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ABSTRACT

Owing to the water crisis, the development of innovative and clean advanced oxidation processes to decompose a variety of harmful organic compounds in wastewater becomes the main challenge for many research teams. Cold discharge plasma is one of the most widely studied and developed processes, owing to its low energy cost and easy to operate. The impact of different factors on the decontamination effectiveness of discharge plasma are detailed in this review. The generation and reaction mechanisms of reactive species in discharge plasma systems have also gained a significant interest and hence discussed. Several potentials and laboratory-scale reactor design recently reported are discussed and schematically presented. The recent combination of discharge plasma decontamination and other processes in both post and pre-treatment configuration are reported. Some applications of water treatment based on discharge plasma at the pilot scale have been addressing.

1. Introduction

The increasing demand for clean water, due to the fast-growing world population, has pushed scientists to develop various conventional processes for wastewater treatment such as physical, chemical, and biological water treatment. Among them, chemical techniques are widely used. However, all these techniques have several disadvantages, like inorganic solid and unreacted chemical residues [1–3]. To overcome these issues, researchers have focused on advanced oxidation processes (AOPs), like photo-Fenton [1] and non-thermal plasma (NTP) techniques [2–4].

The application of plasmas in water treatment has gained immense attention due to its simplicity, effectivity for destroying toxic organic compounds in both raw water and wastewater, economy, and environmentally friendly side [5,6].

In the last years, plasma-liquid systems have attracted considerable attention of researchers on a variety of applications domains that include, **i**.e., environmental and wastewater remediation [7,8], nanomaterial processing [9,10], cancer cells treatment [11], sterilization and disinfection [12], bacterial biofilm prevention and eradication [13] and analytical chemistry [14,15] … etc. In particular, plasma treatment techniques have been tested: i) for the decomposition of many harmful compounds from water, including; herbicide [16], pesticides [17], phenols [7,18], organic dyes [19], As^{3+}, Cr^{6+}, Fe^{3+}, Cu^{2+} and Zn^{2+} [20,21], biomolecules, Virus and bacterial Inactivation [22–25], pharmaceuticals [26,4] and veterinary antibiotics [27]; ii) For the possible removing of chemical and biological wastes in all three states; gas, liquid and solid [28]; for the enhancement of biodegradability of coking wastewater [29].

In this review, various aspects dealing with wastewater treatment by electrical discharge plasma are discussed in detail. Among them, the reaction mechanism for different discharge plasma conceptions, numerous classifications according to various criteria, the effect of different parameters on the process efficiency, some advanced reactor designs, and the development of combined processes involving discharge plasma as a post or pretreatment step. On the other hand,

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some practical advantages and disadvantages in plasma applications have been reported and the scaling up of this technology at the pilot-scale to treating contaminated water. At our knowledge, there are a very few reviews on plasma combined with other processes at different scales are available in literature.

2. Principles and mechanisms of plasma for Water treatment

In recent years the use of discharge plasma for the elimination and/or the complete mineralization of both hazardous organic compounds (Pharmaceuticals products and synthetic dyes) and pathogenic bacteria in wastewater have gained continuous growth and development [30,31]. Plasma is a source of high electric field, energetically charged particles, oxidizing species (radicals and molecules), reductive species (Hydrogen atom, Nitrogen atom and aqueous electron…etc) [7] (Fig.1), ultrasound, UV light, electrohydraulic cavitations, as well as even shockwaves [3,5,32].

Two distinguishing effects can be caused by discharge plasma in contaminated water purification: i) direct effects resulting from electron collisions and ii) other impacts generated by ionic, molecular (H_2O_2, O_2 and O_3) and a host of chemically active species in-situ generated ($^{\bullet}OH$, $O^{\bullet}$, $O_2^{\bullet-}$ and $H^{\bullet}$) via pyrolysis and photolysis reactions [33]. As reported in Fig.2, theses oxidants/reactants have different life times and oxidation potentials [7,34,35]. Nevertheless, aqueous pollutants can also be destroyed by reductive degradation pathways under the effect of some reductive species, such as aqueous electron (E° H_2O/e_{aq} = -2.77 V) [36] and $H^{\bullet}$ radicals (E° $H_2O/H^{\bullet}$ = -2.30 V) [37].

It is worthy to note that electrical discharges in liquids launch various chemical and physical impacts responsible for the harmful microorganisms deactivation: i) the electromechanical compression phenomenon is responsible for the inactivation of bacterial cells by initiating pores in the membrane cells; ii) a deformation of cells structure could be caused by shock waves in the range of 5 – 20 kBar; iii) organism mutation may be prompted by intense UV radiations when ranged between 200 and 400 nm [31].

Some aspects must be noted in view to understand what happened in an aqueous medium during discharge plasma. Firstly, when the plasma is initiated there is a considerable increase in temperature (heating rate $\approx 10^9$ K.s^{-1}) and secondly a limited volume of water prevents the plasma to spread freely and then a high pressure (10^5-10^7 MPa) is generated as a result [28]. In an incompressible medium, the high pressure would transform into impact shockwave, leading to plasma generation as final step [5,38,39], as the result of high-voltage discharge followed by electromagnetic field that can sterilize water and wastewater [40,41]. The resultant shockwave can produce pyrolysis [42] and chemical reactions in the bulk liquid indirectly by electrohydraulic cavitations [43].

The mechanism of wastewater decontamination by high-voltage electrical discharge can be divided, according to the plasma-phase

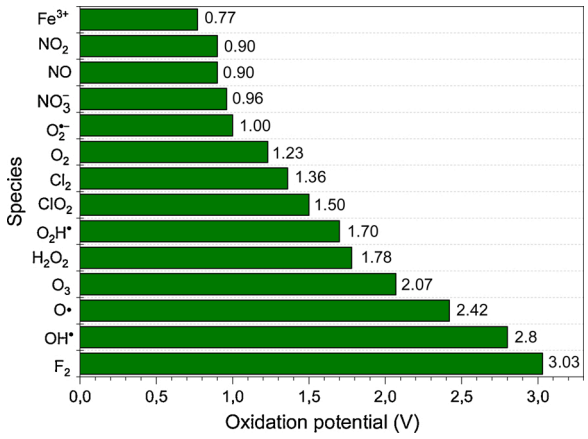

Fig. 2. Comparison of oxidation potential of major oxidants produced by discharge plasma in water.

distribution or reactor operating principles, mainly into three categories, specifically, electrical discharges above the liquid surface, interface plasma-liquid discharge, and direct electrical liquid discharge [5]:

a) Discharge in the gas phase above the liquid (Fig.3.a),
b) Direct discharges in the liquid phase (Fig.3.b),
c) Discharge in multiphase environments or Hybrid gas-liquid system (bubbles/vapor in liquids) (Fig.3.c).

2.1. *Electrical discharges plasma above liquid surface (nonthermal plasma)*

Plasma driven in the gas phase and contact with water vapor (with the help of both plasma electrons and excited species energies), so-called indirect discharge, creates a large number of reactive oxygen species, as well as nitrate, nitrite and NO radicals as reported in Eq. 1 to 15 [5,44,45].

$$e^{-*} + O_2 \rightarrow O^{\bullet} + O^{\bullet} + e^{-} \quad (1)$$

$$e^{-*} + H_2O \rightarrow H^{\bullet} + HO^{\bullet} + e^{-} \quad (2)$$

$$e^{-*} + O_2 \rightarrow O_2^{+} + 2e^{-} \quad (3)$$

$$e^{-*} + O_2 + M \rightarrow O_2^{-} + M \quad (4)$$

$$O^{\bullet} + O_2 + M \rightarrow O_3 + M \quad (5)$$

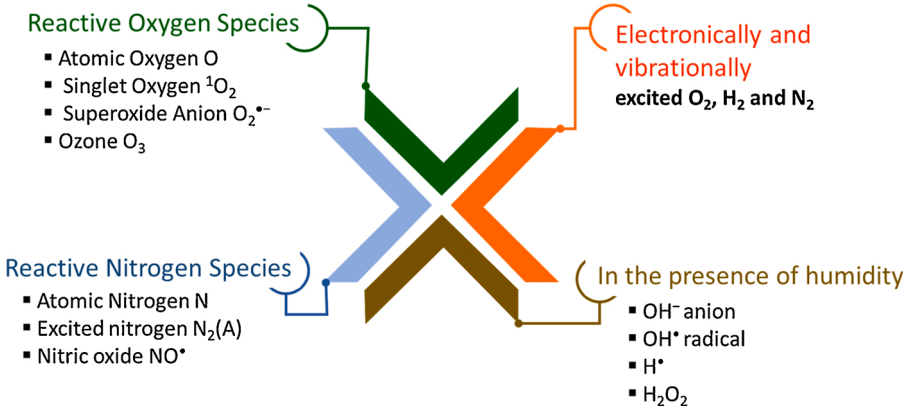

Fig. 1. The reactive species produced in gas and liquid phase by discharge plasma systems.

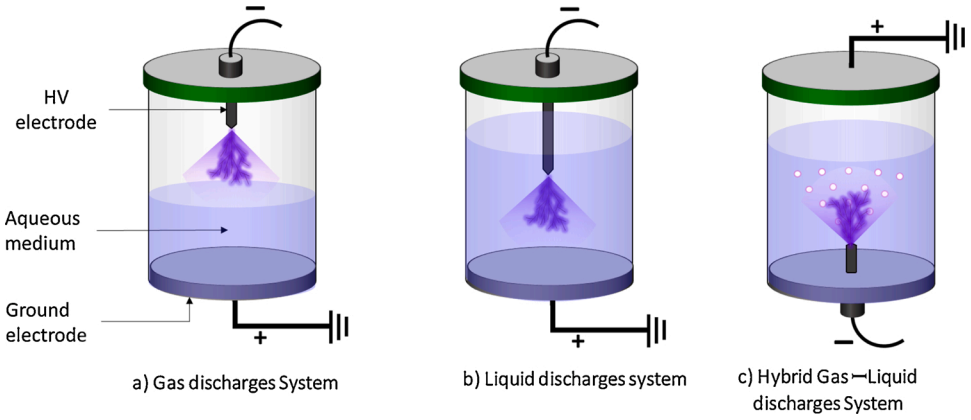

Fig. 3. Schematic of different discharge plasma systems used for water treatment.

$$e^{-*} + 2H_2O \rightarrow H_2O_2 + H_2 + e^- \quad (6)$$

$$HO^\bullet + H_2O_2 \rightarrow H_2O + HO_2^\bullet \quad (7)$$

$$O + H_2O \rightarrow HO^\bullet + HO^\bullet \quad (8)$$

$$O_3 + H_2O_2 \rightarrow HO^\bullet + O_2 + HO_2^\bullet \quad (9)$$

$$O_3 + HO_2^\bullet \rightarrow HO^\bullet + O_2 + O_2^- \quad (10)$$

$$O_3 + h + H_2O \rightarrow H_2O_2 + O_2 \quad (11)$$

$$H_2O_2 + h \rightarrow 2HO^\bullet \quad (12)$$

$$e^{-*} + N_2 \rightarrow 2N^\bullet + e^- \quad (13)$$

$$N^\bullet + O^\bullet \rightarrow NO \quad (14)$$

$$O^\bullet + NO \rightarrow NO_2 \quad (15)$$

These discharges have resemblances with contact glow discharge electrolysis when the metal anode is flooded in the liquid, and the plasma is produced in a vapor layer [46]. In these discharges, a considerable portion of the discharge power is squandered into the liquid causing high liquid evaporation rates [47]. In such a system, less energy from the discharge plasma is used to generate active species.

2.2. Discharge plasma in liquid-gas interface

Interface plasma-liquid discharge, also called discharges in bubbles/vapor on the liquid surface [7], is characterized by a large contact surface area. The latter conducts to high effectiveness for gaseous species to diffuse inside the liquid and less energy required to produce the plasma than direct discharge in the liquid. Also, at the gas-liquid interface, extremely Reactive Species such as H_2O^+, H', O_3, OH, or HO'$_2$ could be produced', and these species would support the destruction of the target pollutants [48–50]. On the one hand, the reaction formation of hydrogen peroxide by hydroxyl radical recombination is notified in Eq. 16 to19 [51].

$$2HO^\bullet \rightarrow H_2O_2 \quad (16)$$

It can take place in the gas, the liquid, or at the interface through the following reactions.

$$2HO^\bullet_{(g)} + M \rightarrow H_2O_{2(g)} + M \quad (17)$$

$$2HO^\bullet_{(aq)} \rightarrow H_2O_{2(aq)} \quad (18)$$

$$2HO^\bullet_{(int)} \rightarrow H_2O_{2(int)} \quad (19)$$

On the other hand, in an atmospheric discharge plasma, a massive flow of electrons is colliding at the water-air interface, inducing a highly reactive interfacial dissolved electrons (aq.) in high concentration [52]. That including other ROS significant amount of 'OH radicals are also produced near the interface.

2.3. Direct electrical discharges in liquid (electrohydraulic discharge)

The discharge plasma in an aqueous medium is generated by nanosecond pulsed power, DC voltages [50], and/or AC excitation using 50–60 Hz up to GHz microwave excitation [53]. It is also required more energy than the electrical discharges above the water level. Moreover, the residence time of plasma is very-short in water because of exchanging high energy electrons with the surrounding water medium (in the case of non-thermal plasma, for example). Indeed, the direct electrical discharge applied in water generates elevated temperature plasma channels, creating electrohydraulic cavitations, supercritical water oxidation, as well as the establishment of short-lived radicals by ultraviolet photolysis [54].

Nevertheless, electrical discharges plasma in water include a simple system and produce chemically active species directly in water that could effectively offense the dissolved contaminants [55].

The essential reactions responsible for free radical production and termination, in the case of electrical discharges in water, and their corresponding rate constants (k) are listed below [44,55,56] in Eq., 20 to 35.

$$H_2O \rightarrow H^\bullet + HO^\bullet \quad 9.25 \times 10^{-10}\ \mathrm{M\ s^{-1}} \quad (20)$$

$$H_2O \rightarrow \frac{1}{2}H_2O_2 + \frac{1}{2}H_2 \quad 1.2 \times 10^{-6}\ \mathrm{M\ s^{-1}} \quad (21)$$

$$2H_2O \rightarrow H_3O^+ + e^- + OH^\bullet \quad 2.35 \times 10^{-9}\ \mathrm{M\ s^{-1}} \quad (22)$$

$$H^\bullet + O_2 \rightarrow HO_2^\bullet \quad 1.0 \times 10^{10}\ \mathrm{M^{-1}\ s^{-1}} \quad (23)$$

$$H + H_2O_2 \rightarrow H_2O + HO^\bullet \quad 1.0 \times 10^{10}\ \mathrm{M^{-1}\ s^{-1}} \quad (24)$$

$$HO + H_2O_2 \rightarrow H_2O + HO_2^\bullet \quad 5 \times 10^{7}\ \mathrm{M^{-1}\ s^{-1}} \quad (25)$$

$$e^-_{eq} + HO^\bullet \rightarrow OH^- \quad 3 \times 10^{10}\ \mathrm{M^{-1}\ s^{-1}} \quad (26)$$

$$e^-_{eq} + H + H_2O \rightarrow OH^- + H_2 \quad 2.5 \times 10^{10}\ \mathrm{M^{-1}\ s^{-1}} \quad (27)$$

$$e^-_{eq} + H_2O_2 \rightarrow HO^\bullet + OH^- \quad 1.2 \times 10^{10}\ \mathrm{M^{-1}\ s^{-1}} \quad (28)$$

$$H^\bullet + HO^\bullet \rightarrow H_2O \quad 2.4 \times 10^{10}\ \mathrm{M^{-1}\ s^{-1}} \quad (29)$$

$$2HO^\bullet \rightarrow H_2O_2 \quad 4.0 \times 10^{9}\ \mathrm{M^{-1}\ s^{-1}} \quad (30)$$

$$2HO_2^{\bullet} \rightarrow H_2O_2 + O_2 \quad 2.0 \times 10^6\ M^{-1}\ s^{-1} \tag{31}$$

$$H^{\bullet} + HO_2^{\bullet} \rightarrow H_2O_2 \quad 1.0 \times 10^{10}\ M^{-1}\ s^{-1} \tag{32}$$

$$2H^{\bullet} \rightarrow H_2 \quad 1.0 \times 10^{10}\ M^{-1}\ s^{-1} \tag{33}$$

$$HO_2^{\bullet} + HO \rightarrow H_2O + O_2 \quad 1.0 \times 10^{10}\ M^{-1}\ s^{-1} \tag{34}$$

$$H_3O^+ + OH^- \rightarrow 2H_2O \quad 3.0 \times 10^{10}\ M^{-1}\ s^{-1} \tag{35}$$

3. Parameters affecting electrical discharge in water treatment efficiency

3.1. Input power

Input power applied in discharge Plasma plays an essential role in the degradation and/or mineralization efficiency of pollutants, where the increase in input power leads to an enhancement in the removal rate [45,57]. Chen et al. reported the degradation of phenol by pulsed high-voltage discharge plasma in an aqueous medium. These authors found that increasing supply voltage from 7 to 10 kV increases the removal efficiency from 55 to 86% [2]. A similar result was found by Kim et al. [27,58]. These results were mainly attributed to the increase in the amount of highly chemically active species produced by increasing the input voltage [59].

The increasing electron concentration can explain the relationship between input power and the degradation yield in the reaction medium, more ozone production [60], high intensity of ultraviolet radiation [61] and the generation of a large number of reactive oxygen species such as $O^{\bullet}$ and $O^{\bullet}H$ [62,63]. In other words, as the input power rises, electrons acquire a higher amount of energy in the electric field, and hence, as a result of the collision, further ionization of oxygen and water molecules is induced [64].

3.2. Pollutant`s initial concentration

As reported in several works, the initial concentration of pollutants significantly affects the removal rate. On the one hand, it is apprized that the increase in pollutant concentration increases the reaction rate until the generation of active species becomes the limiting step in the process [57]. On the other hand, the increase in the pollutant concentration decreases the removal efficiency as well as the Chemical Oxygen Demand (COD), as reported by Tao et al., for methyl orange degradation [19]. This finding could be explained by the fact that there is a competition between the available active species produced by discharge plasma and pollutant molecules and/or its generated intermediates species (at low initial pollutant concentration). [45,58,63].

3.3. Water conductivity

It is essential to take into consideration the solution conductivity, to be treated in the discharge plasma process, regarding decontamination efficiency [65]. A high electrical conductivity of water, owing to high ions concentration, can cause a high current flow, and a large amount of chemically active species could be generated. For example, as reported by Clements et al., weak discharge plasma is observed in deionized water [66]. On the contrary, Jiang et al. found that increasing the medium conductivity leads to a decrease in the removal efficiency of methyl orange [67]. They have suggested that the high discharge current leads to more intense UV light and vigorous acoustic waves formation, which favors heating of the solution [5]. Then less energy is exploited to produce H_2O_2, O_3, and to destruct the organic contaminant.

It appears, therefore, that the concentration of produced reactive ions in the plasma is limited by an optimum electrical conductivity of the solution. If the water conductivity is beyond an optimum value, this could decrease the discharge plasma efficiency due to the fast compensation of the space charge electric field on the streamer head (shorter streamer channel length), and hence fewer active species are produced [38].

Also, Abdelaziz and his co-workers reported that the effect of solution conductivity on the power consumption is only noticeable at relatively high applied voltage, i.e., when the plasma channels spread in the bubbles inside the solution. Nevertheless, the solution conductivity had no role in the degradation efficiency and the pH of the treated solution at the same input power [68].

3.4. Effects of reactor design and discharge gap

It has been found that the distance between electrodes, i.e., the discharge gap, plays an essential role in the degradation efficiency of harmful compounds because changing electrode distance leads to a change in the electric field intensity. Accordingly, different electrical discharge types have been tested; some of them are shown in Fig.4 [39]. As reported by Chen et al., the increase in electrodes distance increases the time needed to achieve a given removal efficiency of pollutants [69]. From another hand Sano et al. in their work on the degradation of organic compounds in water reported that cathode-anode gap has an optimal value at a given voltage input to achieve a maximum Rhodamine B degradation yield [70].

Stratton et al. notified that the discharge plasma area increases with increasing the grounded electrode diameter or surface at fixed electrodes distance [71]. It can be concluded that the design of electrodes must assure a uniform plasma distribution and a possible sizeable electrical discharge area [7].

3.5. Feed gas composition

In the discharge plasma treatment technology, the input gas quality and quantity showed a remarkable effect on the treatment process. Kim et al. investigated the effect of oxygen concentration on the degradation of antibiotics and found that oxygen showed better efficiency as compared to dry air [27].

Manoj Kumar Reddy et al. found that the increase in gas flow rate increases the removal efficiency of pollutants by increasing the number of reactive species produced and ozone generated. However, beyond a limit value of gas flow rate, no degradation efficiency enhancement is noticeable. These results are verified by the increasing of measured unreacted O_3 concentration at the system outlet [45].

3.6. Effects of reactive ions on pollutant removal efficiency

Many works focused on the effect of different ions on the removal efficiency and the degradation of energy yield by discharge plasma. Interestingly, the addition of Na_2SO_4 in the treatment system generates $SO_4^{\bullet -}$ ions ($E_0 = 2.6$ V) (Eq. 36), which has a powerful oxidation effect comparable to hydroxyl radicals [45,72].

$$SO_4^{-2} + HO^{\bullet} \rightarrow OH^{\bullet} + SO_4^{\bullet -} \tag{36}$$

Also, sulfate radical has an interesting capacity to attack dye molecules at different positions leading to a fast fragmentation of chromophores [73]. Nevertheless, the increase of $SO_4^{\bullet -}$ ions in the discharge plasma medium leads to an increase in the degradation efficiency until a certain amount. In contrast, beyond this amount, they become inefficient because of the recombination of reactive radicals and the balance generation at the high amount of the reactive species produced (Eq. 37 and 38) [74].

$$HO^{\bullet} + HO^{\bullet} \rightarrow H_2O_2 \tag{37}$$

$$SO_4^{-2} + SO_4^{-2} \rightarrow S_2O_8^{2-} \tag{38}$$

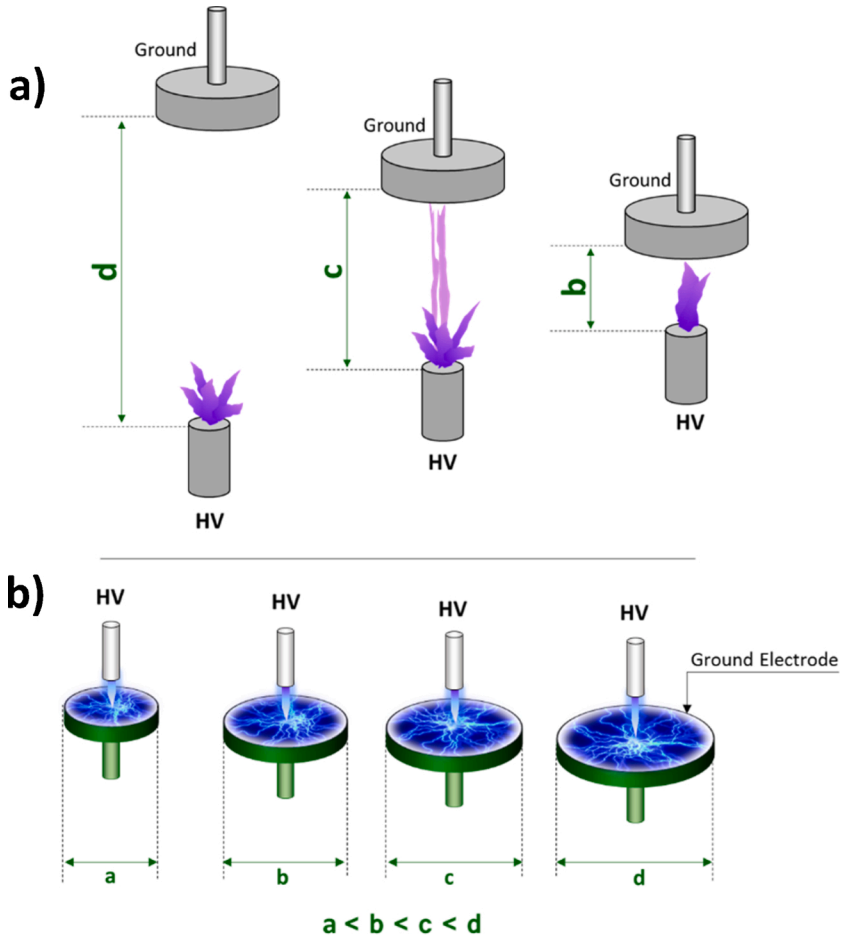

Fig. 4. Reactor design on discharge plasma characteristics: a) discharge gap and b) electrodes surface.

The presence of inorganic mineral pollutants, such as Fe^{2+} **h**as showed a positive effect on the degradation efficiency in both photocatalysis [75] and electrical discharge plasma [76–78], since it can react with H_2O_2 and then leading to the production of more powerful reactive species [79]. For instance, the H_2O_2 reacts with Fe^{2+} ions and produces secondary oxygen species ($HO_2^{\bullet}$) with high oxidizing power, as reported in Eq. 39–41. [80–82]:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{\bullet} + HO^{\bullet} \quad (39)$$

$$Fe^{2+} + HO^{\bullet} \rightarrow Fe^{3+} + OH^{\bullet} \quad (40)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^{\bullet} + H^{+} \quad (41)$$

In their work, Marotta et al. found that there is no signification effect of being noticed with a varying iron concentration in the aqueous solution. They explained that by the deficient H_2O_2 concentration (<1.5 10^{-4} M), namely below the detection limit and too low for observing a catalytic effect by iron ions [54].

The presence of NaCl, as chloride source, was found to harm the degradation efficiency [72] because chloride ion reacts with the hydroxyl radicals, as illustrated in Eq. 42 to 46 [83].

$$NaCl \rightarrow Na^{+} + Cl^{\bullet} \quad (42)$$

$$Cl^{\bullet} + HO^{\bullet} \rightarrow HOCl^{\bullet} \quad (43)$$

$$Cl^{\bullet} + HO^{\bullet} \rightarrow Cl^{\bullet} + OH^{\bullet} \quad (44)$$

$$Cl^{\bullet} + Cl^{\bullet} \rightarrow Cl_2 \quad (45)$$

$$Cl^{-} + Cl^{\bullet} \rightarrow Cl_2 + e^{-} \quad (46)$$

3.7. The pH of the reaction medium

The pH is a parameter that plays a crucial role in the evaluation of the efficiency of discharge plasma-based technique. Several studies showed a proportional relationship between pH and the degradation efficiency of a variety of treated hazardous molecules [27,58,84]. This fact can be explained by the decomposition rate of ozone into more powerful oxidant "hydroxyl radicals" favored under basic pH due to the presence of OH- ions [59], as shown in Eq. 47 to 50 [85,86]:

$$OH^{\bullet} + O_3 \rightarrow O_2^{-} + HO_2^{\bullet} \quad (47)$$

$$O_2^{-} + O_3 \rightarrow O_3^{-} + O_2 \quad (48)$$

$$O_3^{-} + H^{+} \rightarrow HO_3^{\bullet} \quad (49)$$

$$HO_3^{\bullet} \rightarrow HO^{\bullet} + O_2 \quad (50)$$

In alkaline conditions, the ozone was as well described to be decomposed into superoxide ions according to the following reactions (Eq. 51–54) [87]:

$$H_2O + O_3 \rightarrow 2HO^{\bullet} + O_2 \quad (51)$$

$$OH + O_3 \rightarrow O_2^{-} + HO_2^{\bullet} \quad (52)$$

$$OH + O_3 \rightarrow O_2^{-} + H^{+} \quad (53)$$

$$O_3 + HO_2^{\bullet} \rightarrow HO^{\bullet} + 2O_2 \quad (54)$$

Moreover, many works have reported a high degradation rate in acidic conditions for different pollutants, such as dyes and phenols [88–91]. Under acidic condition the ozone is available as molecular ozone and with increasing pH from acidic to alkaline the oxidizing potential of ozone decreased from 2.08 V to 1.4 V [92]. Also, there is a possible consumption of hydroxyls radicals by reacting with; i) carbonate ions [89] and ii) superoxide ions at increasing pH [93].

4. Discharge plasma types

Different criteria can be found in the classification of discharge plasmas: i) according to the gas temperature, ii) electron and ion temperatures and plasma density, iii) operating pressure (Equilibrium plasmas at high pressure $\geq 10^5$ Pa and non-equilibrium plasmas at atmospheric pressure or lower) and current magnitude, iv) powering mode, v) thermodynamic equilibrium (Equilibrium or thermal and non-equilibrium or non-thermal plasmas) and ionization degree (= the density of charged particles/the total density of species including neutral and charged particles, partially ionized (10^{-4}-10^{-6}) to completely ionized (e.g., stars and visible interstellar matter) [94].

Plasma is a partially ionized gas, which can be generated by various electrical discharges and can be classified into two main groups, in other words, thermal and non-thermal, in terms of electronic density, pressure, or temperature. Thermal plasma (usually arc discharges, torches, or radiofrequency) is accompanied by enough energy added to permit plasma elements to be in thermal equilibrium ($T_e \approx T_{gas}$). Whereas non-thermal plasma is attained using less power when an energetic electron temperature much higher than that of the bulk-gas molecules (Fig. 5).

Different technologies, in the discharge plasma, based on high-voltage electrical discharges directly in the water (electrohydraulic discharge) or in the gas phase overhead the water (non-thermal plasma) has been reported in the literature, such as pulsed corona discharges [95,96], dielectric barrier discharges [19], gliding arc [97] and contact glow discharge electrolysis [55,98].

Such mechanisms remain less understood, hence the inferences from various studies available in the literature in certain aspects are contradictory owing to a wide variety of the investigated systems and the involved process variables. Owing to these reasons, the comparison of the discharge reactors' performance considered in proportion to their configurations is not possible, which given their great diversity, would have been interesting. Although the experimental conditions are different in various studies, the results could be discussed based on the energy efficiency (yield) of pollutant removal, and many researchers have used energy efficiency to compare their results to other researchers.

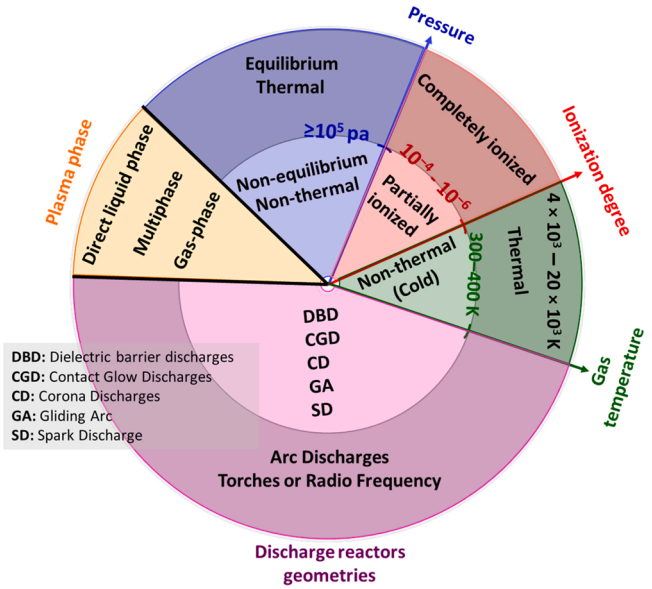

Fig. 5. Different classification of discharges plasma.

The Table 1 summarizes an inexhaustive list about types of plasma applied for pollutant removal as well as operating conditions. A variety of different reactors with liquid phase classified according to the discharge type (DBD, Corona and contact glow discharge) have been developed and tested for the removal of harmful organic compound from water. It is easy to note a great disparity of the removal efficiency and the treatment time for the same compound from a reactor configuration to another whether in the same discharge type or a different one. Also, a wide range of concentrations for treated pollutants have been reported from a few milligrams to hundreds or even thousands of milligrams.

4.1. *Dielectric barrier discharge (DBD)*

This micro-plasma category, which is also called silent discharges, was developed for the first time for ozone generation and then generalized in different applications, such as surface treatment [30,113] and gas purification. In this kind of system, the electrical discharge generally occurs between electrodes, with the condition that a thin layer of dielectric material covers at least one of the electrodes (e.g., quartz, glass, ceramics, mica, and alumina) or one of these materials is placed in the discharge gap [29]. The aqueous solution can also act as the second electrode in the DBD system [30].

Several works underlined the importance of ozone generation during recalcitrant compound degradation because of its long lifetime, compared to the other oxidizing species [27].

This type is classified as cold plasma formed at low/atmospheric pressure systems; it has the advantage of the low operating cost that is ascribed to limited consumption of power and gas. Interestingly, in the past years, various DBD reactors geometries with different electrode configuration were proposed for the treatment of multiple wastewaters [19,114,115]. That emphasizes the fact that water treatment is attracting more attention from the researchers.

4.2. *Contact glow discharges*

Contact glow discharge electrolysis (CGDE) is an unconventional process in the advanced oxidation process (AOPs), where electrochemical reactions take place midst of an electronic conductor (electrodes) and the surrounding ionic conductor surface (electrolyte). This system induces charge transfer across the interface between the electrodes and the electrolyte if the applied voltage is sufficiently high enough [116,117]. Various types of electrolytes have been described in the literature, aqueous electrolytes [118], solid electrolytes [119], and molten salts or ionic liquids [120,121].

In the contact glow discharge electrolysis, the cathode is immersed in water and isolated from the anode using the porous glass. A continuous DC voltage is exercised to a thin wire anode communicating with the water surface [55]. In this type of plasma, the temperature of almost species in the discharge zone (anions, cations, and neutral species) is increased. They are produced with much higher yields than in conventional electrolysis in both gas phase and aqueous solution [122]; so, the plasma generated in the reactors can be called hot plasma [123]. In such a system, H_2O_2 is the main species formed in the liquid phase, and H_2 is formed in the gas phase.

4.3. *Corona discharges*

Pulsed corona discharge is usually generated in a needle to plate electrode configuration (e.g., tips, pinpoints, or thin wires), where a needle is connected to the high-voltage terminal, and the plate is earthed [124,55]. The electrodes materials may be made from copper, ceramic-coated stainless-steel, titanium, etc. [125–127].

Table 1
Some hazardous chemicals and electrical discharge type and reactors

Electrical discharge Reactor	Treated pollutant (Conc.)	Degradation efficiency	Time needed	references
DBD discharges				
A novel pilot two-level dielectric barrier discharge reactor	Methyl orange (240 mg/l)	91.0 %	130 min	[19]
Gas phase surface discharge plasma	Dimethyl phthalate (30 mg/l)	92.1 %	30 min	[44]
DBD plasma combined with TiO2/ activated carbon fibers	Triclocarban (10 mg/l)	89.0 %	30 min	[57]
DBD plasma plume	Methyl violet 5BN (22,4 mg/l)	≈99 %	≈30 min	[30]
DBD Batch plasma reactor.	Atrazine (5 mg/l) Chlorfenvinfos (5 mg/l) 2,4-Dibromophenol (1 mg/l) Lindane (1 mg/l)	93.2 % 94.3 % 98.0 % 86.6 %	5 min 5 min 5 min 5 min	[99]
DBD Coaxial plasma reactor	Atrazine (5 mg/l) Chlorfenvinfos (5 mg/l) 2,4-Dibromophenol (1 mg/l) Lindane (1 mg/l)	40.3 % 93.6 % 73.5 % 79.2 %	5 min 5 min 5 min 5 min	[99]
Ex situ level dielectric barrier discharge reactor	Carbamazepine (20 mg/l)	100 %	5 min	[100]
Parallel plane type coaxial NTP-DBD reactor	Crystal violet (100 mg/l)	90.6 %	25 min	[77]
Water falling film dielectric barrier discharge	Triallyl isocyanurate (1000 mg/l)	100 %	60 min	[78]
Novel two-level dielectric barrier discharge reactor	Methyl orange (80 mg/l)	94.1 %	80 min	[19]
DBD - rotating drum reactor	Carbamazepine (23.6 mg/l)	94.0 %	60 min	[101]
Corona discharges				
Gas discharge circulatory airtight reactor system (GDC)	Methyl orange (60 mg/l)	93.7 %	20 min	[67]
Isothermal batch reactor with continuous gas bubbling	Phenol (100 mg/l)	62.7 %	180 min	[2]
A multi-needle-to-plate pulsed discharge plasma reactor	Polyvinyl alcohol (324 mg/l)	76.0 %	60 min	[102]
a dual pin-to-plate high-voltage corona discharge	Acid Blue 25 dye (10 mg/l)	100 %	35 min	[103]
Corona discharge plasma coupled with natural soil particles (PCDP/SPs)	Tetracycline hydrochloride (50 mg/l)	82.64 %	60 min	[104]
Point-to-plate corona with gas bubbling	Tetracycline (50 mg/l)	61.9 %	24 min	[105]
Corona with liquid shower	Paracetamol (100 mg/l)	100 %	20 min	[106]
Corona above water	Diclofenac (50 mg/l)	100 %	15 min	[107]
Pulsed electrical discharge assisted with modified activated carbon fibers	Methyl orange (100 mg/l)	100 %	≈30 min	[108]
pulsed discharge plasma coupled with MWCNTs-TiO2/ δ-Al2O3	Acid orange II (50 mg/l)	100 %	60 min	[109]
Contact glow discharge				
Contact glow discharge electrolysis (CGDE)	Acid brilliant red B (16 mg/l) Acid flavine G (16 mg/l)	94.99 % 95.55 %	2 min 2 min	[98]
Contact Glow Discharge Electrolysis (CGDE)	Acid Orange 7 (25 mg/l)	95.00 %	90 min	[110]
Contact glow discharge electrolysis (CGDE)	Cationic Blue SD-GTL (25 mg/l)	99.7 %	3 min	[111]
Glow Discharges Generated in Contact with Flowing Liquid Cathode	Methyl red (30 mg/l) Triton X-45 (150 mg/l)	99.0 % 100 %	Not mentioned	[112]

The pulseless in the corona system can generate a high flux of electrons from DC electrical discharge in gas or liquid media. Then a large amount of oxidizing chemical species are produced. The discharge in water with air or oxygen bubbling can produce H_2O_2 and O_3 as the main chemically active molecules [3,38].

As reported in different works, the relative density of ˙OH, H and O ¨generated depends on the nature of bubbled gas [66,128]. The DC corona has the advantage of continuous radical species generation and the disadvantage of high energy consumption as compared to the pulsed electrical corona because of the continuous operation [7].

5. Reactor configuration

In the use of discharge plasma to treat contaminants, there are different influencing parameters; among them, the reactor geometry is a crucial factor. Malik et al. compared the relative energy generates about 27 different types of plasma reactors [129]. The results of energy yield obtained from the degradation of organic compounds in water were found to follow the order: 1) pulsed DC < continuous DC or AC power, 2) pulsed corona discharges < pulsed dielectric barrier discharges < DC discharges, 3) burning plasma in oxygen (Bubbling with oxygen) < air < liquid, 4) treated solution as fine droplets in the plasma zone < thin film < deep layer [129].

A variety of reactor configurations were recently reported in different works; among them, plate to plate DBD to treat liquid film falling by gravity showed the best result [130]. This prototype reactor can be coupled with TiO_2, which drastically enhances the mineralization yield of pollutants and reduces energy consumption. Moreover, in the DBD system, a cascade of two DBD plasma systems can be used in series, separated by an interface in the center (used as supporting gas introduction region), to imply changes in plasma chemistry characteristics and the radical's formation [31].

Hijosa-Valsero et al. reported that the energy efficiency of the DBD coaxial thin-falling-water-film reactor (Fig.6) is higher than a DBD conventional batch reactor. However, the degradation of kinetic is slightly higher in the last reactor configuration [131]. Dojcinovic et al. reported a similar reactor configuration for the degradation of four reactive textile dyes (Reactive Black 5, Reactive Blue 52, Reactive Yellow 125, and Reactive Green 15). The study revealed that the addition of H_2O_2 and Fe^{2+} in the dye solution enhanced the decolonization efficiency at the optimized conditions of applied energy density, pH, and residence time [132].

Tao et al. proposed a novel configuration of a DBD reactor. This continuous pilot DBD system is composed of two co-axial quartz tubes and a stainless-steel disk electrode and one stainless steel mesh electrode (Fig. 7). The wastewater is pumped up through the inner quartz tube to the top of the tube, then falling in the annular region between the two tubes. Thus, two distinct discharges are generated: i) between the stainless-steel disk electrodes and ii) between the steel disk electrode in the water and the stainless-steel mesh electrode. Therefore, these discharges lead to efficient degradation of high concentrated methyl orange solution [19]. A similar reactor, "falling film dielectric barrier discharge," was proposed by Rong and Sun, where the treated solution is directly pumped to the top of the reactor via an external canal [78].

Wang et al. reported in their work for the degradation of dimethyl phthalate a surface discharge plasma reactor in which the plasma was

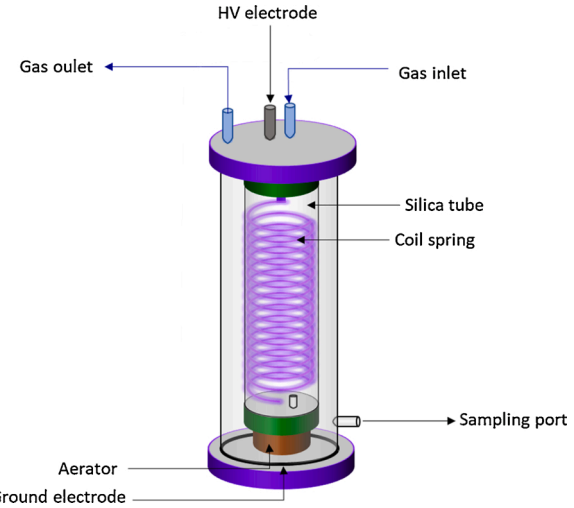

Fig. 6. DBD coaxial thin film water falling surface discharge plasma reactor.

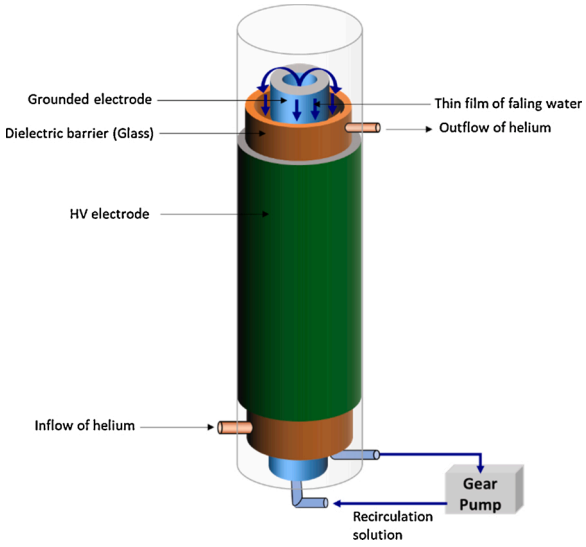

Fig. 7. Two-level dielectric barrier discharge reactor.

initiated in the gaseous environment. Then the generated actives species were transfused quickly into the wastewater [44] This kind of reactor was first developed by Ogata et al. for VOCs treatment [133], and later Li et al. has used this type of reactor for treatment and sterilization of water, wastewater [134,135].

Some reactor geometries, developed by researchers, can have many tasks at the same time. For example, the DBD plasma plume array developed for textile modification by Chen et al., could be used for both fabric-fiber surface treatment and aqueous textile effluent decontamination by using discharge plasma produced by-products such as radicals, ozone, and ions [30].

Jiang et al. have compared the following four prototype plasma reactors for the treatment of dye solution under similar operating conditions: (i) batch reactor with a needle–plate electrodes (BR); (ii) only liquid circulating system (OLC) with gas directly flowing outside from discharge reactor; (iii) hybrid gas-liquid electrical discharge (GL) and ; (iv) gas discharge circulatory airtight reactor system (GDC). They found that the pollutant degradation rate followed the following trend; GDC > GL > OLC > BR. However, interestingly the energy efficiency of pollutant removal decreased in the following order: GDC > OLC > GL > BR [67].

Liu et al. studied the removal of Carbamazepine with two electrical discharge modes, ex-situ and in situ, directly on the water surface. The results showed very high efficiency for the ex-situ mode [136]. This result can be explained by the formation of NOx in-situ mode that leads to a decrease in the pH and their competition with produced ozone.

Besides, scaling up the discharge plasma reactor can enhance the efficiency of the process. Indeed, the improvement of the mass transfer of the reactive species and the interaction between the plasma and solution reduces energy consumption [137].

In 2017, Ajo et al. studied the hydroxyl radical transfer through the Gas-Liquid Interface utilizing a pulsed corona discharge (PCD) reactor, with water sprayed into the plasma zone, through an atomizer array [138]. The vertical wire-plate designed reactor is equipped with an adjustable pulse generator to deliver identical pulses. Water was sprayed, using five axial-flow full cone atomizer nozzles, from the top into direct contact with the plasma, and the treated solution is withdrawn from the bottom in holding tank (Fig. 8). The results reported that the $^{\bullet}OH$ radicals are yielded at the interface on the gas side (i.e., from water vapor), and not in the liquid phase. Furthermore, the gas-liquid interface is a significant barricade for efficient oxidation of aqueous compounds by $^{\bullet}OH$.

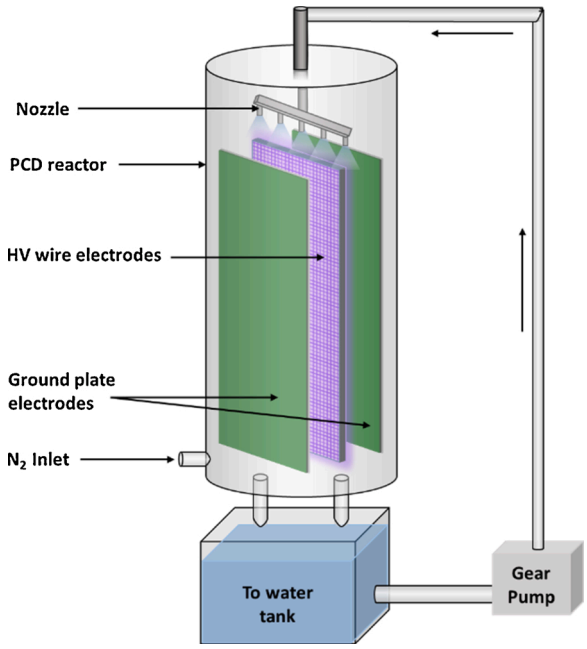

Fig. 8. Pulsed Corona Discharge (PCD) reactor.

6. Plasma in combined processes

Discharges plasma are fast and efficient technique for the treatment of the different family of pollutants. The NTP DBD discharge could mineralize, partially or entirely, the contaminants. Nevertheless, the degradation mechanism is not unique; indeed, it improves the biodegradability of the organic pollutants [139]. This technique can be used alone or coupled with another process such as: photocatalysis [56,57], biodegradation [29], catalytic effects of activated carbon fibers [108], adsorbents, catalysts, ultrasound, ozone and other oxidants [140].

The combined DBD-plate plasma and suspended-TiO2 photocatalysis could significantly increase the mineralization yield as compared to the DBD-plate plasma system alone, as reported by Ghezzar et al. [56]. Vanraes et al. combined the DBD plasma with the adsorption process on activated carbon textile with additional ozonation as post-treatment or pretreatment. It was found that the configuration with ozonation, as pretreatment, leads to enhanced pesticide removal and shows better efficiency than the second configuration (plasma treatment first) [141]. Li et al. reported the degradation of organic pollutants coupling discharge plasma and in-situ hybrid photocatalytic process. In this technique, authors have used discharge plasma to produce active species on the TiO_2 surface and also used external UV light to enhance the process efficiency [142].

It was reported that the treatment of contaminated aqueous solutions with a combined corona discharge-ozonation system increased the rate of degradation and also significantly improved the mineralization yield as compared to ozonation alone [143]. The combined DBD plasma-Activated carbon fibers (ACFs), compared to DBD plasma alone, showed an enhanced degradation efficiency and mineralization yield at low pH. This synergistic effect in the presence of ACFs can be explained by a high adsorption capacity, stable discharge, and small discharge gap [144].

The synergetic degradation performance of Acid Orange 7 (AO7) by dielectric barrier discharge plasma (DBDP) and persulfate (PS) was examined by Shang et al. the addition of persulfate during treatment had little effect on the degradation pathways of dye, but significantly improved the mineralization, the authors mainly attribute this effect to the enhanced production of sulfate radical and hydroxyl radicals produced from the activated PS [74].

Shang et al. reported the degradation of p-nitrophenol (PNP) by DBD plasma / Fe^{2+}/ persulfate oxidation process. It was found that the tertiary system of plasma/PS/Fe^{2+} with an optimal quantity of PS and Fe^{2+} presented higher PNP degradation efficiency for the same time of treatment than discharge plasma alone and the binary systems of plasma/PS, plasma/Fe^{2+} and PS/Fe^{2+} [145].

7. Advantages and drawbacks in discharge plasma for water treatment

In recent years, extensive attention has been drawn to water treatment using plasma techniques, as it has many advantages such as:

i) Importantly, no external oxidants are required, and no need to adjust the temperature and pH of the treated effluents. Therefore, it can be discharged directly, or it can be used in the irrigation field [146].

ii) High-efficiency process

iii) The reactors used to generate plasma in the gas phase can be used for water treatment with small modifications, since similar kind of active species are produced in the gas and the liquid phase [147].

Indeed, discharge plasmas have several advantages, as mentioned above, it also has some drawback such as;

1) Possible generation of highly toxics degradation by-products [148].
2) A decline in the efficiency of increasing concentrations of organic pollutants and mineral salts [132].
3) In some plasma technologies, the price and the high gas consumption present a severe limitation for a large-scale application [132].

8. Plasma treatment in pilot scale

Nowadays, there is still a need to develop systems with pilot or industrial-scale reactors that have an excellent capacity for resolving a real problem of chemically and biologically contaminated water [149]. Such as the system proposed by Fröhling et al. in the study of the Impact of a Pilot-Scale Plasma-Assisted Washing Process on the Culturable Microbial Community [150]. The development of the Pilot-Scale Pulsed Corona Discharge system by Ajo et al., for the removal of high concentrated pharmaceutical residues gives a reduction of 87% of this last from raw sewage. However, 100% elimination was obtained for wastewater treated biologically with less energy consumption [151]. Some researchers team has proposed the scaling-up of a specific plasma process utilizing a microwave plasma torch, where this system can generate antimicrobial active water at a near industrial scale. The process was competitive with industrial habitual chemical proceedings [152]. On the other hand, this technology has shown high efficacy in the degradation of trace organic compounds such as pharmaceuticals at a pilot-scale [153], or for the degradation of wide concentration range (~10–10^5 ng/L) of poly- and perfluoroalkyl species from liquid investigation-derived waste [154]. A supplementary ozone injection in pilot scale plasma system for some organic pollutants removal has proved to be more efficient [155]. However, the entire investment costs and reliability of this reactors at a large-scale are still relatively ambiguous. Also, given that molecular ozone oxidation may be the dominant mechanism for many pollutants, future studies could provide direct comparisons of electrical discharge plasma with traditional ozone generators to compare process performance. From another hand, in order to enhance its commercial viability for full-scale water and wastewater treatment, plasma technology could undergo a long-term reliability test on its desired performance durability and normal working life as well as operational safety.

9. Conclusions

In the last years, the demand for the application of electrical discharge plasma technology for raw water and industrial wastewater cleaning has significantly increased. The medium of discharge considerably affects both the reactive species production and the degradation

reaction mechanism. In the plasma reactor, the efficiency is controlled by the discharge plasma type, the physical state of the contaminated solution and its introduction system, the internal conception, and the scale of the reactor. The discharge plasma process can be combined with different processes of treatment, and the yield depends on the type of the coupled process, the configuration of the two processes in post or pre-treatment arrangement, and the number of passes. As for all treatment techniques, the discharge plasma has many advantages for water cleaning but also presents some drawbacks.

Plasma technologies combined with other processes seem to be practical, inexpensive, effective and forward-looking methods for the removal of aqueous contaminants at both laboratory and pilot scales. But, looking forward, several challenges remain to be addressed for more research and development in order to satisfy consumer demand. Future use of plasma discharge for water treatment will primarily depend on its efficacy and energy consumption relative to other advanced oxidation process and treatment methods in general, but certain requirements must also be taken into account. Sustainability, ease of use, capital and operating costs, gas supply will all decide when the device can be implemented on a wide scale. In addition, a thorough analysis of the produced oxidation by-products and long-lived oxidants in treated water is required to ensure that total toxicity is continuously and adequately decreased after treatment.

The novelty of this study " Review on Plasma discharge for water treatment: mechanism, reactor geometries, active species and combined processes"

This manuscript presents a review on a serious environmental problem (contamination of the water).

In our knowledge, paper on plasma reactors configurations and actives species for water remediation presents a new review for Journal of Water Process Engineering readers.

These summarize of findings (over more than 150 ref) would certainly be of great interest to the readership of Journal of Water Process Engineering for advancing in their investigation about nonthermal plasma for environmental applications.

Declaration of Competing Interest

The authors report no declarations of interest.

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