

Review

Plasma-Activated Homogeneous Catalysis for Water Decontamination: Mechanisms, Synergies, and Future Perspectives

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Abstract

The pervasive contamination of water bodies by refractory organic pollutants necessitates the development of advanced purification technologies. Plasma has emerged as a promising solution, capable of generating a broad spectrum of reactive oxygen and nitrogen species (RONS), UV photons, and electrons in situ, thereby directly degrading contaminants. However, the practical application of plasma-alone systems is often constrained by limited energy efficiency and insufficient mineralization capacity. To overcome these challenges, the integration of plasma with homogeneous advanced oxidation processes (AOPs) has been established as a highly effective strategy. By coupling plasma with catalysts such as peroxymonosulfate (PMS), peracetic acid (PAA), periodate (PI), and Fenton reagents ($\text{Fe}^{2+}/\text{Fe}^{3+}$), a remarkable synergistic effect is achieved. This synergy arises from the multi-modal activation of catalysts by plasma via energetic electrons, UV photolysis, and radical-induced reactions, while the catalysts, in turn, consume long-lived plasma products and regulate reaction pathways. The resultant ‘plasma/catalytic’ system significantly enhances the degradation rate and mineralization efficiency of pollutants, broadens the operational pH window, and improves overall energy utilization. This review systematically examines the mechanisms, performance, and influencing factors of these hybrid systems, and discusses current challenges and future prospects to guide the development of this synergistic technology for sustainable water remediation.

Keywords: plasma; homogeneous catalysis; synergy effect; reactive oxygen species; pollutants



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1. Introduction

The preservation of water resources is fundamental to ecosystem integrity and human society. However, the escalating contamination of aquatic environments by a wide array of recalcitrant organic compounds has emerged as a critical global challenge [1–3]. These synthetic pollutants often possess complex molecular structures that render them resistant to conventional biological degradation processes, allowing them to persist in water bodies and pose long-term risks to environmental safety and public health [4]. This pressing scenario has intensified the demand for innovative and highly efficient water treatment technologies capable of achieving complete and rapid pollutant elimination [5].

In this context, plasma technology has surfaced as a formidable and versatile advanced oxidation platform [6]. As a partially ionized gas, plasma serves as a rich source of diverse reactive species, including high-energy electrons, ions, excited atoms/molecules,

radicals (e.g., $\cdot\text{OH}$, $\cdot\text{H}$), and molecular agents (e.g., H_2O_2 , O_3) [7–9]. Upon interaction with aqueous solutions, these plasma-generated species initiate a complex suite of physiochemical processes directly in the liquid phase or at the gas–liquid interface, enabling the direct degradation of a wide spectrum of organic pollutants [10]. Unlike conventional single-mode activation methods, plasma provides a unique multi-component ‘reactive species library’ and an energetic field, establishing a versatile foundation for water decontamination [11]. Despite these advantages, the practical application of plasma-alone systems is hampered by inherent limitations, such as relatively low energy efficiency, where a significant portion of electrical energy is dissipated as heat, and poor reaction selectivity, which can lead to incomplete mineralization and the formation of toxic by-products [12]. These bottlenecks underscore the need to enhance the efficacy and sustainability of plasma-based water treatment.

Homogeneous catalysts play a pivotal role in advanced oxidation processes (AOPs) due to their high reactivity and efficiency [13]. Their effectiveness stems from the ability to generate powerful radical species (e.g., $\cdot\text{SO}_4^-$, hydroxyl radicals $\cdot\text{OH}$, and organic oxygen-centered radicals) upon activation, which can aggressively attack and break down pollutant molecules [14–16]. However, the conventional methods for activating these catalysts, including thermal energy, ultraviolet radiation, and transition metal ions, often entail significant drawbacks, such as high energy input, limited pH operating windows, and the risk of secondary pollution from metal leaching or sludge formation [17,18]. These inherent limitations have motivated the exploration of more efficient and sustainable activation strategies.

Despite the efficacy of homogeneous AOPs, their conventional activation methods (e.g., thermal energy, UV radiation, transition metal ions) present inherent drawbacks that hinder their broad application. These include high energy consumption, a narrow effective pH range (particularly for Fenton reactions), the risk of secondary pollution from metal leaching or chemical residues, and the limited utilization efficiency of the oxidants. These challenges collectively create a demand for an alternative activation platform that is more energy-efficient, environmentally benign, and operationally flexible.

The convergence of these two fields, plasma chemistry and homogeneous catalysis, has given rise to the innovative strategy of ‘plasma/catalytic synergy’ [19]. Recently, the strategy of integrating plasma with homogeneous catalysts has emerged as a frontier in environmental catalysis for the synergistic degradation of pollutants [20]. A growing body of evidence delineates a complex synergy where the plasma environment does not merely co-exist with the catalyst but actively and reciprocally interacts with it. Plasma can directly activate catalysts via multiple parallel pathways (e.g., electronic, photolytic, and radical-induced), while the presence of certain catalysts can, in turn, modulate plasma chemistry and consume long-lived plasma products (e.g., H_2O_2), driving the entire system towards greater efficiency and often different reaction pathways compared to either process alone [21]. This interplay results in a synergistic effect where the combined system’s performance significantly surpasses the sum of its parts. For instance, high-energy electrons and UV photons from plasma can directly cleave the O–O bond in persulfates to generate $\cdot\text{SO}_4^-$; meanwhile, plasma-derived $\cdot\text{OH}$ and $\cdot\text{H}$ atoms can drive the metal redox cycle in Fenton-like systems (e.g., enhancing the reduction of Fe^{3+} to Fe^{2+}), thereby significantly improving efficiency and broadening the effective pH range [22,23].

Therefore, this review aims to provide a comprehensive and critical analysis of the recent advances in the application of plasma-activated homogeneous catalyst systems for the degradation of organic pollutants. It will commence with an overview of the fundamental principles and synergistic advantages of this coupled technology. Subsequently, it will delve into a detailed and systematic discussion of the activation mechanisms, degradation

performance, and influencing factors for various homogeneous catalysts, including PMS, PAA, periodate (PI), H_2O_2 and $\text{Fe}^{2+}/\text{Fe}^{3+}$. Finally, the review will critically assess the current challenges hindering practical application and offer insightful perspectives on future research directions, encompassing the development of novel catalysts, reactor optimization, enhancement of energy efficiency, and compatibility with real-world water matrices. The overarching goal is to provide a valuable reference and stimulate further innovation in the development of this promising green technology for water purification.

2. Fundamentals of Plasma Technology

Plasma, often referred to as the fourth state of matter, is a partially ionized gas composed of electrons, ions, neutral atoms, and various reactive species [24,25]. It was first systematically described by Tonks and Langmuir in 1929 as an “ionized gas” with collective behavior governed by electromagnetic forces. Plasma can be broadly classified into thermal (or equilibrium) plasma and non-thermal plasma (NTP) [26]. Thermal plasma, characterized by local thermodynamic equilibrium among all species, is typically generated at high temperatures ($>10,000$ K) and is used in industrial applications such as waste treatment and material synthesis [27]. In contrast, NTP, also known as cold plasma, maintains a high electron temperature (1–10 eV) while the bulk gas remains near ambient temperature, making it particularly suitable for environmental and biomedical applications where thermal damage must be avoided [28].

Plasma can be generated through various electrical discharge methods, each with distinct mechanisms and reactor configurations. Common types include dielectric barrier discharge (DBD), corona discharge, glow discharge, pulsed discharge, and gliding arc discharge [29,30]. DBD reactors incorporate one or more dielectric barriers between electrodes to prevent arc formation, enabling uniform and stable discharge at atmospheric pressure [31]. Corona discharge, typically generated in a point-to-plane or wire-cylinder geometry, produces highly localized electric fields and is effective for gas-phase pollutant removal [32]. Glow discharge operates at low pressure and is characterized by a luminous plasma region, often used in surface modification and thin-film deposition [33]. Pulsed discharge systems employ short-duration high-voltage pulses to generate high-energy electrons with minimal heating, enhancing energy efficiency [34]. Gliding arc discharge, a hybrid between thermal and non-thermal plasma, utilizes a diverging electrode configuration where the arc is extended and cooled by gas flow, enabling efficient degradation of volatile organic compounds [35].

Figure 1 shows the number of published studies on plasma degradation of pollutants in water and the proportion of articles with plasma types activating different homogeneous catalysts on the Web of Science in the past ten years. The effectiveness of plasma in wastewater treatment stems from its ability to generate a multitude of reactive oxygen and nitrogen species (RONS), including $\cdot\text{OH}$, O_3 , H_2O_2 , atomic oxygen (O), and superoxide anions ($\cdot\text{O}_2^-$) [36–38]. These species are produced through collisions between high-energy electrons and water or gas molecules, leading to the dissociation, excitation, and ionization of reactants. For instance, electrons with energies between 1 and 10 eV can dissociate water molecules into $\cdot\text{OH}$ and $\cdot\text{H}$, while O_2 can be converted into O_3 or atomic oxygen. Additionally, plasma emits ultraviolet (UV) radiation, produces shockwaves, and induces local electric fields, all of which contribute to the degradation of organic pollutants through physical and chemical pathways [39,40]. It is noteworthy that O_3 , a common by-product of plasma discharge in oxygen-containing atmospheres, is managed with strict safety precautions. In experimental systems, the reactor is typically operated within a sealed or ventilated enclosure (e.g., a fume hood), and the effluent gas is often treated with an ozone

destruct unit (e.g., catalytic converter) before release to ensure operator safety and prevent environmental release.

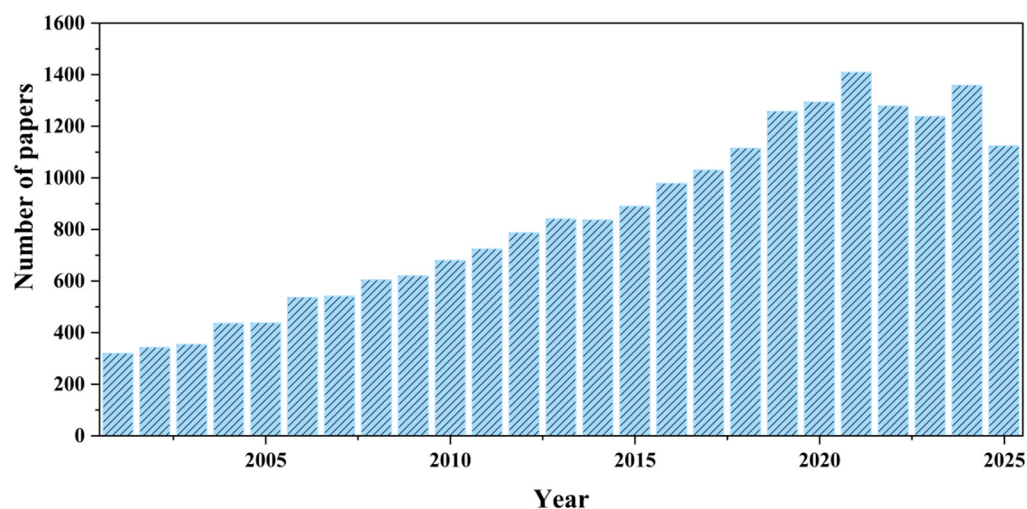


Figure 1. The number of literature sources related to wastewater treatment by plasma from 2001 to 2025 (data from ‘Web of Science’).

Different plasma reactors exhibit varying efficiencies and applicability depending on the target pollutants and operational conditions [41]. For example, DBD reactors are widely used for liquid-phase treatment due to their ability to form stable plasma in contact with water, as seen in falling-film, coaxial, and plate-type configurations [42]. Corona discharge, while energy-efficient for gas-phase applications, shows limited liquid penetration and is often combined with aeration or spraying systems for aqueous treatment [43]. Pulsed discharge systems offer high instantaneous power and are effective in generating strong UV and shockwaves, promoting the breakdown of refractory compounds [44]. Gliding arc reactors demonstrate good scalability and energy efficiency for continuous-flow systems, particularly in treating high-concentration organic wastes [45]. Table 1 summarizes the key characteristics and typical applications of these plasma types. See Figure 2 for some common plasma reactors. All reactors contain high-voltage electrodes and grounded electrodes that produce plasma by supplying enough energy to the gas to ionize it.

Despite these advantages, plasma technology still faces several challenges that limit its large-scale application [46–48]. The energy efficiency of plasma processes remains relatively low, with a significant portion of electrical energy dissipated as heat or lost in non-reactive pathways [49]. Moreover, the selectivity of plasma-induced reactions is often poor, leading to incomplete mineralization and the formation of toxic by-products [50]. The stability and longevity of plasma reactors under continuous operation, especially in complex wastewater matrices, also require further improvement [51,52]. To address these issues, recent research has focused on integrating plasma with homogeneous or heterogeneous catalysts, which can enhance the formation of reactive species, improve energy utilization, and promote selective degradation pathways [53–55]. The plasma treatment of water pollution has always been a hot issue, and specific solutions are emerging one after another. The following sections will elaborate on the synergistic effects of plasma/catalytic systems, with an emphasis on homogeneous catalytic mechanisms.

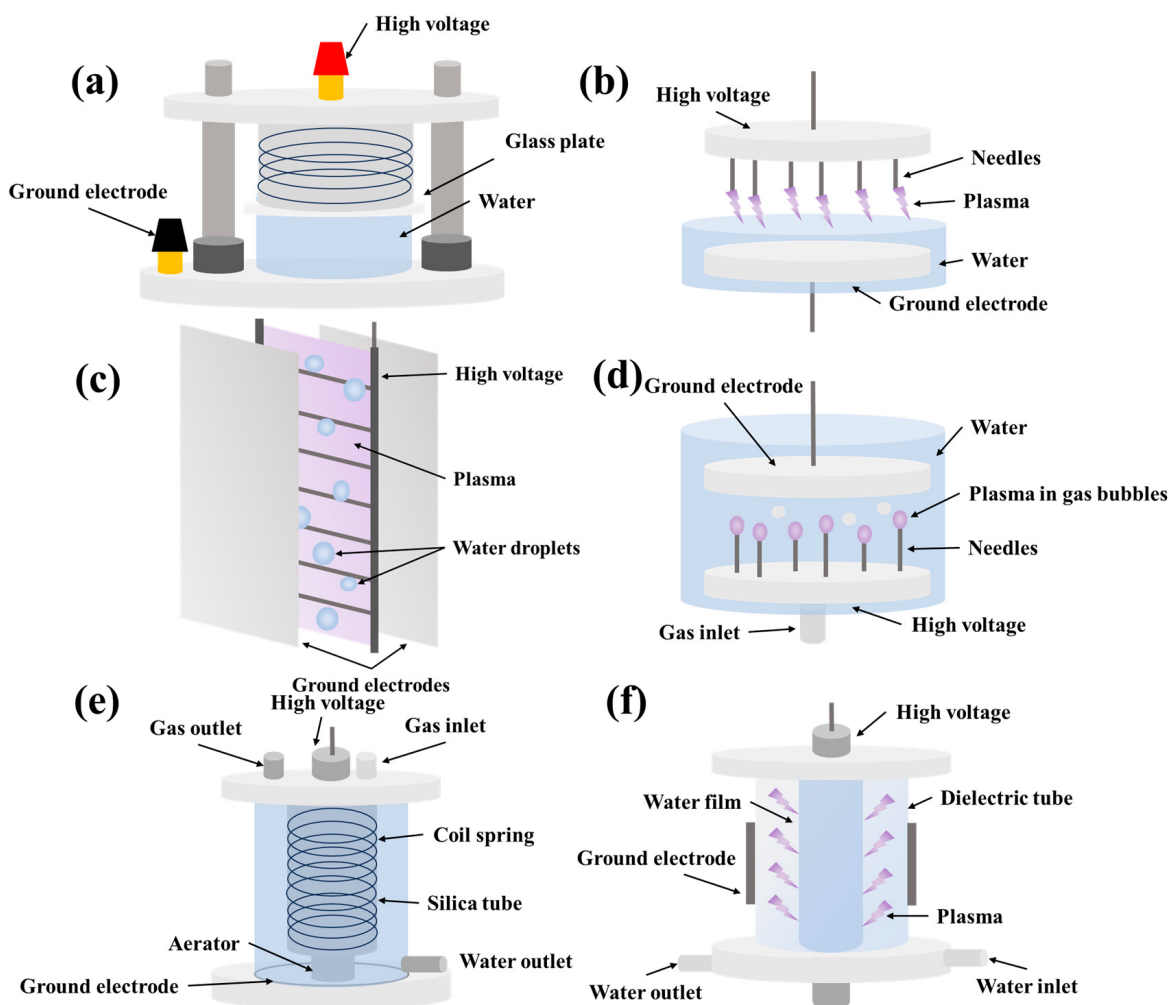


Figure 2. Common plasma reactor structures: (a) plate–plate reactor; (b) needle array–plate reactor; (c) corona with water shower reactor; (d) corona in bubbles reactor; (e) tube–tube reactor; (f) falling liquid film reactor.

Table 1. Different discharge types of plasma.

Character	DBD	Corona	Pulsed Corona	Gliding Arc	Glow Discharge
Structure	Plate-to-plate, coaxial	Needle-plate, thread-cylinder	Thread-cylinder, needle-plate	Knife-shaped electrode	Parallel plate electrode
Modality	Filiform/Uniform	Streamer/Corona layer	Streamer/Corona layer	Arc of gliding arc	Be diffused and uniform
Energetic efficiency	Medium	Medium	High	Medium	Medium
Power density	Medium	Low	High	High	Low
Active substances	$\cdot\text{OH}$, O_3 , UV	$\cdot\text{OH}$, O_3	$\cdot\text{OH}$, UV, Shock wave	$\cdot\text{OH}$, O_3 , NO_x	$\cdot\text{OH}$, e^-
Interact with the liquid	Direct contact	Gas phase mass transfer	Gas-liquid interface	Gas-liquid interface	Spraying or sputtering
Gas temperature (K)	300–1000	300–500	300–500	~1 (arc core), ~10 (wake flame)	1–10
Power source	AC HV	AC/DC HV	Nanosecond pulse HV	AC/DC HV	AC/DC LF
System complexity	Medium	Low	High	Medium	High
Major areas	VOCs degradation, water treatment	Flue gas purification, ozone generation	Refractory wastewater treatment	Surface modification of materials	Precision cleaning, material composition
Advantage	The structure is simple and stable	Simple equipment, low energy consumption	High energy efficiency with fewer by-products	Uniform discharge and good controllability	Plasma density is high, no electrode pollution
Disadvantage	The electrode may corrode	Uneven discharge	Expensive price	System requirements are high	The system is complex and the investment is large
Ref.	[56]	[57]	[58]	[59]	[60]

3. Plasma-Activated Homogeneous Catalysts

In order to better analyze the combination of plasma and different homogeneous catalysts, five homogeneous catalysts (PMS, PAA, PI, H₂O₂, Fe²⁺/Fe³⁺) were taken as representatives for analysis. By analyzing the visualization diagram of the key words (Figure 3), it can be known that the relatively large node sizes for terms such as ‘hydrogen peroxide’ and ‘iron’, aside from the core keywords ‘degradation’ and ‘plasma’, highlight them as prominent research foci within the field of pollutant degradation using plasma coupled with homogeneous catalysts.

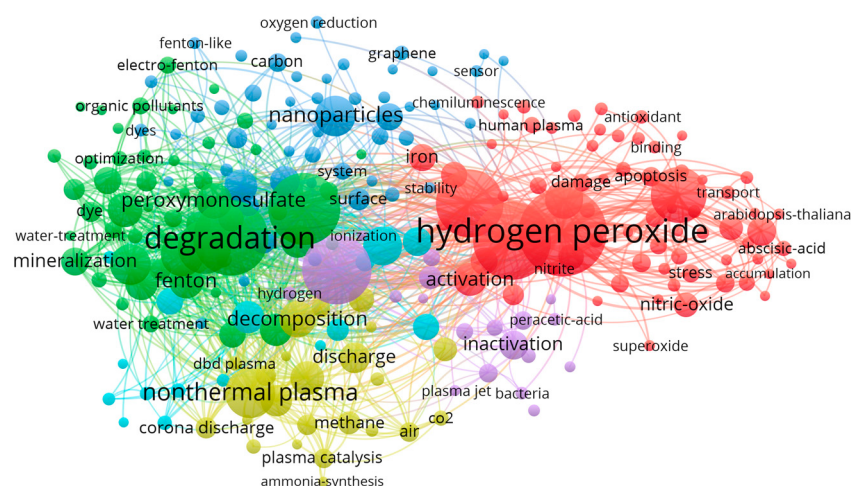
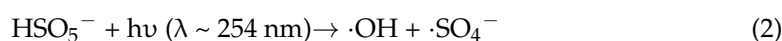
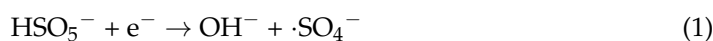


Figure 3. Average year publication volume and keyword frequency of plasma in collaboration with homogeneous catalysts degrades pollutants from 2015 to 2025 (data from ‘Web of Science’).

3.1. Peroxymonosulfate (PMS)

PMS is a versatile oxidant and a primary precursor to $\cdot\text{SO}_4^-$, which has garnered significant attention in advanced oxidation processes. With a standard redox potential of 1.82 V, PMS itself possesses moderate oxidizing capability [61]. However, its true potential is unlocked upon activation to generate $\cdot\text{SO}_4^-$, a radical species with a higher redox potential ($E^0 = 2.5\text{--}3.1$ V), longer persistence, and greater selectivity for electron-rich organic pollutants compared to the hydroxyl radical. Despite its promise, a critical limitation of PMS-based AOPs lies in the requirement for effective activation. Traditional methods, including transition metals, heat, and UV light, often face challenges such as metal leaching and sludge formation, limited energy efficiency, or high operational costs.

Plasma technology presents an ideal solution to these limitations by serving as a multi-modal activation platform. A plasma discharge system simultaneously produces a suite of physical and chemical effects, including high-energy electrons, UV photons, ozone, and other reactive species, that can synergistically activate PMS. The primary activation mechanisms in a plasma/PMS system involve: the direct cleavage of the O-O bond in PMS by high-energy electrons and UV photolysis of PMS (e.g., Equations (1) and (2)) [62]. Additionally, other plasma-generated radicals like $\cdot\text{O}_2^-$ can participate in secondary activation cycles, ensuring a continuous and robust production of $\cdot\text{SO}_4^-$ and $\cdot\text{OH}$. This multi-pathway activation not only enhances the utilization efficiency of PMS but also creates a diverse mix of powerful oxidants, leading to a remarkable synergistic effect that significantly accelerates the degradation of contaminants.



$\cdot\text{SO}_4^-$ mainly attacks pollutant molecules through the electron transfer mechanism. It has a remarkable effect on the cleavage of carbon–halogen bonds (C-X), and can efficiently dehalogenase halogenate organic compounds, thereby reducing their toxicity and persistence. As an electrophilic reagent, it preferentially attacks sites with high electron cloud density, such as phenolic hydroxyl groups, amino groups and other electron-rich functional groups, and initiates the ring-opening reaction of aromatic rings. $^1\text{O}_2$ in the system can also selectively oxidize electron-rich functional groups and is less disturbed by background water quality.

Substantial research efforts have validated the superior performance of plasma/PMS systems [63–65]. Hua et al. reported a synergistic system in which the combined action of water surface plasma and Fe^{2+} /PMS activation generated a multitude of reactive oxygen species, resulting in a remarkable enhancement in the degradation rate, energy efficiency, and mineralization of Malachite Green compared to the plasma process alone [66]. In a study targeting a notoriously recalcitrant perfluoroalkyl substance, Wang et al. employed a DBD plasma/PMS system to degrade perfluorooctanoic acid (PFOA) [67]. Their findings confirmed that the plasma-activated PMS generated substantial $\cdot\text{SO}_4^-$, which were crucial for breaking the robust C-F bonds, leading to efficient PFOA removal. These studies consistently underscore that the integration of plasma with PMS not only boosts the degradation rate and mineralization efficiency but also operates effectively across a wider pH range. See Table 2 for relevant studies on plasma activation of PMS.

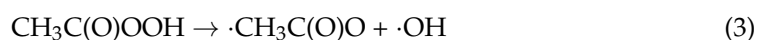
Table 2. Research on plasma activation of PMS.

Author	Target	PMS Concentration	Conditions	Time/min	Degradation Rate
Wu et al. [68]	Benzotriazole (BTA)	2.52 mM	Initial concentration: 10 mg/L Voltage: 12 kV Conductivity: 920–1000 $\mu\text{S}/\text{cm}$ pH: 6.2	20	87%
Luo et al. [69]	Potassium ethyl xanthate (PEX)	Molar ratio of PMS and PEX = 30:1	Initial concentration: 20 mg/L Voltage: 27.5 kV pH: 7	25	91%
Shang et al. [70]	Sulfamethoxazole (SMX)	0.8 mM	Initial concentration: 0.08 mM Voltage: 26 kV Conductivity: 26–512 $\mu\text{S}/\text{cm}$ pH: 10	25	Almost 100%
Deng et al. [71]	Sulfadiazine (SDZ)	0.4 mM	Initial concentration: 10 mg/L Voltage: 12 kV Frequency: 9.2 kHz pH: 7	15	92%
Sang et al. [72]	Chlortetracycline (CTC)	0.8 mM	Initial concentration: 75 mg/kg Voltage: 12 kV Frequency: 10 kHz	30	82%
Rahman et al. [73]	Complex organic compounds	10 mM	Initial concentration: 130 mg/L Voltage: 9 kV Current: 50 mA Power: 7.77 W	60	82%
Jia et al. [74]	Perfluorooctanoic acid (PFOA)	20.8 mM	Initial concentration: 75 mg/L Power: 38.52 W Energy consumption: 399.14 mg/kWh pH: 2	60	99%

3.2. Peracetic Acid (PAA)

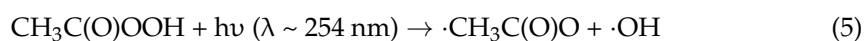
PAA represents an emerging oxidant in advanced oxidation processes, characterized by its high standard redox potential ($E^0 = 1.96 \text{ V}$) and minimal formation of hazardous disinfection by-products [75]. While demonstrating inherent oxidative capability, PAA's full potential is realized through activation to generate highly reactive radical species,

primarily acetyloxyl radicals ($\cdot\text{CH}_3\text{C}(\text{O})\text{O}$) and hydroxyl radicals ($\cdot\text{OH}$), according to the fundamental decomposition pathway (Equation (3)) [76]:



Conventional activation methods, including UV photolysis ($k = 0.02\text{--}0.05 \text{ M}^{-1}\text{s}^{-1}$), transition metal catalysis, and alkaline hydrolysis, often encounter limitations such as narrow pH optima, secondary contamination risks, and insufficient radical yield [77]. These constraints have motivated the development of alternative activation strategies.

Plasma technology establishes an advanced activation platform through its multi-mechanistic action [78]. The plasma discharge generates diverse reactive components, high-energy electrons (1–10 eV), UV photons (200–400 nm), and radical species ($\cdot\text{OH}$, $\cdot\text{O}_2^-$), that synergistically activate PAA through parallel pathways (Equations (4)–(7)) [79]:



$\cdot\text{CH}_3\text{C}(\text{O})\text{O}$ has an extremely strong destructive power on the azo bond ($-\text{N}=\text{N}-$) and other chromophoric groups in dye molecules, leading to rapid decolorization. Most free radicals can work in synergy with $\cdot\text{OH}$ to attack the aromatic ring structure, causing hydroxylation and eventually ring-opening cleavage. They can destroy the cell membrane structure of microorganisms and inactivate key enzymes, achieving efficient disinfection and sterilization.

Plasma-activated PAA technology demonstrates significant performance improvements in both pollutant degradation and microbial inactivation by enhancing the generation of active species, increasing reaction rates and energy efficiency [80–82]. In the treatment of refractory organic substances, Su et al. conducted a systematic study on the degradation of bisphenol A (BPA) [83]. The results show that the DBD/PAA system has a degradation rate of BPA as high as 93.4% within 15 min, which is 39.8% higher than that of the DBD system alone. The energy efficiency of this system reaches 100.7 mg/kWh, and its G_{50} value (211.5 g/kWh) is approximately 3.5 times that of the DBD treatment alone (G_{50} : the energy yield for 50% pollutant removal.). Through free radical quenching experiments and electron spin resonance analysis, it was confirmed that $\cdot\text{OH}$ and $\cdot\text{CH}_3\text{C}(\text{O})\text{O}$ play a leading role in the degradation process. Further, through liquid chromatography-mass spectrometry and density functional theory calculations, the degradation pathway of BPA was analyzed, and it was confirmed that the ecological toxicity of the intermediate reaction products was significantly reduced, demonstrating the environmental safety of this technology. In terms of microbial control, Yang et al. investigated the inactivation effect of DBD plasma in combination with PAA on *Chlorella* [84]. The research found that the DBD/PAA system achieved an inactivation rate of 89% for *Chlorella* within 15 min, which was approximately 21 percentage points higher than that of DBD treatment alone. Mechanism studies have shown that plasma activation of PAA significantly enhances the generation of active species within the system, with the signal intensities of $\cdot\text{OH}$ and $^1\text{O}_2$ increasing to 2.5 times and 1.3 times that of the DBD system alone, respectively. These active species eventually lead to the death of algal cells by destroying the integrity of the algal cell membrane, oxidizing key photosynthetic pigments such as chlorophyll and causing DNA damage. This process

maintains stable treatment effects in different water body matrices, demonstrating good practical application potential. See Table 3 for relevant studies on plasma activation of PAA.

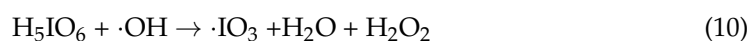
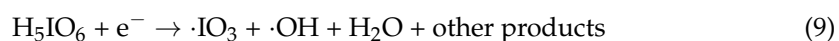
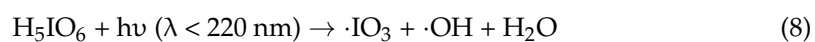
Table 3. Research on plasma activation of PAA.

Author	Target	PAA Concentration	Conditions	Time/min	Degradation Rate
Wu et al. [78]	Tetracycline (TC)	60 mg/L	Initial concentration: 20 mg/L Voltage: 21 kV Frequency: 50 Hz Conductivity (σ): 200 μ S/cm pH: 5	60	86%
Li et al. [79]	Sulfamethoxazole (SMX)	20 mg/L	Initial concentration: 20 mg/L Power: 120 W pH: 6.7	30	52%
Su et al. [81] and Han et al. [83]	Bisphenol A (BPA)	3 mM	Initial concentration: 40 mg/L Power: 445 W/450 W Frequency: 3500 Hz pH: 4.5/4.8	15	93%
Yang et al. [84]	Chlorella	0.15 g/L	Liquid flow rate: 100 mL/min Initial concentration: 0.100 (OD680) Voltage: 180 V pH: 3.5	15	95%

3.3. Periodate (PI)

PI is an emerging oxidant in advanced oxidation processes, characterized by its high oxidation potential and unique iodine-centered reactivity [85,86]. As a periodate-based oxidant, PI possesses a complex oxidation chemistry that can be directed toward efficient pollutant degradation through controlled activation. The standard activation pathways for PI, including ultraviolet irradiation, sulfite reduction, and heterogeneous catalysis, often face challenges related to slow reaction kinetics, pH dependency, and potential catalyst contamination [87]. These limitations have prompted investigation into alternative activation methods that can more effectively harness PI's oxidative potential.

Plasma technology offers a sophisticated platform for PI activation through its ability to generate multiple reactive species simultaneously [88]. The plasma discharge environment produces energetic electrons, ultraviolet radiation, and various radical species that can activate PI through complementary pathways. The primary mechanisms include reductive activation by UV photolytic decomposition (Equation (8)), plasma electrons (Equation (9)) and radical-mediated activation (Equation (10)) [89].



It is noteworthy that the UV photolysis pathway (Equation (8)), requiring photons with $\lambda < 220 \text{ nm}$, presents a challenge for conventional mercury-based UV lamps, which exhibit low emission intensity in this spectral range. This limitation, however, is mitigated in plasma-based systems. The plasma discharge itself acts as an intrinsic source of broad-band ultraviolet radiation, including vacuum ultraviolet (VUV) photons capable of cleaving the O-I bond in periodate. More importantly, the practical efficacy of the plasma/periodate system is secured by its multi-modal activation design. Even if the photolytic contribution is limited under certain conditions, the concurrent activation by energetic electrons (Equation (9)) and radicals (Equation (10)) ensures continuous and ro-

bust generation of reactive species, underscoring the synergistic advantage of the plasma process over single-mode activation methods.

The generated iodate radical ($\cdot\text{IO}_3$) exhibits strong oxidizing capability with a redox potential of approximately 1.6 V, while the simultaneous production of hydroxyl radicals and other reactive species creates a multi-oxidant system that can effectively degrade various organic contaminants. The reactions of $\cdot\text{IO}_3$ and other iodoxyl radicals are highly selective. They can preferentially attack electron-rich functional groups through hydrogen extraction or electron transfer mechanisms. This enables them to exhibit reaction rates and efficiencies far exceeding those of other systems when dealing with specific pollutants such as phenols and anilines. To clarify: In aqueous solution, periodic acid (H_5IO_6) and periodate salts dissociate to form various species, with the periodate ion (IO_4^-) being the key oxidizing agent involved in the activation processes described.

According to Kim et al., the integration of plasma with periodate markedly improved the degradation of tetramethylammonium hydroxide (TMAH) [90]. Their results showed that under optimized conditions (40.56 W CP power and 4 mM PI), the kinetic constant for TMAH removal reached 1.6250 min^{-1} , representing a 54-fold enhancement compared to the low-power CP system (2.2 W) alone. This remarkable improvement was attributed to the generation of reactive species such as $\cdot\text{OH}$, O_3 , and $^1\text{O}_2$ by plasma, which activated PI to produce more reactive radicals like $\cdot\text{IO}_3$ and $\cdot\text{O}_2^-$. In a complementary study, Jiang et al. developed plasma system coupled with Fe-N-C catalyst for PI activation to degrade sulfadiazine (SDZ) [91]. They reported that the DBD/Fe-N-C/PI system achieved complete removal of SDZ within 12 min, with a reaction rate constant of 0.53 min^{-1} , substantially higher than that of the DBD system alone. Jiang et al. identified that besides the contribution of reactive oxygen species (ROS), the electron shuttle mechanism mediated by Fe-N-C played a pivotal role. The catalyst not only reduced the activation energy barrier for PI but also facilitated efficient electron transfer between PI and pollutants, introducing a non-radical degradation pathway alongside conventional radical oxidation.

The integration of plasma with PI establishes a promising advanced oxidation platform that overcomes the limitations of conventional PI activation methods. This hybrid approach leverages the synergistic interaction between plasma-generated species and PI-derived radicals, creating an efficient and adaptable system for water treatment applications. See Table 4 for relevant studies on plasma activation of PI.

Table 4. Research on plasma activation of PI.

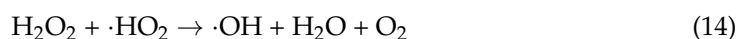
Author	Target	PI Concentration	Conditions	Time/min	Degradation Rate
Puyang et al. [89]	Sulfadiazine (SDZ)	6 mM	Initial concentration: 20 mg/L Power: 560 W pH: 3.8	12	99%
Kim et al. [90]	Tetramethylammonium hydroxide (TMAH)	4 mM	Initial concentration: 10.4 mg/L Power: 40.56 W pH: 10	60	30%
Jiang et al. [91]	Sulfadiazine (SDZ)	3 mM	Initial concentration: 40 mg/L Power: 510 W pH: 3.2	12 min	94%
Zhang et al. [92]	Atrazine (ATZ)	0.01 mM	Initial concentration: 10 mg/L Power: 68 W pH: 5	10 min	87%

3.4. H_2O_2

H_2O_2 stands as one of the most fundamental and environmentally benign oxidants in advanced oxidation processes, characterized by its high oxidation potential ($E^0 = 1.78 \text{ V}$) and the clean decomposition products of water and oxygen [93–95]. While widely utilized,

the relatively slow reaction kinetics of H_2O_2 with many recalcitrant organic pollutants at ambient conditions limits its direct application [96]. The activation of H_2O_2 to generate highly reactive hydroxyl radicals ($\cdot\text{OH}$, $E^0 = 2.8 \text{ V}$) is therefore crucial, with conventional methods including Fenton's reagent, UV photolysis, and alkali activation [97–99]. However, these approaches often contend with constraints such as a narrow optimal pH range, the cost of UV lamps and electrical energy, or the requirement for continuous chemical addition [100].

Plasma technology presents a robust and versatile alternative for H_2O_2 activation, functioning both as an in situ generator and an activator of exogenous H_2O_2 [101,102]. The plasma discharge creates a reactive milieu rich in energetic electrons, ultraviolet photons, and a plethora of radicals (e.g., $\cdot\text{OH}$, $\cdot\text{HO}_2$), which can initiate multiple decomposition pathways for H_2O_2 : electron impact dissociation (Equation (11)), UV photolysis (Equation (12)) and radical-induced decomposition (Equations (13) and (14)) [103–105].



This multi-mechanistic activation ensures the sustained and high-yield production of $\cdot\text{OH}$, significantly amplifying the oxidative capacity of the system beyond what either plasma or H_2O_2 could achieve individually. $\cdot\text{OH}$ is one of the most oxidizing free radicals, and its attack is almost non-selective. For pollutants, their main attack methods are classified as: 1. Hydrogen extraction: It attacks the C-H bond on the adipose chain to generate water and an organic free radical, triggering a chain reaction. 2. Electrophilic addition: Directly adding to the C=C double bond or aromatic ring, making its structure unstable and ultimately leading to ring opening and chain breakage.

In the field of plasma-activated degradation of pollutants, Fan et al. and Lee et al. conducted studies focusing on different applications and mechanisms [106,107]. Fan et al. investigated the use of cold plasma-activated hydrogen peroxide aerosols to inactivate food-borne pathogens such as *Salmonella Typhimurium* and *Listeria innocua* on tomato surfaces. Their research demonstrated that the efficacy of the treatment was primarily influenced by the concentration of hydrogen peroxide, with higher concentrations leading to greater bacterial reduction. Specifically, hydrogen peroxide concentrations above 4.2% achieved an average 5-log reduction in *L. innocua* on smooth tomato surfaces, while concentrations above 5.7% were required for similar reductions in *Salmonella*. However, bacterial reduction on stem scars was limited to less than 2 logs, highlighting the challenges of treating complex surface structures. The study emphasized the synergistic effect of combining cold plasma with hydrogen peroxide, enhancing microbial inactivation compared to either treatment alone. Lee et al. demonstrated that combining hydrogen peroxide with in-package cold plasma (HCP) synergistically inactivated indigenous bacteria, *E. coli* O157:H7, and *Listeria monocytogenes* in mixed vegetables, achieving reductions of 1.0–1.3 log CFU/g. The HCP treatment also inhibited microbial growth during refrigerated storage, extended shelf life, and reduced the abundance of spoilage-related bacteria like *Pseudomonas* and *Pantoea*, without adversely affecting vegetable quality or inducing endocrine disruptor leaching from PET packaging. Their study highlighted the potential of plasma-based advanced oxidation processes for efficient and rapid degradation of persistent organic pollutants.

The integration of plasma with H_2O_2 establishes a highly synergistic and efficient advanced oxidation platform [108]. By leveraging the multi-faceted activation capabilities

of plasma, this hybrid system surmounts the kinetic limitations of H_2O_2 , enabling rapid and cost-effective degradation of a broad spectrum of organic pollutants [109]. Its operational flexibility, environmental compatibility, and proven efficacy in both synthetic and real wastewater position the plasma/ H_2O_2 process as a robust and promising technology for modern water remediation challenges. See Table 5 for relevant studies on plasma activation of H_2O_2 .

Table 5. Research on plasma activation of H_2O_2 .

Author	Target	H_2O_2 Concentration	Conditions	Time/min	Degradation Rate
Wu et al. [78]	Tetracycline (TC)	95 mg/L	Initial concentration: 20 mg/L Voltage: 21 kV Frequency: 50 Hz Conductivity (σ): 200 $\mu\text{S}/\text{cm}$ pH: 5	60	52%
Ma et al. [104]	Auricularia auricula polysaccharide (AAP)	2% (<i>w/v</i>)	Initial concentration: 0.3% (<i>w/v</i>) Electrodes distance: 2 mm	180	91%
Lee et al. [107]	Bacillus coli	20%	Initial concentration: 4.6 log CFU/g Voltage: 24.5 kV	3	94%
Boscariol et al. [108]	Bacillus subtilis var. niger ATCC 9372 (Bacillus atrophaeus)	5%	Power: 400 W	40	Almost 100%
Kim et al. [109]	Salmonella	20%	Voltage: 24.5 kV	2 min	Inactivation level: 0.9 ± 0.1 log CFU/g

3.5. Fenton Reagents ($\text{Fe}^{2+}/\text{Fe}^{3+}$)

The Fenton reaction, employing iron ions ($\text{Fe}^{2+}/\text{Fe}^{3+}$) and H_2O_2 generate hydroxyl radicals ($\cdot\text{OH}$), represents a cornerstone technology in advanced oxidation processes [110,111]. Its efficacy primarily relies on the catalytic cycle between Fe^{2+} and Fe^{3+} , as described by the classical reaction (Equation (15)) [112]. However, the conventional homogeneous Fenton process faces significant operational challenges, including a stringent optimal pH range (2.5–3.5), the accumulation of Fe^{3+} sludge requiring post-treatment, and the inefficient decomposition of H_2O_2 which leads to high chemical consumption [113–115].



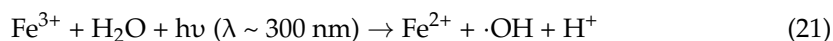
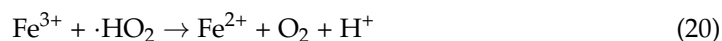
Plasma technology offers a sophisticated solution to these limitations by serving as a multi-functional enhancer for the Fenton system [116]. The integration of plasma introduces several complementary mechanisms that profoundly intensify the $\text{Fe}^{2+}/\text{Fe}^{3+}$ catalytic cycle and radical generation [117]. The enhancement pathways are multi-faceted:

In situ H_2O_2 Production: Plasma discharge in or over water directly generates H_2O_2 , providing a continuous and endogenous source of the essential Fenton reagent, thereby reducing or eliminating the need for external chemical addition (Equations (16)–(18)) [118].



Accelerated Iron Redox Cycling: The plasma environment actively promotes the reduction of Fe^{3+} back to Fe^{2+} , which is the rate-limiting step in the classical Fenton cycle. This is achieved through direct electron reduction, radical-mediated reduction and UV Photoreduction (Equations (19)–(21)) [119].





Multi-Radical Synergy: Beyond $\cdot\text{OH}$ from Fenton reactions, plasma itself produces a rich array of additional reactive species (e.g., O_3 , $\cdot\text{O}_2^-$, $^1\text{O}_2$) that can contribute to pollutant degradation through non-Fenton pathways, creating a multi-oxidant attack strategy [120].

The core of it is the classic Fenton reaction, which generates a large amount of $\cdot\text{OH}$ through Fe^{2+} catalyzing H_2O_2 . These $\cdot\text{OH}$ groups launch indiscriminate and explosive attacks on pollutants, effectively disrupting stable aromatic ring structures, breaking long-chain alkanes, and ultimately mineralizing organic matter into CO_2 and H_2O . This system is particularly effective for the overall mineralization (COD removal) of high-concentration and complex organic wastewater.

A large number of studies have demonstrated the profound synergy of the plasma/Fenton system. Kavian et al. demonstrated that integrating plasma with the Fenton/photo-Fenton process significantly enhances phenol degradation in wastewater [121]. Their study showed that adding 0.4 mmol/L of Fe^{2+} increased the phenol removal efficiency from 56.8% (plasma alone) to 86.93% within 10 min, while the pseudo-first-order kinetic constant rose from 0.0824 min^{-1} to 0.2073 min^{-1} . The system promoted in situ H_2O_2 generation and facilitated $\text{Fe}^{2+}/\text{Fe}^{3+}$ cycling under plasma irradiation, which contributed to continuous $\cdot\text{OH}$ production. Moreover, the efficiency was strongly influenced by parameters such as initial phenol concentration, pH, and treatment time, with acidic conditions favoring the process. Similarly, Liu et al. constructed a “plasma + iron” system coupling discharge plasma with iron microelectrolysis for Cu-EDTA decomplexation [122]. This combination demonstrated a distinct synergy, with the decomplexation efficiency and energy efficiency being 1.53 and 2.31 times higher, respectively, than in the plasma-alone system. The synergy originated from multiple mechanisms: the plasma’s acidic and oxygen-rich environment activated the iron powder, inducing Fenton reactions that converted plasma-generated H_2O_2 into $\cdot\text{OH}$; Fe^{3+} displaced Cu^{2+} from the stable Cu-EDTA complex due to a higher complexation constant; and the resulting iron hydroxides flocculated and removed the released copper ions. This multi-process catalysis, triggered by the plasma, allowed for efficient operation over a wider pH range and mitigated the negative effects of radical scavengers, as $\cdot\text{OH}$ could also be generated on the iron particle surfaces. See Table 6 for relevant studies on plasma activation of $\text{Fe}^{2+}/\text{Fe}^{3+}$. The comparison of synergy between plasma and different homogeneous catalysts is shown in Table 7 and Figure 4.

Table 6. Research on plasma activation of $\text{Fe}^{2+}/\text{Fe}^{3+}$.

Author	Target	$\text{Fe}^{2+}/\text{Fe}^{3+}$ Concentration	Conditions	Time/min	Degradation Rate
Tao et al. [119]	Methyl orange (MO)	$\text{FeSO}_4 \cdot 7\text{H}_2\text{O} : \text{FeCl}_3 \cdot 6\text{H}_2\text{O} = 1 \text{ mol} : 5 \text{ mol}$	Initial concentration: 200 mg/L	6	99%
Grbić et al. [120]	Lignin	$\text{FeCl}_3 \cdot 6\text{H}_2\text{O} : \text{H}_2\text{O}_2 = 1 : 25$	/	30	53%
Kavian et al. [121]	Phenolic	0.4 mM	Initial concentration: 100 mg/L pH: 5.95	10	87%
Liu et al. [122]	Cu-ethylenediaminetetraacetic acid (Cu-EDTA)	2.0 g/L	Initial concentration: 0.3 mM Voltage: 6 kV Frequency: 7 kHz Power: 25.6 W pH: 2	2	72%
Kim et al. [123]	Rhodamine B (RhB)/Reactive black 5 (RB5)	1.0 g/L	Initial concentration: 10 mg/L Frequency: 22 kHz Power: 100 W Gas flow rate: 20 LPM	30	Almost 100%

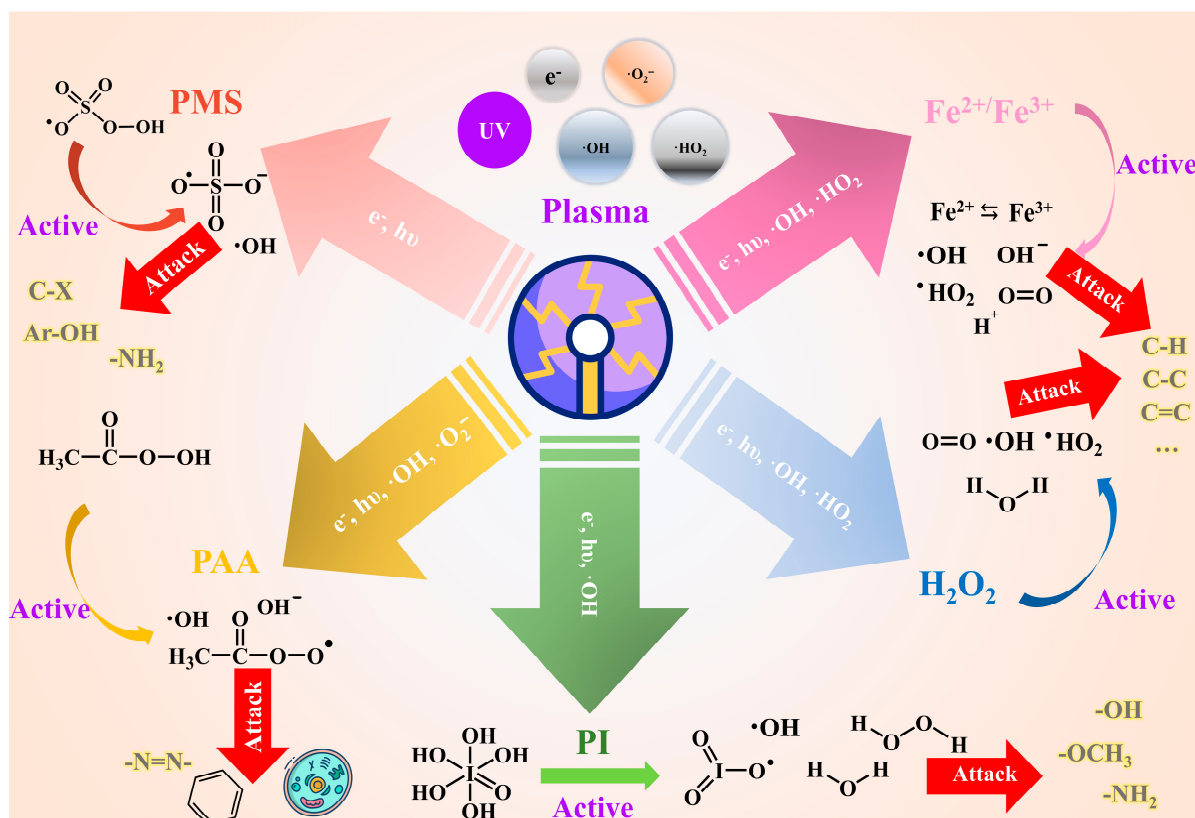


Figure 4. Classification of plasmas coupled to homogeneous catalyst.

Table 7. Comparison of different homogeneous catalysts assisted by plasma.

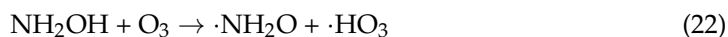
Catalyst System	Primary Active Species	Core Synergistic Mechanism	Advantages	Disadvantages	Ref.
PMS	$\cdot\text{SO}_4^-$, $\cdot\text{OH}$, $\cdot\text{O}_2^-$, $^1\text{O}_2$	Multi-path activation by electrons/UV	Broad pH range, high selectivity	Cost, sulfate residue	[124]
PAA	$\cdot\text{CH}_3\text{C(O)O}$, $\cdot\text{OH}$, $^1\text{O}_2$	O-O bond cleavage to organic radicals	Eco-friendly, diverse pathways	High cost, complex byproducts	[125]
PI	$\cdot\text{IO}_3$, $\cdot\text{IO}_4$, $\cdot\text{OH}$	Electron transfer to iodine radicals	Self-sufficient, low cost	Limited yield, mass transfer issues	[126]
H_2O_2	$\cdot\text{OH}$, $\cdot\text{HO}_2$	In situ generation and activation	Neutral pH operation, less sludge	Iron separation, anion interference	[127]
$\text{Fe}^{2+}/\text{Fe}^{3+}$	$\cdot\text{OH}$, $\cdot\text{HO}_2$	Enhanced $\text{Fe}^{2+}/\text{Fe}^{3+}$ cycle and H_2O_2 production	High selectivity, fast kinetics	Cost, iodinated byproducts	[120]

3.6. Other Homogeneous Catalysis

In addition to the well-established homogeneous catalysts such as PMS, PAA, PI, H_2O_2 , and $\text{Fe}^{2+}/\text{Fe}^{3+}$, plasma technology has also demonstrated significant synergistic effects with other homogeneous catalytic systems. These systems leverage the unique reactive environment of plasma to activate or enhance the performance of less conventional catalysts, thereby broadening the scope of plasma-enhanced AOPs [128–130].

One notable example is the use of hydroxylamine (HA) as a homogeneous catalyst in conjunction with discharge plasma. Huang et al. investigated the synergistic degradation of tetracycline (TC) using a plasma/hydroxylamine system [131]. Hydroxylamine, which contains $-\text{NH}_2$ functional groups, can be activated by plasma-generated electrons and ozone to produce additional hydroxyl radicals ($\cdot\text{OH}$), thereby enhancing the degradation

efficiency of organic pollutants. The activation mechanisms can be described as follows (Equations (22) and (23)):



In their study, the addition of 0.25 mmol/L hydroxylamine increased the degradation efficiency of tetracycline from 70.7% (plasma alone) to 90.0%, with the kinetic constant rising from 0.067 min^{-1} to 0.108 min^{-1} . G_{50} value also improved from 2.28 g/kWh to 3.18 g/kWh. The synergy was attributed to the enhanced generation of $\cdot\text{OH}$, $\cdot\text{O}_2^-$, and $^1\text{O}_2$, as confirmed by ESR and quenching experiments.

Another promising homogeneous catalyst is persulfate (PS), which has been widely used in sulfate radical-based AOPs. Shang et al. demonstrated the synergistic degradation of Acid Orange 7 (AO7) using a DBD plasma–persulfate system [132]. The activation of persulfate by plasma occurs via multiple pathways, including UV photolysis and electron-induced cleavage (Equations (24) and (25)):



The combined system achieved a 60% increase in decolorization efficiency and a 6.7-fold enhancement in the degradation rate constant compared to plasma alone. The mineralization efficiency was also significantly improved, with TOC removal increasing from 14% to 58% under optimal conditions.

The integration of plasma with other homogeneous catalysts such as hydroxylamine and persulfate exemplifies the versatility and adaptability of plasma-catalytic systems [133,134]. These combinations not only enhance the generation of reactive species but also improve degradation kinetics, energy efficiency, and operational flexibility. The multi-pathway activation mechanisms, including electron transfer, UV photolysis, and radical-mediated reactions, underscore the potential of plasma to synergize with a wide range of homogeneous catalysts, opening new avenues for the efficient degradation of refractory organic pollutants in water.

To quantitatively address the critical aspect of energy consumption, Table 8 compares the energy efficiency metrics of various plasma-catalyst systems reported in the literature. Key indicators such as the G_{50} value and energy yield highlight the significant synergy achieved by plasma-catalytic processes, while also underscoring the need for further optimization to reduce absolute energy costs (kWh/m³) for large-scale applications.

Table 8. Comparison of Energy Efficiency for Different Plasma-Catalyst Systems in Degrading Organic Pollutants.

Plasma System	Target Pollutant	Key Energy Efficiency Metric	Value	Ref.
Pulsed Corona	Refractory Wastewater	Energy Consumption (kWh/m ³)	10–100	[58]
DBD/PMS	Perfluorooctanoic acid (PFOA)	Energy Yield (mg/kWh)	399	[74]
DBD/PAA	Bisphenol A (BPA)	G_{50} (g/kWh)	212	[81]
Plasma/PI	Sulfadiazine (SDZ)	Pseudo-first-order k (min ^{−1})	0.5	[91]
Plasma/ Fenton	Phenol	Pseudo-first-order k (min ^{−1})	0.2	[121]
DBD/ Hydroxylamine	Tetracycline (TC)	G_{50} (g/kWh)	3	[131]
DBD/PS	Acid Orange 7 (AO7)	Pseudo-first-order k (min ^{−1})	7	[132]

The examples of hydroxylamine and persulfate, while less commonly featured as the central focus compared to PMS or PAA, serve to underscore the versatility and broad applicability of the plasma activation platform. A systematic analysis reveals that their synergy with plasma adheres to the same fundamental principles governing the primary catalysts: multi-modal activation (via electrons, UV photons, and radicals) and the subsequent generation of secondary reactive species that enhance the degradation process. This observation strengthens the central thesis that plasma is not merely a co-treatment technology but a universal activator. The choice to employ these “other” catalysts is often dictated by specific scenarios: persulfate (PS) is a viable alternative to PMS, particularly when sulfate radical-based chemistry is desired but cost or availability is a concern; hydroxylamine, as a reducing agent, excels in systems where accelerating the metal redox cycle (e.g., in a plasma-Fenton context) is the rate-limiting step. Therefore, this category does not represent a disparate set of outliers but rather expands the toolbox of plasma-activated homogeneous catalysis, demonstrating that the synergistic framework is applicable to a wider range of chemical additives whose properties can be exploited for tailored remediation strategies.

4. Influencing Factors

The efficacy of plasma-enhanced homogeneous catalysis is governed by a complex interplay of parameters that control the formation of reactive species, the activation pathways of catalysts, and the subsequent degradation kinetics of pollutants.

4.1. Plasma Operational Parameters

The plasma source serves as the primary engine for the process, and its operational conditions directly dictate the initial energy input and the physicochemical environment [135]. The input power and applied voltage are fundamental drivers. Higher power typically increases electron density and energy, intensifying the dissociation of background gases (e.g., O_2 , H_2O) and leading to enhanced yields of primary reactive species, including $\cdot OH$, O atoms, and O_3 . This not only facilitates the direct attack on pollutants but also provides a more potent flux of activators (e.g., UV photons, e^- , radicals) for the homogeneous catalyst (e.g., PAA) [136]. However, a non-linear relationship often exists, with an optimal power level beyond which diminishing returns or inhibitory effects are observed. For instance, excessive power can lead to the overproduction of H_2O_2 , which may act as a scavenger for certain critical radicals (e.g., $\cdot CH_3C(O)O$ in PAA systems), thereby reducing the overall catalytic efficiency [137].

The composition of the plasma-forming gas (e.g., air, O_2 , Ar) profoundly influences the spectrum of generated species. Oxygen-rich atmospheres favor the formation of O_3 and $\cdot O$, while inert gases like Ar can enhance UV emission and direct electron impact processes [138]. The reactor geometry, such as the electrode design and the mode of plasma-liquid contact (e.g., submerged discharge, falling film DBD), governs the stability of the plasma discharge, the efficiency of mass transfer of short-lived species from the gas phase to the liquid bulk, and the utilization of concomitant physical effects like UV radiation and shockwaves [139].

4.2. Catalyst Dosage and Activation Kinetics

In homogeneous systems enhanced by plasma, the dosage of the catalyst is a critical variable that must be carefully balanced [140]. A characteristic optimal concentration exists for a given set of conditions. An insufficient dosage fails to fully capitalize on the plasma's activation capacity, limiting the generation of secondary radicals (e.g., $\cdot SO_4^-$ from persulfate or $\cdot R-O$ from PAA). Conversely, an excess of the catalyst can initiate scavenging reactions. For example, high concentrations of PAA can react with the very $\cdot OH$ radicals

it aims to supplement, and high persulfate levels can quench $\cdot\text{SO}_4^-$ radicals [141]. This self-quenching effect can lead to a plateau or even a decrease in the degradation rate of the target pollutant.

4.3. Pollutant-Directed Catalyst Selection

The efficacy of plasma-catalytic systems is profoundly influenced by the intrinsic characteristics of the target pollutants, thereby guiding the selection of an appropriate co-catalyst. Understanding the molecular structure and physicochemical properties of the contaminants is crucial for designing a highly efficient and tailored treatment process. As shown in Figure 5, for recalcitrant organic pollutants, the plasma/PMS system is exceptionally suitable [142]. The $\cdot\text{SO}_4^-$ operates primarily through an electron-transfer mechanism, which is highly effective in mitigating the formation of toxic intermediates. Conversely, when treating pollutants rich in electron-donating functional groups, such as phenols and anilines, the plasma/PI system demonstrates superior performance. The iodine-oxygen radicals (e.g., $\cdot\text{IO}_3$) exhibit remarkable selectivity and high reaction kinetics towards these electron-rich moieties, enabling a targeted degradation approach.

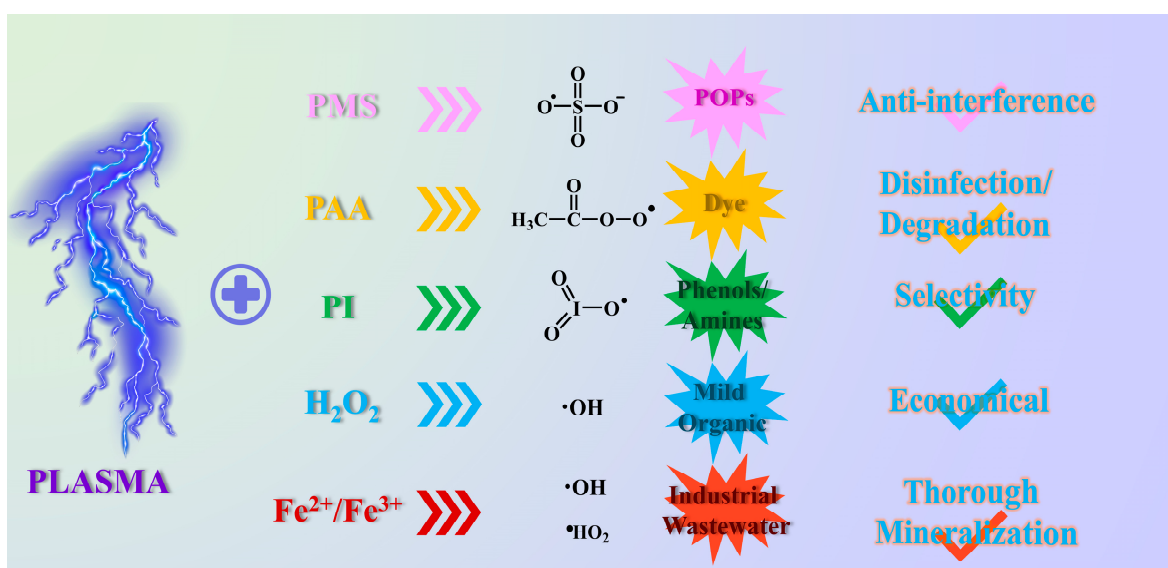


Figure 5. Application scenarios of plasma synergy with different homogeneous catalysts.

Furthermore, the nature of the water matrix itself dictates the optimal strategy. In complex industrial wastewater with significant background organic matter, the non-radical pathway (e.g., $^1\text{O}_2$) prevalent in the plasma/PMS system offers strong anti-interference capability, allowing for selective oxidation of the target contaminants [143]. For aqueous environments requiring concurrent disinfection and contaminant degradation, such as medical wastewater, the plasma/PAA system emerges as the optimal choice [144]. The organic oxygen radicals generated from PAA activation, combined with PAA itself, create a powerful synergistic effect for efficient microbial inactivation and organic pollutant removal without generating harmful halogenic by-products. In scenarios involving high-concentration, broad-spectrum organic pollution, such as the production of textile or chemical processing wastewater, the robust and non-selective $\cdot\text{OH}$ produced by the plasma/Fenton system provides the intense oxidative power necessary for effective mineralization and COD reduction [145].

Optimizing the plasma-coordinated homogeneous catalytic process requires systematic consideration of the synergy and trade-off between operating parameters, catalyst

metering, water matrix and target pollutant characteristics, so as to achieve efficient, economical and targeted wastewater treatment applications.

5. Mechanisms of Pollutant Degradation

The foundation of pollutant degradation in plasma systems lies in the generation of diverse reactive oxygen and nitrogen species (RONS). These species include short-lived radicals ($\cdot\text{OH}$, $\cdot\text{O}$, $\cdot\text{O}_2^-$), long-lived oxidants (H_2O_2 , O_3), and nitrogen-derived species, as well as excited molecules generated under different plasma conditions. Figure 6 summarizes the major plasma-generated reactive species and compares the oxidation potentials of key oxidants involved in plasma-activated water chemistry.

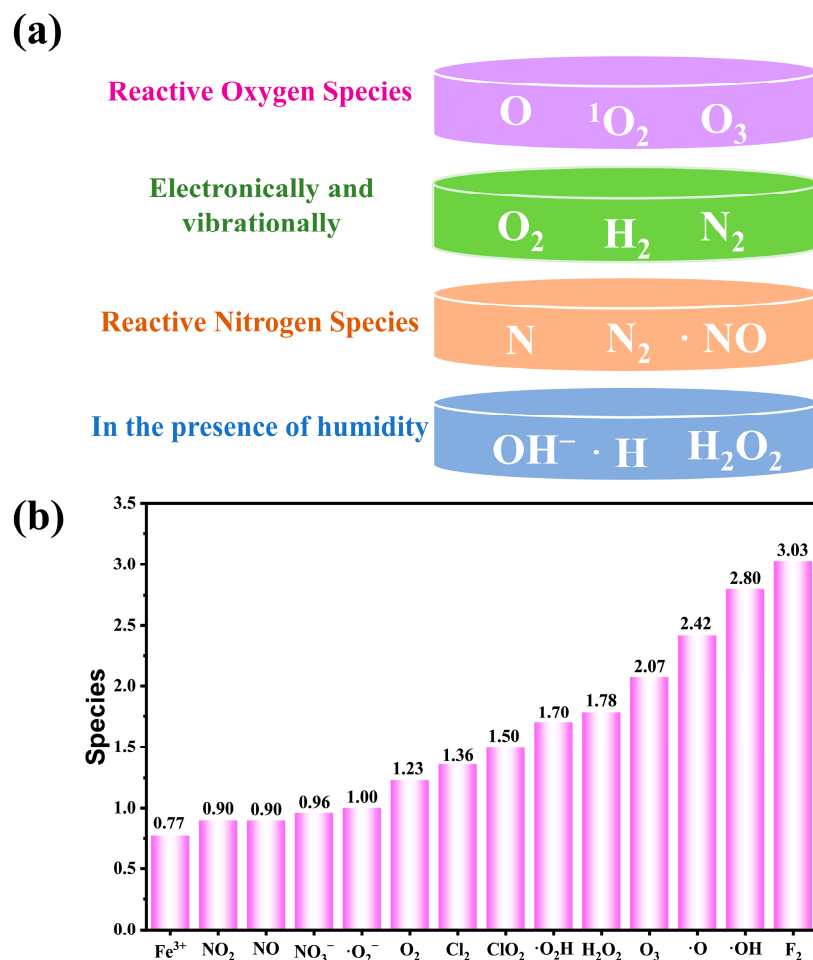


Figure 6. (a) The reactive substances produced by the discharge plasma system in the gas phase and liquid phase; (b) Comparison of the oxidation potentials of the main oxidants produced by discharge plasma in water.

In gas-phase discharge (e.g., DBD, corona over water), high-energy electrons primarily collide with gaseous molecules (H_2O , O_2 , N_2). This dissociates and excites these molecules, initiating a cascade of reactions to form the primary reactive oxygen and nitrogen species (RONS). These gas-phase RONS must then diffuse and dissolve into the liquid phase to attack pollutants, a process where the gas–liquid interfacial area and mass transfer efficiency become critical limiting factors.

Conversely, in liquid-phase discharge, electrons directly impact water molecules and dissolved substances within the liquid bulk. This creates an intense reaction zone at the plasma–liquid interface and within plasma channels, resulting in the direct, in situ

production of radicals. The high electric field at the plasma–liquid interface can also cause molecular dissociation.

A key advantage of liquid-phase discharge is the direct aqueous production of $\cdot\text{OH}$, which avoids mass transfer limitations and leads to extremely high localized concentrations of radicals. Furthermore, it can generate aqueous electrons (e^-), a potent reductant, and UV photons that can directly photolyze pollutants and activate catalysts within the solution. The subsequent activation of homogeneous catalysts and the degradation of pollutants are then powered by these reactive species generated from these two fundamental discharge modes [146].

e^- is the primary energy carriers in plasma, it induces the dissociation and excitation of background gas and water vapor molecules through electron impact. This leads to the formation of primary radicals and excited species, as shown in reactions (Equations (26)–(29)) [147]:



These species, particularly $\cdot\text{OH}$ and O_3 , are potent oxidants that can directly attack a wide range of organic pollutants.

At the same time, UV photons can directly photolyze certain pollutants that possess specific chromophores. More importantly, it plays a critical role in activating homogeneous catalysts, such as the photodissociation of the O–O bond in persulfate (Equation (30)) [148]:



The introduction of a homogeneous catalyst significantly expands the reactive species portfolio [149]. The primary plasma-generated species activate these catalysts, producing highly oxidizing secondary radicals. Primary radicals like $\cdot\text{OH}$ are highly effective in activating catalysts. For instance, $\cdot\text{OH}$ can react with PAA to generate organic radicals, and with persulfate to yield sulfate radicals.

The combined action of primary and secondary reactive species opens multiple parallel and sequential degradation pathways for pollutants [150]. $\cdot\text{OH}$ primarily attacks organic pollutants via two mechanisms: (1) Electrophilic Addition to unsaturated bonds or aromatic rings, leading to hydroxylated intermediates; (2) Hydrogen Abstraction from aliphatic C–H bonds, generating carbon-centered radicals that subsequently react with oxygen to initiate decomposition. Organic radicals (e.g., $\cdot\text{CH}_3\text{C}(\text{O})\text{O}$, $\cdot\text{CH}_3\text{C}(\text{O})\text{OO}$) and singlet oxygen ($^1\text{O}_2$) exhibit high selectivity towards electron-rich functional groups. They are particularly effective in attacking specific moieties like phenol rings, anilines, and sulfur-containing groups, which might be less reactive toward $\cdot\text{OH}$ or $\cdot\text{SO}_4^-$ under certain conditions. This selectivity complements the non-selective attack of $\cdot\text{OH}$ and can lead to distinct degradation pathways.

6. Practical Considerations and Challenges

The transition of plasma-catalytic systems from laboratory research to industrial application is primarily hindered by scale-up challenges and complex real wastewater matrices. Scale-up efforts face hurdles in maintaining energy efficiency in continuous-flow reactors, ensuring electrode longevity, and achieving cost-effective large-volume treatment, as pilot

studies on industrial effluents like paper mill wastewater have shown [151]. Simultaneously, real wastewater matrices impose significant performance barriers. Background organic matter (e.g., humic acids) and ubiquitous inorganic anions (e.g., Cl^- , HCO_3^- , NO_3^-) act as potent scavengers of reactive radicals ($\cdot\text{OH}$, $\cdot\text{SO}_4^-$), drastically reducing degradation kinetics [152]. Suspended solids can shield pollutants and foul electrodes. To mitigate these effects, strategies such as employing matrix-resilient non-radical pathways (e.g., $^1\text{O}_2$ in the PMS system) and implementing simple pre-treatment (e.g., filtration) are crucial for enhancing the practical viability of this technology.

Beyond these practical challenges, a critical analysis of the literature reveals apparent contradictions in the reported efficacy of plasma-catalytic systems. These performance variations often originate from fundamental differences in experimental conditions rather than true inconsistencies. Key factors include the type of plasma reactor, which dictates the dominant activation mechanism; the water matrix composition, where the presence of radical scavengers can drastically alter the dominant reaction pathways; and the molar ratios between oxidant, catalyst, and pollutant, which can shift the system from being radical-limited to scavenger-dominated. Therefore, direct comparison of performance between studies requires careful consideration of these contextual parameters, and the optimal system is highly specific to the target wastewater and operational constraints.

7. Conclusions and Prospects

This review has systematically examined the recent advances in plasma-enhanced homogeneous catalysis for the degradation of environmental pollutants. See Figure 7 for summary and outlook. The integration of plasma with homogeneous catalysts such as PMS, PAA, PI, H_2O_2 , and $\text{Fe}^{2+}/\text{Fe}^{3+}$ has demonstrated remarkable synergistic effects, leading to enhanced degradation rates, improved mineralization efficiency, and broader operational pH ranges compared to individual processes. The multi-modal activation capability of plasma, through energetic electrons, UV photons, and radical species, enables the efficient and continuous generation of reactive oxidants, thereby overcoming many limitations of conventional activation methods. Key influencing factors, including plasma operational parameters, catalyst dosage, aqueous matrix composition, and pollutant characteristics, play critical roles in determining system performance. The complex interplay among these factors underscores the need for optimized and context-specific system design.

Despite significant progress, several challenges remain to be addressed before widespread practical implementation can be achieved: (1) Although plasma-catalytic systems show improved energy utilization compared to plasma-alone processes, further enhancements are needed to reduce energy consumption and improve cost-effectiveness. (2) Current plasma reactors are often lab-scale. Developing scalable, robust, and efficient reactor configurations suitable for industrial applications is essential. (3) The design of novel homogeneous catalysts with higher stability, lower cost, and better compatibility with plasma environments is a promising direction. (4) Future studies should focus on testing systems in complex real wastewater to evaluate the impact of natural organic matter, ions, and other constituents on treatment efficacy. (5) Understanding and minimizing the formation of toxic intermediates or by-products during degradation is crucial for environmental safety. (6) Combining plasma-catalytic systems with other treatment technologies (e.g., adsorption, membrane filtration) could enhance overall treatment performance and feasibility.

In conclusion, plasma-enhanced homogeneous catalysis represents a highly promising and versatile strategy for the effective degradation of refractory organic pollutants. With continued research focused on overcoming existing challenges, this technology holds great potential to become a mainstream solution for sustainable water purification.

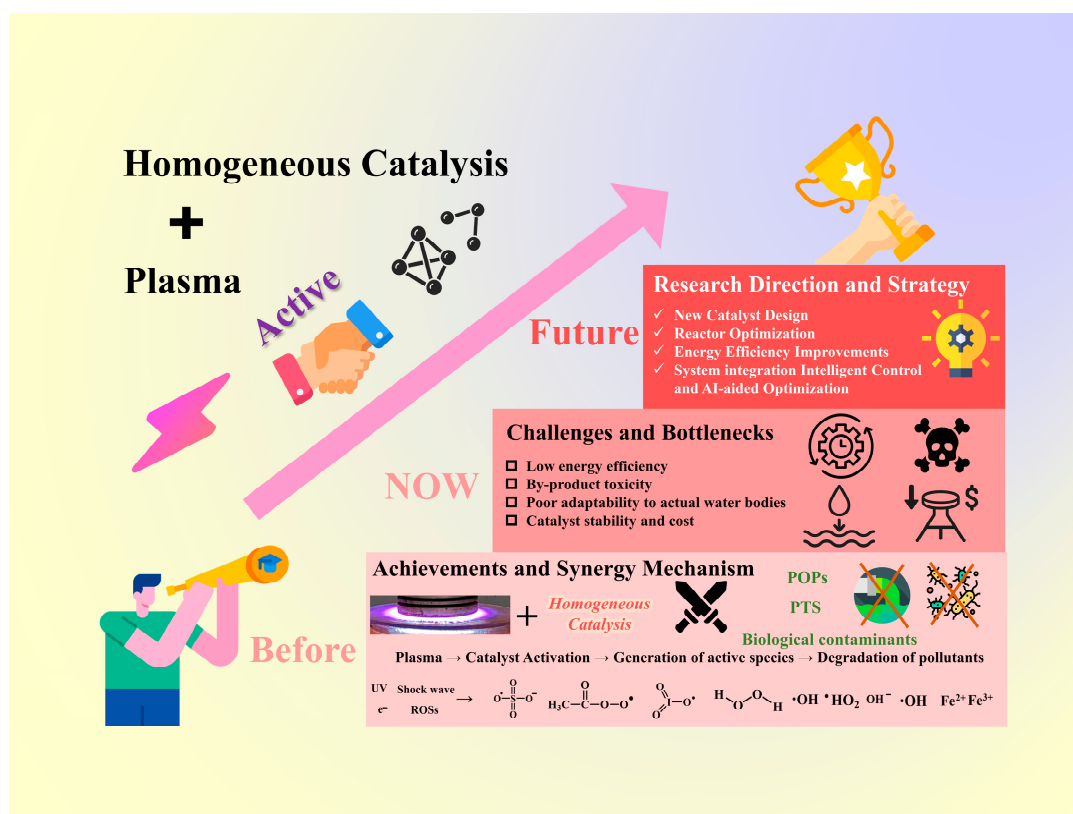


Figure 7. Summary and outlook of synergy between plasma and homogeneous catalysts.

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