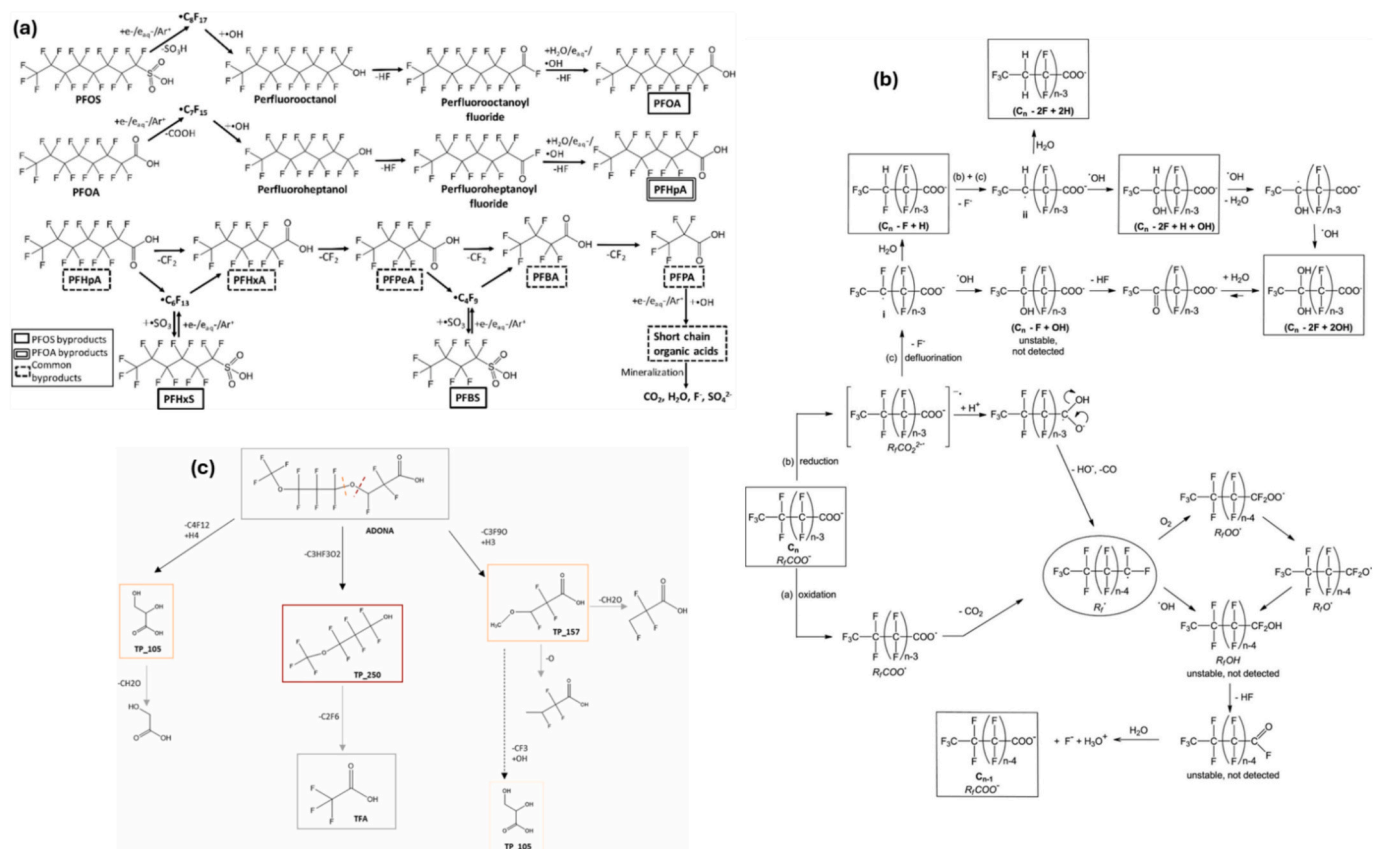


Fig. 7. Representative degradation pathways of different PFAS reported in the literature; (a) PFOS, PFOA, PFHpA, (b) PFCAs, (c) ADONA. Figure generated with permission based on data reported in refs 107 (a), 106 (b), 84 (c).

of reactive species in the plasma-treated medium. Increasing the frequency generally enhances the steady-state density of radicals and electrons, thereby improving PFAS degradation rates [102]. For example, in the nanosecond-pulsed degradation of PFOS, increasing the frequency from 0.5 kHz to 10 kHz accelerated PFOS removal. However, this improvement came at the cost of energy efficiency: higher frequencies increased power consumption more than tenfold while only reducing the time required to achieve 50 % degradation by approximately 50 % [102]. These results suggest that although high-frequency operation can enhance degradation kinetics, it must be carefully balanced with energy input to ensure practical and cost-effective treatment.

In conclusion, tailoring the electrical input parameters such as pulse duration, voltage amplitude, and frequency, is essential for optimizing plasma systems for PFAS degradation. While higher voltages and μ -pulses can boost degradation rates, ns-pulsed discharges offer superior energy performance. Pulse frequency, too, must be optimized to balance reactivity and efficiency. A mechanistic understanding of these parameters will be critical for the rational design and scale-up of plasma technologies for real-world PFAS remediation.

Evaluating each of the above-mentioned parameters in sections 3.1–3.5, it is observed that despite promising results, significant knowledge gaps remain for critical parameters governing plasma performance. For gas composition, the optimal balance between the various reactive species for effective PFAS degradation is not yet well defined, particularly under realistic water chemistries. A promising approach could be the use of air-argon mixtures, which combine the generation of reactive nitrogen and oxygen species (RNS/ROS) from air with the production of hydrated electrons and argon ions from argon, thereby exploiting complementary degradation pathways. Matrix effects, including natural organic matter and co-contaminant interactions,

introduce complexity that is insufficiently understood and can hinder reproducibility across studies. Similarly, the influence of electrical parameters such as voltage and frequency, on both degradation efficiency and energy demand may differentiate depending on the reactor configuration, underscoring the need for guidance toward reactor optimization in the context of upscaling and further deployment.

4. Mechanisms of PFAS degradation by cold plasma

4.1. Degradation pathways

The high variety of legacy and emerging PFAS has resulted a complex roadmap of degradation pathways that they follow. To that end, most recent analytical method for PFAS in air, water, abiotic solid matrices and biological matrices are comprehensive requiring meticulous analysis steps [103].

PFAS degradation by plasma involves a series of complex, multi-step reactions primarily driven by interactions between PFAS molecules and reactive plasma species. As already mentioned, among these species, e_{aq}^- are widely recognized as particularly effective, given their strong reducing potential that enables direct cleavage of C–F bonds. Recent ab initio molecular dynamics simulations have further revealed that the interaction between hydrated electrons and PFAS is more intricate than previously understood, differing significantly from simple excess electron interactions [104].

In addition to hydrated electrons, plasma electrons and argon ions (Ar^+) in argon-fed plasma systems also contribute to PFAS degradation. These species can initiate charge-transfer reactions, generating fluoro-carbon radicals that propagate further degradation [105,106].

The overall degradation pathway typically follows a stepwise mechanism (Fig. 7). Initially, an electron or ion attack cleaves a C–F

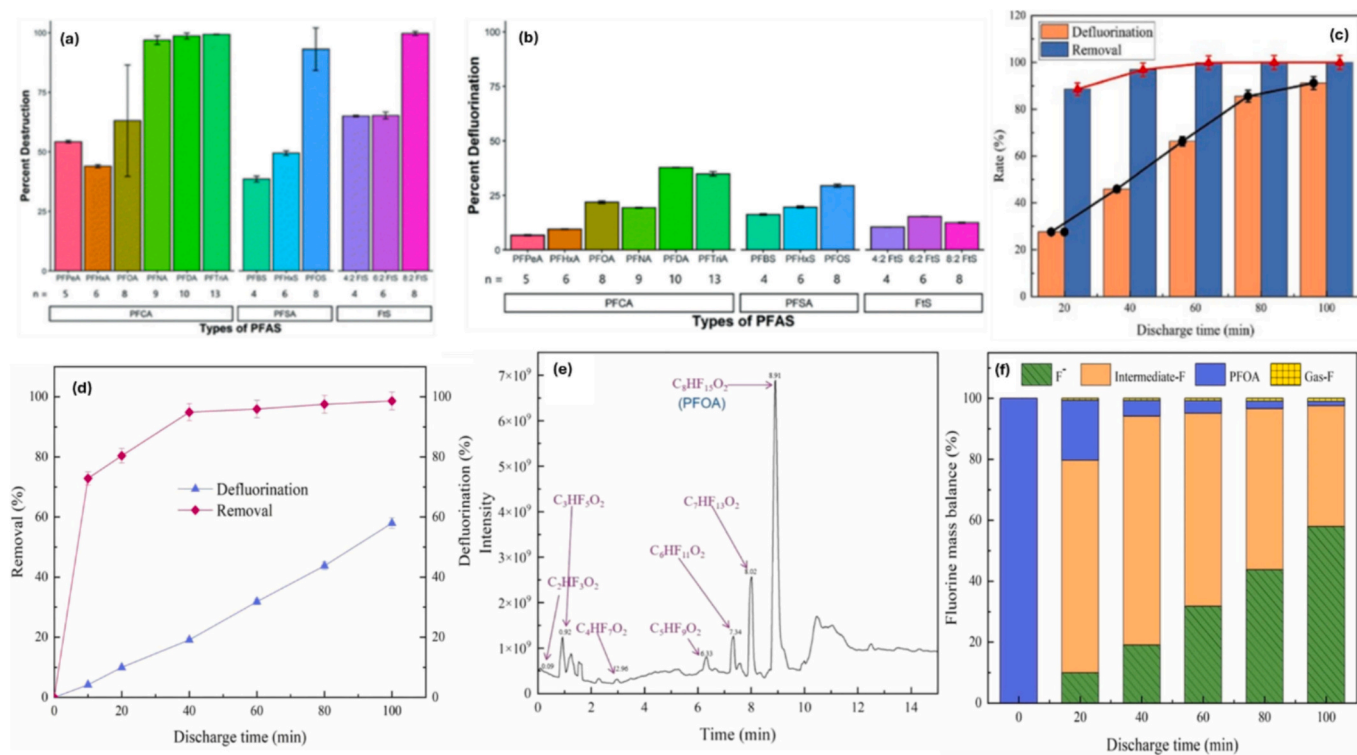


Fig. 8. (a) Percent destruction of PFCAs, PFSA, FtS as a function of chain length (C_nF_{n+1}). (b) Percent defluorination of PFCAs, PFSA, FtS as a function of chain length (C_nF_{n+1}). Experimental conditions: ~100 mg/L PFAS, 50 L/min air flow, 150 W power output setting, 1 h of treatment time. Figure generated with permission based on data reported in ref 86. (c) Degradation and defluorination of PFOA treated by microwave discharge plasma in liquid Figure generated with permission based on data reported in ref 100. (d) Degradation of PFOA treated by microwave discharge plasma in liquid combined with catalytic ions Fe²⁺. (e) Intermediate products of PFOA decomposition. (f) Mass balance of fluorine from PFOA degradation varies with time. Fluorine-containing intermediates (Intermediate-F), Fluorine containing gas (Gas-F) Figure generated with permission based on data reported in ref 116.

bond, releasing a fluoride ion and forming shorter-chain perfluoroalkyl carboxylic acids (PFCAs). These intermediates undergo further breakdown to yield CO₂, fluoride ions, and water. The degradation efficiency and progression along this pathway are strongly influenced by plasma operating conditions.

The specific degradation mechanisms can vary depending on the type of PFAS compound [84,87,94,106–109]. For example, in PFCAs and perfluoroalkyl sulfonic acids (PFSAs), degradation involves decarboxylation [110] or desulfonation [111], respectively, resulting in CO₂ release or the formation of SO₃. In these systems, •OH radicals, primarily produced via electron impact on water vapor, also contribute, albeit with low reaction rate constants compared to most organic pollutants.

While •OH radicals are often considered ineffective for directly cleaving C–F bonds, emerging evidence suggests they may participate in secondary degradation stages [112]. For instance, multiple parallel degradation routes have been proposed. In PFCAs, both direct decarboxylation and a hydroxylation–elimination–hydrolysis (DHEH) pathway have been identified [109]. In PFOS degradation, initial cleavage of the C–S bond, initiated by electrons or Ar⁺, and forms the reactive C₈F₁₇• radical, which can then generate shorter-chain PFCAs [105,107].

Overall, PFAS degradation by plasma can proceed via both reductive and oxidative pathways. Reductive mechanisms, dominated by hydrated electrons, are generally considered more efficient for direct C–F bond scission and fluoride release [113]. In contrast, oxidative species such as •OH, O₃, and reactive nitrogen species (e.g., •NO, ONOO⁻) primarily contribute to secondary transformations, including decarboxylation, desulfonation, and the oxidation of intermediates, which accelerate chain-shortening but often without full defluorination [114]. This interplay between reductive defluorination and oxidative fragmentation highlights the importance of tailoring plasma conditions (e.g., gas

composition, discharge environment) to favor pathways that maximize mineralization while minimizing the accumulation of partially fluorinated byproducts.

Advanced analytical techniques such as ion chromatography (IC) and ultra-performance liquid chromatography-mass spectrometry (UPLC-MS) have confirmed the formation of short-chain degradation products including PFPrA, PFBA, PFPeA, PFHxA, and PFHpA, as well as partially defluorinated intermediates. These intermediates typically follow a sequential chain-shortening pathway, ultimately leading to C–C bond cleavage and mineralization [92,115]. In emerging PFAS such as GenX and ADONA, degradation pathways differ from legacy PFCAs and PFSAs due to the presence of ether linkages and branched structures. Recent plasma studies indicate that ether bond scission is often the initiating step, generating perfluoroalkyl radicals that undergo subsequent chain-shortening reactions [84]. These intermediates may follow similar oxidative and reductive routes as PFCAs, ultimately producing shorter-chain perfluorinated acids and fluoride release, though with slower kinetics compared to PFOA or PFOS. Importantly, partially defluorinated products from GenX and ADONA have been detected, underscoring the need for more systematic mechanistic studies to fully elucidate their breakdown routes and assess potential toxicity [84]. Importantly, trifluoroacetic acid (TFA) has been identified as a terminal byproduct during the plasma degradation of both legacy and emerging PFAS, including GenX and ADONA. Owing to its high persistence and mobility, TFA frequently accumulates in treated solutions and is often still detected even after prolonged treatment times [101]. This underscores the critical challenge, that in spite of the plasma processes that effectively break down long-chain PFAS, achieving complete mineralization and preventing the buildup of stable short-chain acids including TFA remains an unresolved issue. Collectively, these findings highlight the versatility of cold plasma technologies in addressing both legacy and

modern PFAS compounds and underline the need for continued mechanistic studies to optimize treatment conditions for full mineralization.

4.2. Defluorination efficiency

Recent advances have demonstrated the potential of plasma for PFAS defluorination, though efficiencies remain highly variable depending on the compound and treatment conditions. For example, in a hybrid non-thermal plasma-ozonation reactor that achieved high overall PFAS degradation, defluorination efficiencies after 60 min were 8 % for PFBA, 12 % for PFHxA, 27 % for PFOA, and 40 % for PFDA [98]. Using a gliding arc plasma stabilized in a reverse vortex for PFAS removal, high degradation efficiencies were achieved; however, defluorination efficiency remained low (Fig. 8a-b) [86]. In another study, a microwave discharge plasma in liquid system achieved complete degradation of PFOA within 100 min, with 91.2 % defluorination (Fig. 8c) [100]. In a similar microwave discharge plasma in liquid, the degradation of PFOA reached 98.6 % after 100 min, whilst its defluorination was 58 % (Fig. 8d), highlighting that defluorination of PFOA is more difficult since its degradation produced short-chain PFAS (Fig. 8e) [116]. The corresponding fluorine mass balance is illustrated in Fig. 8f. Therefore, the defluorination of PFAS is more valuable for the actual water purification treatment than the removal of PFAS, since PFAS can produce short-chain fluorine-containing intermediates in the process of degradation, which will still cause serious secondary pollution to the water environment.

Therefore, while plasma treatments frequently report high PFAS degradation efficiencies, these values often overestimate true mineralization, as defluorination efficiencies typically lag well behind overall PFAS removal, and therefore PFAS complete defluorination remains a significant challenge. This disparity reflects the fact that plasma species readily disrupt headgroups or partially cleave carbon backbones, producing shorter-chain intermediates that retain C–F bonds [107]. This raises significant environmental and health concerns, as incomplete defluorination could result in the accumulation of intermediates that are equally or more harmful than the original PFAS. Factors such as PFAS chain length, molecular structure, co-contaminants, and reactor design can all influence the generation and interaction of plasma species. Notably, short-chain PFAS are often more resistant to degradation due to their high solubility and low interfacial adsorption, hindering their exposure to reactive species at the plasma-liquid interface and creating the need for additional agents to facilitate their transfer. Approaches such as using surfactants to enhance interface transfer have been attempted, but challenges including complete mineralization of parent molecules and degradation of intermediate byproducts such as TFA remain, highlighting the need for optimized reactor designs and complementary treatment methods to improve contact with reactive species. These strategies are critical for achieving more consistent and complete defluorination of environmentally persistent short-chain PFAS [101]. This fact highlights that complete fluoride release requires successive scission steps, which are kinetically and energetically more demanding, especially for short-chain PFAS.

In conclusion, the formation of partially defluorinated intermediates raises serious concerns [101,107]. Another major challenge in evaluating cold plasma treatment of PFAS lies in the accurate quantification of partially defluorinated intermediates. As already mentioned above, these intermediates can include shorter-chain PFAS, perfluoroalkyl carboxylic acids, and other fluorinated compounds that retain C–F bonds, exhibiting enhanced mobility, persistence, and potential toxicity compared to their parent compounds [84,87,94,106–109]. Therefore, alongside measuring PFAS degradation and defluorination efficiency, rigorous profiling of intermediate species and toxicity assessments are critical to accurately evaluating the safety and effectiveness of plasma treatment strategies.

5. Lessons learned, perspectives & challenges, and future outlook for plasma-based PFAS remediation

5.1. Comparative evaluation of PFAS remediation technologies: strengths and limitations

As already mentioned, a range of physical, chemical, and biological technologies has been explored for PFAS removal or destruction, each with distinct strengths and limitations.

Adsorption and membrane-based processes are widely employed for PFAS removal. These technologies can achieve over 95 % removal efficiencies for long-chain PFAS under environmentally relevant conditions [117]. Their performance is governed by PFAS physicochemical properties, membrane characteristics, and operating parameters. However, these methods are non-destructive and merely transfer PFAS into secondary waste streams, creating a need for further treatment or disposal.

AOPs, such as photocatalytic ozonation and UV/persulfate systems, provide destructive pathways for PFAS [118–121]. Although they can achieve high degradation efficiencies for long-chain PFAS, defluorination rates remain low [122]. In addition, their typically high energy demands pose significant challenges for large-scale application.

Electrochemical oxidation has shown promise in both removal and mineralization, with reported efficiencies exceeding in some cases 90 % for TOC removal [31,110,123–125]. Yet, its applicability is constrained by the cost and durability of electrode materials and energy requirements, which can range from 1.4 kWh/m³ to over 1470 kWh/m³ depending on PFAS type and concentration [31,126,127].

Bioremediation, though environmentally friendly and cost-effective, remains largely ineffective for PFAS. Studies have shown minimal degradation over multi-year timescales when PFAS-containing biosolids were applied to soil [128–132].

In summary, while several technologies have demonstrated effectiveness for PFAS mitigation, none yet combine complete degradation, low energy consumption, and scalable operation, underscoring the need for transformative alternatives.

5.2. Cold plasma: advancing energy-efficient and sustainable PFAS treatment

Multiple studies have demonstrated plasma's remarkable potential in achieving high degradation efficiencies while maintaining significantly lower energy inputs than traditional technologies. To further contextualize energy efficiency, metrics such as the electrical energy per order (E_{EO} , kWh/m³) and mass-normalized energy consumption (mg PFAS degraded/kWh) can provide standardized comparisons across systems. From an economic perspective, cold plasma exhibits significant advantages over conventional PFAS treatment technologies. Cold plasma systems typically require E_{EO} of 1–220 kWh/m³ for PFAS degradation [57,77,87,97], substantially lower than photocatalysis (>100 kWh/m³ and sometimes up to 6000 kWh/m³) [133] or electrochemical oxidation (1.4–1470 kWh/m³) [31]. At typical electricity prices (~0.10–0.15 €/kWh), these energy requirements translate into operational costs of ~0.15–30 €/m³ for plasma treatment, markedly lower than AOPs or other methods. In addition, other methods not only require high energy inputs but also involve chemical agents, adsorbents or catalysts, leading to additional costs associated with reagents, secondary waste management, and post-treatment. For example, PFOA degradation applying atmospheric radial plasma in tap water was >99 % with an E_{EO} of 1.02–13.8 kWh/m³ depending on the conditions [87]. In addition, a DBD reactor with ozone recirculation achieved 94.8 % degradation with an energy yield up to 120 mg/kWh in the presence of peroxymonosulfate (PMS) [95]. Notably, a streamer discharge system treating landfill leachate containing diverse PFAS achieved >90 % reduction of long-chain compounds and precursors, with an average energy cost of just 16 kWh/m³ [57]. In this context there is an elevating

Table 1
Indicative studies of PFAS destruction by means of cold plasma treatment.

Discharge features	PFAS/initial concentration (mg/L)	Water matrix	Treated volume (mL)	Gas type	Treatment time (min)	Degradation efficiency (%)	Energy consumption (kWh/m ³)/ energy yield (mg/kWh)	Ref
Coaxial DBD falling-film reactor-pulsed-AC	PFOA/0.08	Ultrapure water	5000	O ₂ and N ₂ mixture (60/40)	120	63.75	–/15.31	[134]
Coaxial DBD + O ₃ + PMS	PFOA/5	Ultrapure water	150	O ₃	120	94.8	–/120	[95]
Pulsed discharge + bubbles	PFOS/80	Ultrapure water	25,000	Ar	120	99.5	1.6 to 8.4 depending on the bubble size/–	[77]
Self-pulsing discharge (SPD) plasma reactor	PFOA/41.4	Ultrapure water	15	Air	30	84	–/87.4 (G50)	[135]
DC plasma within O ₂ bubbles	PFOA/41.4	Ultrapure water	20	O ₂	180	98	–/6, 2.2 (defluorination)	[115]
Gas-liquid DBD-Nanopulses	PFOS/60	Ultrapure water	15	Ar	60	100	>99.9	[79]
Falling film DBD	PFOA/1	Tap water	15	Ar	60	>99.9	23.6/19.0	[79]
7-wires Corona-DC plasma reactor operated in a negative corona discharge	PFOA/8.2	Ultrapure water	500	Air	100	98	90.6/–	[81]
7-wires Corona-DC plasma reactor operated in a negative corona discharge	PFOA/41.4	Ultrapure water	170	Air	300	12	–/0.23 (G50)	[135]
Glow discharge above water	PFOA/41.4	Ultrapure water	50	O ₂	30	<30	–/–	[136]
Reverse vortex gliding arc plasma	PFOA/100	Ultrapure water	1000	Air	60	75	213.4/–	[86]
Reverse vortex gliding arc plasma	PFOS/100	Ultrapure water	1000	Air	60	93.1	23.2/–	[86]
Radial plasma discharge over liquid surface + bubbling	PFOA/4.14	Tap water	100	Ar	15	>99	1.02/2070 (G50)	[87]
Nanopulses-dual DBD-jet	PFOA in PFAS mixture (ng/L)	Groundwater	500	Argon	60	92.3	45/–	[83]

number of studies related to the plasma treatment of PFAS which highlight the overall significant performance of the method (Table 1). In addition to energy and operational cost savings, plasma offers broad-spectrum PFAS degradation, and the potential for modular, scalable deployment. This combination of high treatment efficiency, low energy consumption, and minimal secondary waste underscores the economic viability of plasma as a sustainable and versatile solution for PFAS remediation. When evaluated alongside conventional methods, cold plasma emerges as a transformative technology capable of reducing both per-unit treatment costs and overall lifecycle expenditures.

5.3. Key determinants of efficiency: Insights into plasma reactivity and process optimization

The efficiency of PFAS degradation by plasma depends on several tightly coupled factors: plasma type, reactor configuration, plasma gas composition, water matrix characteristics, and power input parameters, all of which were thoroughly discussed in Sections 3.1–3.5.

Plasma operating parameters cannot be considered in isolation from reactor design, as trade-offs often emerge. PFAS studies highlight that each plasma type/configuration presents distinct operational limitations beyond degradation efficiency. For example, DBD reactors have the ability to maintain uniform plasma fields over extended surface areas [134]. The surfactant-like behavior of PFAS, which preferentially localizes them at the gas-liquid interface, makes gas-over-liquid systems (e.g., air-liquid DBD) particularly effective [79]. However, despite the fact that lab-scale gas-liquid DBD reactors facilitate uniform treatment at the plasma-liquid interface, suffer from limited penetration of short-lived species, which constrains energy efficiency despite high degradation yields. On the other hand, bubbling reactors coupled with above-liquid plasma have shown promoting effect in degradation performance [77] while bubbling mode that allows plasma generation within gas bubbles injected into the liquid, has emerged as a particularly promising strategy with interest to be thoroughly examined [39,70–72]. Pilot-scale plasma bubble systems energized by HV nanosecond pulses

excel in mass transfer efficiency, producing higher concentrations of short-lived species and achieving remarkably low energy consumption ($E_{EO} < 1 \text{ kWh/m}^3$) for different classes of pollutants (e.g., dyes and pharmaceuticals) whereas gas-liquid discharges favor more uniform but less efficient contact [67]. Looking ahead, one promising pilot-scale concept would be a hybrid reactor capable of delivering plasma discharges both above the liquid surface (targeting interfacial PFAS) and within the liquid phase via injected plasma bubbles (enhancing bulk contact). Such dual-interface systems could maximize plasma-pollutant interaction and represent a major step toward full-scale solutions.

However, in all plasma/reactor types, maintenance costs are also involved. Lab- and pilot-scale corona discharges are relatively simple and scalable but often display non-uniform fields and electrode corrosion. On the other hand, in the case of DBD plasma bubbles, unlike other reactor designs that generate plasma discharges directly within the aqueous phase [137], this configuration minimizes the risk of unwanted contamination from particles or metal ions that could be released through electrode corrosion [39]. In plasma jet configurations, intense localized reactivity enables high PFAS degradation in ultrapure water; however, their narrow interaction zone, electrode sensitivity, and sharp efficiency losses in real matrices highlight operational instability and increased energy demand.

In parallel, plasma gas composition determines the dominant degradation pathway, oxidative vs. reductive. Studies show air plasma performs well due to its mixed ROS/RNS environment [81], while argon plasma excels by generating high densities of hydrated electrons, crucial for efficient C–F bond cleavage [79,88]. Therefore, the maximization of the interaction of PFAS with the key-role reactive species is a challenging yet decisive factor for the efficiency of the process. In parallel, the limited degradation of short-chain PFAS needs to be further investigated and optimized in order to be able to obtain a permanent solution of the PFAS pollution problem.

Water matrix effects are equally critical, as the distribution and reactivity of plasma-generated species vary significantly with the surrounding environment. Factors such as organic matter, ionic strength,

and pH strongly influence species composition and diffusion [79], ultimately impacting PFAS degradation efficiency. These dependencies highlight the need for systematic studies to better understand and optimize plasma performance in different matrices. Tap water, with its moderate ionic strength and slightly alkaline pH, was shown to promote degradation by stabilizing hydrated electrons and enhancing interfacial transport, even outperforming ultrapure water under certain conditions (see Section 3.3) [79,94]. Moreover, the presence of bicarbonates and sulfates can promote the formation of secondary radicals (e.g., $\bullet\text{CO}_3^-$, $\bullet\text{SO}_4^-$), improving reactivity in some systems [95].

Power supply characteristics, such as voltage, frequency, and pulse duration, directly shape plasma energy density and species formation. Tuning these parameters alters the balance and distribution of reactive species, thereby impacting PFAS degradation efficiency and energy use. Increasing voltage generally enhances degradation, though excessively high values can reduce energy efficiency [138]. Similarly, HV microsecond pulses promote greater reactive species generation and faster PFAS degradation, while nanosecond pulses offer higher energy efficiency by minimizing thermal losses [79].

5.4. Scaling-up cold plasma for PFAS destruction: Opportunities and barriers for large-scale deployment

Although efforts to scale up plasma systems for water treatment remain limited, both laboratory- and pilot-scale studies demonstrate a promising trajectory toward real-world application. While most research is still conducted at the bench scale, recent pilot-scale demonstrations show that with careful reactor design and operational control, plasma technologies can sustain or even enhance treatment efficiency at larger volumes. A representative example is the nanopulsed plasma bubble reactor [67], where scaling from 70 mL to 2.5 L not only preserved degradation performance across diverse pollutant classes (e.g., dyes, pharmaceuticals) but also improved energy efficiency. For methylene blue, the electrical energy per order decreased from 0.37 to 0.18 kWh/m³, for sulfamethoxazole from 0.50 to 0.42 kWh/m³, and for valsartan from 1.54 to 0.88 kWh/m³, illustrating how reactor hydrodynamics at larger volumes can synergistically improve plasma efficiency.

A PFAS targeted pilot-scale study comes from Clarkson University, where a plasma reactor was deployed at a fire training site to treat PFAS-contaminated groundwater. Operating two 4-L plasma units in semi-batch mode at flow rates up to 8.4 L/min, the system achieved over 90 % degradation of long-chain PFAS and precursors (e.g., PFOA, PFOS) in a single cycle, reaching concentrations below the USEPA's 70 ng/L health advisory [57]. The energy efficiency of the process was satisfactory with E_{EO} ranging from 9.2 to 31 kWh/m³. However, short-chain PFAS exhibited lower degradation efficiency due to both their formation as degradation byproducts and poor transport to the plasma interface [57]. To address the more recalcitrant short-chain PFAS, in that study additional batch-mode lab-scale experiments with the cationic surfactant CTAB achieved up to 88 % degradation by enhancing interfacial transport.

Pilot-scale plasma systems introducing bubbles were also used to enhance mass transfer and hydrated electron availability, achieving markedly lower E_{EO} values (e.g., 1.6 kWh/m³ with fine bubbles) [77]. Another recent pilot-scale comprising a coaxial DBD falling-film non-thermal plasma reactor (5 L) was recently used to treat both PFOA standards and PFAS-contaminated groundwater. Under optimized conditions, the system achieved up to 63.75 % PFOA reduction with an energy efficiency of 24.63 mg/kWh. However, when applied directly to contaminated groundwater, results were variable, with some samples showing concentration decreases and others increases, likely due to precursor release and by-product formation [134].

Therefore, across multiple laboratory and pilot-scale studies [57,67,87,95], plasma systems have consistently demonstrated energy-efficient PFAS degradation and scalable performance. These outcomes

highlight the technical feasibility and emerging economic viability of cold plasma, underscoring its potential for full-scale PFAS remediation. Yet, the scarcity of pilot systems shows that scaling up remains challenging. Given the stability and surfactant-like nature of PFAS, future reactor designs must balance bulk water treatment with effective exploitation of interfacial PFAS localization.

Long-term stability under complex wastewater conditions still requires validation to ensure reliable operation and manageable maintenance. Electrode durability (e.g., corrosion and erosion), maintenance frequency, and operational stability are critical factors to weigh alongside degradation performance when selecting plasma configurations. Throughput is another major bottleneck: municipal systems process >3700 L/h, demanding plasma designs that maintain high mass-transfer rates without sacrificing energy efficiency. Hydrodynamic solutions such as vortex flows and bubble-diffuser systems, studied mainly at laboratory and pilot scale, show promise for meeting these demands [77,139].

Overall, scaling cold plasma systems for PFAS remediation remains an engineering challenge. Key priorities include sustaining effective mass transfer, ensuring uniform plasma-liquid interactions, and optimizing energy use at larger volumes, while aligning these advances with regulatory frameworks to enable commercial adoption.

5.5. Strategic research directions and innovation needs for next-generation plasma systems

To fully realize the potential of cold plasma in PFAS remediation, several strategic research priorities must be addressed. First, hybrid plasma configurations that combine gas-liquid interface and bubble-phase discharges should be developed and systematically evaluated for their ability to maximize plasma-PFAS interactions.

Plasma-catalysis is also a strategy that has been promising for other pollutants and may also offer an additional boost in TOC removal [140–143], but catalysts must be immobilized to avoid secondary pollution, and their viability should be confirmed through life-cycle and techno-economic evaluations. The development of hybrid systems, such as plasma-ozonation, plasma-adsorption, plasma-electrochemistry, or the combination of plasma with homogeneous catalysts or inorganic salts (including Fenton-like synergy or/and persulfate/percarbonate contribution) may offer further potential to expand treatment versatility, lower costs, and accelerate adoption. Moreover, integrating plasma with pre- or post-treatment processes could further enhance PFAS removal and achieve near-complete mineralization. For instance, pre-treatment steps such as adsorption or membrane separation, would allow plasma reactors to operate more efficiently, while post-treatment oxidation or catalytic enhancement could target residual intermediates to maximize defluorination and ensure full mineralization.

As mentioned in the previous section, one of the main challenges in PFAS remediation is scaling treatment to large water volumes. A promising strategy is to first concentrate PFAS using Surface Active Foam Fractionation (SAFF) and then apply plasma treatment to the smaller enriched waste stream. This combined approach creates a closed loop system that separates, concentrates and ultimately destroys PFAS, offering a more energy efficient and sustainable pathway toward full scale remediation. Systematic studies exploring such hybrid treatment trains are a promising avenue for future research.

Advanced analytical diagnostics, including UPLC-MS, TOC balance, and fluorine mass balance, should be standardized to thoroughly assess degradation pathways, the formation and fate of intermediates, and the completeness of mineralization, as discussed in Section 4. Additionally, experiments using real wastewater matrices are urgently needed to evaluate the impact of matrix complexity, fouling behavior, and potential by-product toxicity under operationally relevant conditions.

Nevertheless, monitoring of plasma-generated by-products remains essential to ensure environmental safety. Although studies on PFAS treatment have not reported significant formation of NO_x or ozone under

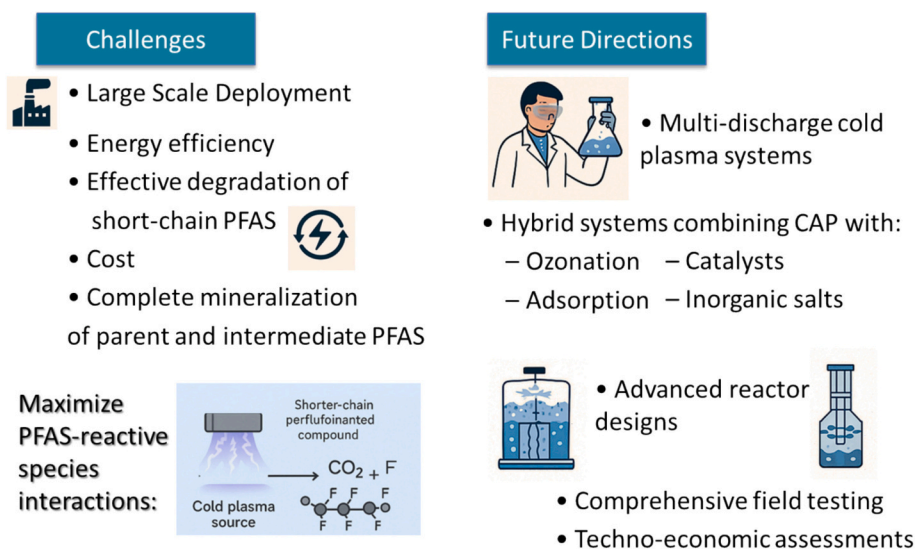


Fig. 9. Challenges and future directions for the degradation of PFAS by cold plasma.

typical operating conditions, such evaluations become particularly important when cold plasma is applied to complex wastewater matrices. A prudent contingency measure could involve capturing and recirculating ozone from exhaust gases, allowing its reuse to support the treatment process through complementary ozonation.

Furthermore, complementary techno-economic analyses and life-cycle assessments (LCA) are necessary to quantify sustainability metrics and benchmark plasma against existing treatment technologies. Moving forward, plasma systems should be benchmarked against conventional treatments such as ozonation, UV, and AOPs under real-world conditions to establish their comparative performance. Optimization of operational parameters, coupled with thorough scalability and economic feasibility studies, will be essential for successful technology transfer.

Effective stakeholder engagement including regulatory bodies, water utilities, and technology developers will be critical to define treatment objectives and develop performance standards for plasma-based water purification systems. Finally, regulatory acceptance is a key consideration for large-scale deployment. Cold plasma is not yet specifically referenced in existing PFAS treatment guidelines; however, its demonstrated ability to achieve high PFAS degradation and, in some cases, mineralization positions it as a complementary or alternative technology alongside conventional approaches. Integrating cold plasma within existing regulatory frameworks, such as using it after approved removal processes or as part of hybrid treatment trains, can facilitate early adoption while ensuring compliance with established water quality standards.

In conclusion, future research directions (Fig. 9) can be prioritized based on technical feasibility, scalability, and environmental impact. High-priority areas include developing hybrid plasma configurations that maximize the contact between PFAS and plasma reactive species, integrating plasma with complementary pre- or post-treatment processes to ensure complete PFAS mineralization, and optimizing reactor designs for high throughput and long-term operational stability. Next in turn, studies should focus on real wastewater testing, effective defluorination assessments, and detailed monitoring of plasma-generated species. Subsequent efforts include refining regulatory frameworks and performing comprehensive lifecycle and techno-economic analyses to support sustainable, large-scale implementation.

6. Conclusions

Cold plasma is undeniably a highly promising approach for PFAS

destruction since it can produce both reductive and oxidative reactive species, most importantly hydrated electrons (e_{aq}^-), which enable direct C–F scission, while operating at ambient conditions with competitive energy footprints. However, it needs to be considered that PFAS present unique, system-specific challenges (strong C–F bonds, surfactant behavior and matrix sensitivity) that make reactor design, plasma gas selection, and control of the plasma-liquid interface decisive for success. Systems that maximize plasma-liquid contact and driven by pulsed or tailored HV waveforms show the best trade-off between degradation rate and energy efficiency. Pilot demonstrations already report high degradation of long-chain PFAS at considerable energy efficiency, yet short-chain PFAS and full mineralization (avoidance/monitoring of persistent byproducts such as TFA) remain key gaps. Therefore, the near-term pathway is clear: prioritize hybrid reactor concepts that combine interfacial and bulk plasma delivery, standardize diagnostics, perform real-matrix and long-duration trials, and pair techno-economic and life-cycle analyses with early regulatory engagement to translate cold plasma from promising lab/pilot results into reliable field solutions. By overcoming these challenges, cold plasma has the potential to emerge as a transformative technology in the global strategy for sustainable PFAS remediation, offering an innovative solution to the unique chemical and engineering complexities of this contaminant class.

CRedit authorship contribution statement

Christos A. Aggelopoulos: Writing – review & editing, Writing – original draft, Visualization, Resources, Investigation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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