



Disinfecting Water: Plasma Discharge for Removing Coronaviruses

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Abstract

At COVID-19 time, viruses in water become gravely dangerous to human health and life and very resistant to traditional disinfection methods. As a type of encouraging endeavor for contamination removal, plasma discharge shows good results in dealing with viruses' removal. Indeed, more efficient, cheaper, and environmentally-friendly than conventional disinfection techniques, electrical discharge technologies are confirmed as. UV emission from plasma dispositions and the impacts of irradiation on microorganisms become broadly studied. Throughout ozonation, implementing pulsed high-voltages can lead to better diffusion of ozone in water and quicker transformation of ozone into free radicals. Via direct electrical discharges, purifying water has trends to be examined on a large-scale. Both in water and above water level, the electrical discharges possess their advantages and disadvantages. Above water level, which is in the gas phase, electrical discharges need less energy for the discharge to occur; however, in water, electrical discharges need an easier setup and form the chemically active species that could immediately bombard the aqueous contaminants. One of the kinds of electrical discharges, pulsed corona discharge remains the most tried and looks to be the most encouraging for treating water. Such methods could be methodically experimented with determining the optimal circumstances for killing COVID-19 and different pathogens from water. Merging plasma discharge, electrocoagulation, and magnetic field implementation can lead to better performances. As a secure physical separation, the final step has to involve activated carbon adsorption pursued by a membrane process to retain organic matter liberated from the cellular cytoplasm throughout oxidation and disinfection methods.

Subject Areas

Chemical Engineering & Technology, Infectious Diseases, Public Health

Keywords

Coronaviruses, COVID-19, Corona Discharge, Dielectric-Barrier Discharges (DBDs), UV Radiation, Disinfection

1. Introduction

As well-known as barrier discharges or silent discharges, dielectric-barrier discharges (DBDs) have for a long period been seen as the ozonizer discharge [1]. In 1932, Buss [2] discovered that in a plane parallel gap with insulated electrodes, air breakdown takes place in a number of individual tiny breakdown channels. Lately, it was seen that plasma could be affected, modeled and optimized for a specific utilization via micro-discharges [3] [4] [5]. The most remarkable feature of DBDs remains that non-equilibrium plasma circumstances could be furnished in a much easier manner than with other options such as low-pressure discharges, fast pulsed high-pressure discharges or electron beam injection. Further, the DBDs could breakdown most of the gases at around atmospheric pressure in a big number of independent current filaments or micro-discharges. The dielectric barrier restricts the quantity of charge and energy deposited in a micro-discharge and distributes the micro-discharges across the full electrode surface. At what time a DBD is run in rare gases or a rare gas halogen mixture, plasma circumstances in a micro-discharge channel are identical to those in pulsed excimer lasers. As a result, each micro-discharge could operate like an intense source of ultraviolet (UV) or vacuum ultraviolet (VUV) radiation. The absorption coefficient of most substances augments at shorter wavelengths. Thus, in several conditions, the UV radiation is absorbed in a very thin surface layer. The xenon excimer lamp can form photo-cleavage of water and oxygen [1] [4].

In 1979, Donohoe and Wydeven [6] acquired a uniform glow discharge with pulsed excitation in a helium/ethylene mixture from which the term *atmospheric pressure glow* (APG) was originated. In 1956, for producing uniform glow discharges at atmospheric pressure in helium, air, argon, oxygen, and nitrogen via 50 Hz power source, Gambling and Edels [7] employed an electrode arrangement comprising two metal foils masked with a particular metal mesh and ceramic plates [1]. Electret dielectrics could collect charges on the surface, and are trapped uniformly on the surface through the applied voltage. The charge carriers are ejected spontaneously from the surface conducting to homogeneous discharge because of the alteration in the polarity of the electric field surpassing the threshold value [1].

Using gas plasmas efficiently demobilized microbes caught the attention of

several scientists [1]. It is established that there are abundant charged particles, chemically reactive species, and UV photons in the plasma discharge, all of which could provoke harm to cells, so attaining demobilization or alteration [3] [8]. Biologically, biochemical effects are noticed at what time microbes are revealed to plasmas, for instance, 1) protein denaturalization, 2) enzyme deactivation and 3) deoxyribonucleic acid (DNA) mutation [1].

It is known that atmospheric pressure plasma discharge possesses a huge influence on *Escherichia coli* thanks to the deterioration of the cell membrane that conducts to cell lysis [7]. Until 2015, Paunekar *et al.* [1] affirmed that they are incapable to assess the impact of plasma treatment on the bio-macromolecules such as cell wall and membrane, made up of polysaccharides and membrane-bound proteins. In order to comprehend the pathway of plasma treatment, it is crucial to study the phenomenon of cell lysis and death, and also the mutation [1].

This work examines the chronicle of DBDs, kinds of disinfection techniques, the necessity for substitutional disinfection process (thermal plasma and non-thermal plasma), proposed theories for adequate clarification of pathways supporting such a discharge process for the induction of corona discharges in water, impact of sterilization employing DBD setup and impacts of DBD plasma, water purification via electrical discharges, ozone for treating water, and arc discharge-mediated disassembly for killing viruses.

2. Chronicle of Dielectric-Barrier Discharges (DBDs)

In 1857, Siemens [9] first proposed DBDs for the objective of ozone production in the air; however, in 1778, Lichtenberg [10] performed its first test. At the commencement of the 20th Century, investigations were conducted leading to a better comprehension of the DBD and diverse utilizations [1].

Relating to public health issues (like multi- and extensive drug-resistant microbes, bioterrorism, etc.), there is a demanding necessity for reinforced endeavors to avoid transmission of infections employing ecological controls [1] [11] [12] [13]. Because of this, ultraviolet germicidal irradiation (UVGI) has attracted large regard. Some interrogations concerning performance and security restrict the implementation of UV founded techniques for disinfecting, even with that UVGI could be secure and greatly efficient in disinfecting the air, water, and surfaces that way avoiding transmission of a set of infections. Right now, low-pressure mercury (Hg) discharge lamps are utilized in UVGI implementations and release shortwave ultraviolet-C (UV-C, 100 - 280 nm) [14] radiation, mostly at 254 nm. Nevertheless, mercury-based lamps may play a part in heavy metal contamination and linked health risks [1].

Downes and Blunt [15] established that the potential of sunlight to demobilize microbes was a function of intensity, time, and wavelength, with the shorter wavelengths of the solar spectrum being the most performant [1]. UV-C radiation demobilizes microorganisms via destroying their DNA. Many types of research

were dedicated to locating the wavelength dependence of the germicidal work of light [16]. The main manner of demobilization happens at the time that the absorption of a photon generates pyrimidine dimers among adjacent thymine bases and makes the microorganism unable of replicating [1].

Paunekar *et al.* [1] presented a list of the main steps throughout history for developing VUV/UV DBDs and their corresponding efficiencies in killing pathogens (Table 1).

3. Kinds of Disinfection Techniques

In terms of water disinfection engineering, numerous conventional sterilization techniques (like thermal sterilization, chemical sterilization, and irradiation of UV and gamma rays) become currently utilized on a large scale. However, throughout recent years, plasma-founded apparatuses have been valued for biological sterilization [1]. Such devices possess a large extent for utilization relative to traditional techniques. Irradiation of UV and gamma rays produces energetic photons which could occasion grave harm to DNA and are toxic for human being [1]. Nevertheless, most of the classical sterilization manners are time-consuming and are run in closed space. The plasma-founded setups could be implemented to open space, possess a very short sterilization period (1 min), produce several reactive species (such as ozone, hydroxyl radical, and oxygen atom) and possess a great benefit over different sterilization devices [43]. The physical technique performs via positive and negative ions in the discharge's streamer, and the chemical process is realized through ozone, atomic oxygen, hydroxyl radical, etc. [1] [44] [45] [46].

Some of the classically applied sterilization techniques for water treatment involve [47] [48] [49] [50]: boiling, chlorine [47] [51] [52] [53] [54], chlorine dioxide, dry heat sterilization, ozone [55], UV light [14], etc. [1].

4. Necessity for Substitutional Disinfection Process

Recently, the engineering of killing pathogens employing non-thermal plasma formed via gas discharge at atmospheric pressure has attracted considerable interest [56]. Conventional disinfection and sterilization technologies possess numerous disadvantages [56]. Such techniques require extended treatment periods. Numerous polymer-founded tools and several very expensive equipments (like endoscopes) could not be disinfected via autoclaving. Virulent substances like formaldehyde, ethylene oxide, and glutaric dialdehyde not only hurt the human body but as well as contaminate nature [1].

In the 1960s, surveys on plasma sterilization begun. Following this period, large investigations have been performed on plasma sterilization. As juxtaposed to incineration and thermal plasma treatment, the merit of utilizing a non-equilibrium discharge is that most of the discharge energy could be employed to speed electrons and produce free radicals [43]. Moreover, DBDs possess benefits over classical techniques if numerous contaminants should be handled at the same instant. DBDs are adopted particularly when contaminant levels

Table 1. Chronicle of R&D of VUV/UV DBD and their disinfecting performance [1].

Year	Accomplishment
1885	Duclaux [1] noted dissimilarities in sensitivity to sunlight among diverse species of bacterial spores.
1892	Geisler [1] demonstrated that UV radiation from sunlight and electric lamps was more efficacious in neutralizing microbes than longer wavelength radiation; nevertheless, he as well observed that the deadly impacts of longer wavelength radiation were boosted at augmented intensities.
1904-1905	Hertel <i>et al.</i> [1] were the premier to illustrate the mutagenic impacts of UV radiation.
1929	Gates [1] presented the premier analytical bactericidal action spectrum. Utilizing a mercury arc lamp, he generated the same shaped action spectra for <i>Staphylococcus aureus</i> and <i>Bacillus coli</i> , both with peak performance at 265 nm.
1930	Gates [1] announced an analytical bactericidal action spectrum with peak performance at 265 nm, very near to the 254 nm output of low-pressure Hg germicidal lamps.
1935	Wells and Fair [1] established that airborne infectious organisms can be efficiently destroyed in a short time employing aerosolized <i>E. coli</i> at 254 nm radiation in commanded circumstances.
1934-1955	Wells [1] suggested the idea of airborne infection via “droplet nuclei”—evaporated droplets carrying infectious microbes that could stay suspended in the air for prolonged times. Wells and Fair [1] established the capability of UVGI to efficaciously demobilize airborne microbes and demonstrated the notion of infection through the airborne pathway. They utilized upper-room UVGI to avoid the epidemic diffusion of measles. Overholt and Betts [17] widen the implementation of UVGI in hospitals by applying many dispositions of cubicle-like UVGI “light curtains” conceived to avert respiratory cross-infections. Whisler <i>et al.</i> [18] estimated the influence of physical and ecological parameters on UVGI performance, comprising humidity and air circulation—two key variables in the effectiveness of UVGI. The Council on Physical Therapy [19] agreed on UVGI for disinfecting targets. Hollaender and Oliphant [20] declared that the high UV Germicidal Irradiation for Air Disinfection vulnerability of several agents at around 260 nm is founded on the essential work of DNA in biological actions of organisms.
1957-1976	Riley <i>et al.</i> [21] revealed Guinea pigs to air emerging from an occupied tuberculosis (TB) ward and established that TB is diffused through the airborne pathway [22] [23]. Riley <i>et al.</i> [24] established that virulent tubercle bacilli and <i>Bacillus Calmette-Guérin</i> (BCG) are in the same manner vulnerable to UVGI and quantified the vanishing rate of aerosolized BCG in a model room with and without upper-room UVGI. Further, UVGI efficiently demobilized <i>E. coli</i> in the ward and stopped rabbits from developing TB. On the contrary, revealed rabbits were infected with TB without employing UVGI. Investigations have emphasized both that TB can easily be diffused via droplet nuclei and that UVGI can enough demobilize the infected air [25] [26] [27]. Beukers and Berends [28] revealed frozen solutions of thymine to UV-C radiation leading to the generation of thymine dimmers. McLean [29] blocked the diffusion of influenza in Veterans Hospital TB patients utilizing upper-room UVGI throughout the 1957 pandemic, presenting testimony for the airborne transmission of influenza. Riley <i>et al.</i> [21] examined the influences on disinfection rates in the lower room from air mixing via convection and a ceiling fan and mathematically modeled it. Riley and Kaufman [25] [27] followed the impact of relative humidity (RH) on the performance of UVGI, with an acute slop observed in the portion of organisms neutralized at RH estimates bigger than 60% to 70%. UV-C wavelengths are the most biologically energetic radiation and, ironically, much less hazardous to human beings. This is due to the fact that UV-C radiation is absorbed by the outer dead layer of human skin, while UV-B and UV-A radiation infiltrate deeper. The contrast has to be performed among the biological effect and the infiltration depth of UV radiation, a fundamental notion in UVGI security in the direction of quantitatively assessing UVGI performance and integrity actions for the appropriate employment of UVGI [30]-[35].
1985-1992	Investigations estimating different ecological and physical variables on UVGI efficiency (like air mixing and ventilation, humidity, microbial vulnerability, fixture irradiance and configuration, and photo-reactivation) were performed [36].
2001	The dielectric barrier has a crucial contribution in prevention of arcing and in the so-called non-thermal excitation of the plasma. Further, DBD produces uniform discharge plasma at atmospheric pressure [37].
2004	Plasma-based apparatuses have been assessed for biological sterilization. Cooper <i>et al.</i> [38] focused on the impact of plasma on <i>Bacillus stratosphericus</i> in three viability states (<i>i.e.</i> , viable, cultivable at low plasma dose, and viable but non-cultivable (VBNC) at high plasma dose). <i>B. stratosphericus</i> possesses the capacity to turn into VBNC across plasma implementation. Yating <i>et al.</i> [39] examined the influence of atmospheric pressure non-equilibrium plasmas (APNPs) on <i>N. gonorrhoeae</i> . APNPs are apt to efficiently and rapidly neutralize the <i>N. gonorrhoeae</i> ; further, the neutralizing impact is linked to the structural deterioration of the cell membrane. Employing non-thermal plasmas for disinfecting multidrug-resistant microorganisms such as <i>S. aureus</i> , <i>Pseudomonas aeruginosa</i> , and <i>Candida albicans</i> in environmental settings and substantiate ongoing clinical applications for plasma devices. Maisch <i>et al.</i> [40] assessed the influence of cold atmospheric plasma for numerous time periods or UVC radiation doses on <i>D. radiodurans</i> . They found <i>D. radiodurans</i> sensible to the cold atmospheric plasma treatment, identical to the methicillin-resistant <i>Staphylococcus aureus</i> (MRSA) strain. Conversely, <i>D. radiodurans</i> was more resistant than MRSA to UVC radiation treatment. Using cold plasma, Pan <i>et al.</i> [41] killed <i>E. faecalis in vitro</i> biofilms in dental root canal treatment, and Xu <i>et al.</i> [42] eliminated yeast cells in water.
2010-2020	

are small, *i.e.*, in the 10 - 1000 ppm span. Utilizing DBD sources, the degradation of greenhouse gases (CO and CH) below diverse running circumstances is examined across a large temperature and pressure extent. Further, the DBD remediation of big parts is made possible at atmospheric pressure. Plasma could be categorized in capacitively coupled plasmas (CCPs) and inductively coupled plasmas (ICP) because of the power input. Two sorts of atmospheric plasma utilized for biological sterilization comprise thermal and non-thermal plasma [1].

4.1. Thermal Plasma

In thermal plasma, energy flux from electrons to heavy particles balances the energy flux from heavy particles to the environment only when the temperature of heavy particles becomes almost equal to the electron temperature. Employing thermal plasma is restricted due to its elevated temperature (2000 K up to 10,000 K). Such a temperature could burn and harm the tissue. In the case of contact, glow discharges almost all the species in the discharge zone, *i.e.* anions, cations, and neutrals, heat up; therefore, the plasma produced in the devices could be named *hot plasma* [1].

4.2. Non-Thermal Plasma

Non-thermal plasma runs at ambient temperature. Its elevated performance and security are convenient for medical and biological aims. DBD stays one of the rapid and credible non-thermal plasma that is employed largely for sterilization. In silent discharges, pulsed corona discharges, only free electrons acquire elevated energy and the residue of the heavier charges and neutrals stay close to room temperature, and the plasma so formed is named *cold plasma* or *non-equilibrium plasma* [1] [57] [58].

Actually, chlorine is being re-estimated as the standard for disinfecting potable water and wastewater [47] [53] [54]. However, because of the price of hypochlorite fabrication and its possible carcinogenic and mutagenic influences (disinfection by-products, DBPs) [59] [60] [61] [62] [63] on aquatic species, its usage stays also restricted. Sterilizing or demobilizing pathogens is indispensable for the most vital domains like medicine, food industry [64], and agriculture. Lately, considerable regard has been accorded to the electrode composition and to the usage circumstances at which non-thermal plasma could efficiently kill microbes. As a rule, it adopted that the electric field and plasma products (UV radiation, charged particles, and reactive oxygen species (ROSs) [65] [66] [67] [68]) are the bactericidal agents. Such species in plasma are extremely complicated and the bacterial demobilization employing non-thermal plasma is linked to numerous themes, like plasma physics, biology, medicine, and disinfection; thus, it stays mostly obscure of what pathways atmospheric-gas plasmas worked in killing microbes [1].

At the speed of light, UV disinfection was adopted as an option to chlorination of wastewater effluents [14] [65] [69] [70] [71] [72]. It has been established

to be both efficient and economically competitive with chlorination [66] [73]-[78]. Below high-pressure circumstances, non-equilibrium discharges in rare gases or rare-gas/halogen mixtures generate excimers, which do not have a stable ground state and disintegrate quickly, liberating in the process radiation in the VUV, UV, or even visible range [79]. **Figure 1** illustrates a usual DBD apparatus devised and optimized for both air and water disinfection investigations. The radiation formed via excimer lamps is incoherent; however, it is intense and spectrally selective. Plasma system makes able the formation of plasma-active species at atmospheric pressure without expensive vacuum setups. Active species could comprise UV or visible photons, charged particles, involving electrons, ions, free radicals [43], and highly-reactive neutral species, like reactive atoms (oxygen, fluorine, ozone, nitrogen oxides, etc.), excited states atoms, and reactive molecular fragments [55]. Emission of UV-light and production of radicals and charged particles take part in the demolition of pathogens in plasmas via fragmenting the strains in the DNA and demolishing the shell of a cell through chemical responses [1].

As aforesaid, in 1857, Siemens [9] employed DBD for producing ozone from air or oxygen. Currently, employing DBD for producing ozone is an efficacious instrument as a substitutional disinfectant thanks to its strong oxidation impacts. Ozone is performant in demobilizing bacteria, viruses [80], protozoa, and endospores. Instantaneously, ozone decays throughout water treatment producing hydroxyl free radicals ($\cdot\text{OH}$), which are viewed as the most efficacious oxidizing agents in water that could ruin the cell of microbes or remerge forming hydrogen peroxide (H_2O_2) that is a powerful oxidant itself [55] [81] [82] [83]. On the other hand, ozonation possesses drawbacks since it could generate mutagenic and carcinogenic agents (DBPs) like bromide in the treated water [1] [84]-[89].

As mentioned above, ozone formed in the plasma zone is a strong oxidizer that can demolish pathogens efficaciously [1] [55]. In nature, ozone level changes from 0.01 ppm to 0.05 ppm, following the season and geographic location.

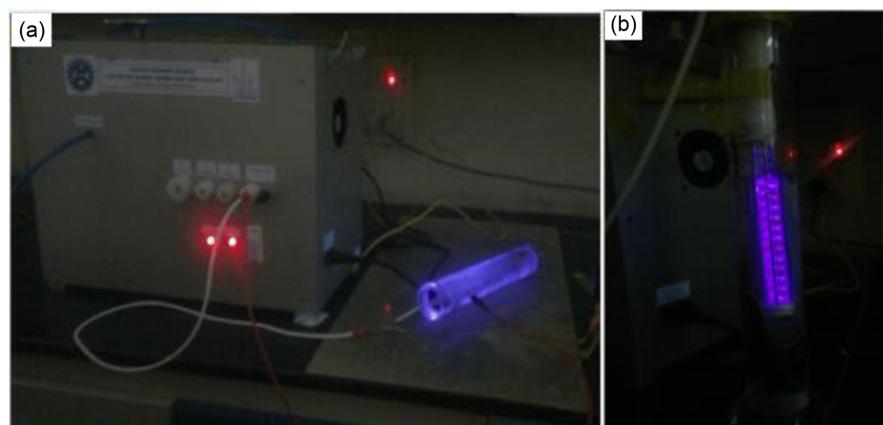


Figure 1. Usual DBD devise for both (a) air disinfection and (b) water disinfection researches [1].

High-voltage ozone generators form ozone/gas mixture that carries 1% to 3% ozone if utilizing dry air, and from 3% to 6% ozone if high-purity oxygen is employed as a feed gas. Practically, it is demonstrated that high-level ozone can be a performant air disinfectant. Thanks to the intrinsic characteristics of DBD plasmas to form active species and UV irradiation, the grid disposition constitutes a low-cost option to traditional disinfection techniques. Such filaments, also famous as micro-discharges, are the active zones of a DBD in which active chemical species and UV/VUV radiation could be formed. Such micro-discharges work as individual discharges that run independently of one another [1].

Theoretically, a DBD source is run on a large range of parameters such as the thickness of the dielectric layer, gas gap, gas pressure, etc. [1].

The interdependent link among all the variables remains fundamental to attain wanted radiation (172 nm peak for Xe) for lengthy functioning. In order to ameliorate the performance of compact sealed-off excimer source, a demountable DBD characterization device of DBD source with optimization of geometrical and electrical variables is needed. Founded on optimization parameters, sealed off DBD tubes should be manufactured. Excilamps could be adopted as interesting choices to mercury lamps and lasers for utilizations in microbial control techniques thanks to the absence of elemental mercury, long lifetime, geometric freedom, high photon flux, and mild running temperatures. The UV excimer sources have been suggested for demobilizing microbes following their wavelength and intensity. Photo-inactivation is engendered via modifying absorption levels of several biomolecules like DNA, membranes, or proteins. Phosphors could be employed to convert its VUV radiation to visible light [79] [90]. This wavelength transformation is used in mercury-free fluorescent lamps [1].

5. Suggested Principles

For adequate clarification of pathways supporting such a discharge process for the induction of corona discharges in water, principles have been suggested [1].

5.1. Electronic Principle

Following the electronic principles, below the implemented electric field, the free electrons speed up and could shock with and ionize the ambient molecules, so forming more free electrons (*electron avalanche*) and conducting to breakdown in water. The fundamental DBD is completely following the electric field utilized. If the used electric field is augmented to the ignition degree, the breakdown will happen and it is recognized as *micro-discharge*. The discharge period of micro-discharges is few nanoseconds and it is uniformly distributed over the dielectric surface. **Figure 2** could be utilized to interpret the general discharge behavior of DBD [1].

Provided that the gap voltage V_g is smaller than the ignition voltage, then there is no discharge activity and the device behaves like a series combination of two capacitance namely gap capacitance C_g and dielectric capacitance C_d . The

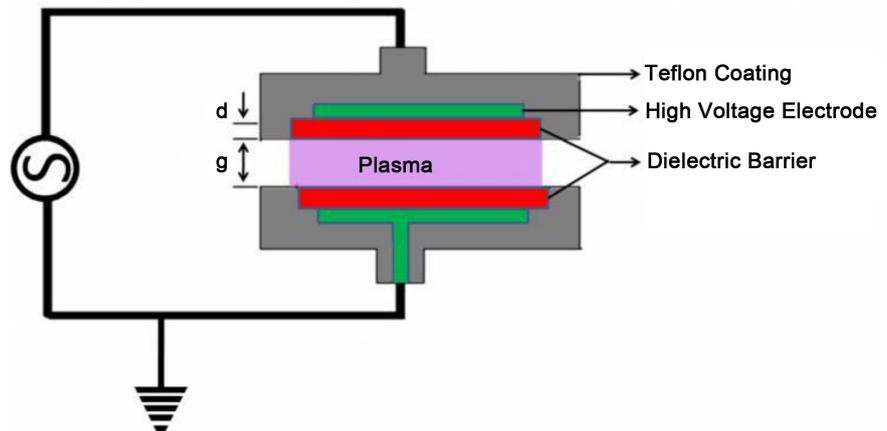


Figure 2. Schematic view of the parallel plate geometry of the DBD [1].

total capacitance C is given as [1]:

$$C = (C_d \times C_g) / (C_d + C_g) \quad (1)$$

At whatever time the gap voltage V_g crosses the ignition voltage level, then the micro-discharges are initiated. During half-cycle, the discharge voltage V_d persists approximately constant ($V_d = V_s = \text{Const.}$) and the current flow through the discharge gap is kept by a large number of micro-discharges (Figure 3). As a rule, the discharge voltage is a function of factors such as gas composition, pressure and, gas spacing. The micro-discharge pathway active in DBD is self-terminating and acts over a great span of supply frequencies with numerous voltage or current shapes. Figure 3 depicts a schematic view of the parallel plate geometry of the DBD employed currently in some labs. Upon inelastic collision, the free electron may ionize an ambient gas molecule, therefore forming more free electrons. The free electrons could repeat the phenomenon and so generate an electron avalanche (streamer). The discharge-generated ions cross the space and aggregate on the dielectric, where they form a reverse electric field and pause the current flow in a few nanoseconds. Because of the short period of the micro-discharge, only electrons, being the lightest charged particles, could earn high energy; however, the remainder of the heavier charges and neutrals stay close to room temperature. The energetic electrons, successively, initiate the plasma chemical reactions that in charge of the generation of free radicals and ions, which in the end ruin the contaminants [1].

5.2. Thermal Breakdown (Bubble) Principle

Following the thermal breakdown (bubble) principle, the current in the high-field region provokes heating and vaporization of the liquid, producing bubbles. Gas breakdown happens inside each bubble, forming more heating and development of the bubble until the total breakdown of the gap takes place. A sole streamer possesses a fraction of a millimeter diameter and could propagate to a distance of more than a centimeter in water [1].

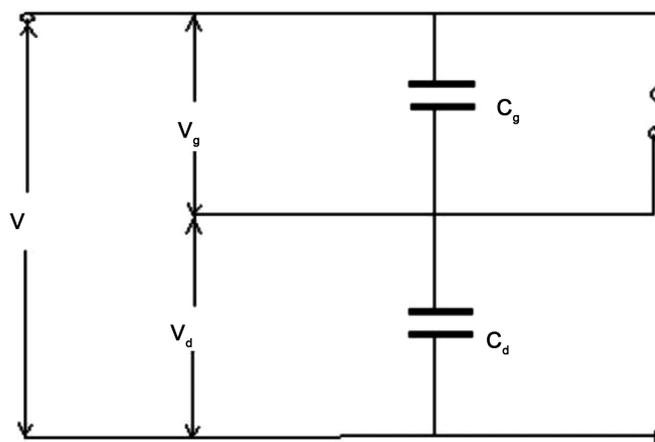


Figure 3. Equivalent circuit diagram of DBD [1].

A spark discharge furnishes a more reactive medium than a corona discharge thanks to its high-energy particles, UV radiation, shockwaves and supercritical water provoking temperature mediated transformation (which may attain 14,000 - 50,000 K) and free radical reactions in and around the plasma channel. At the moment that the high-voltage pulse finishes, the plasma channel cools and transfers its thermal energy to the surrounding water, conducting to the generation of steam bubbles [1].

The high-energy electrons formed in electrical discharges lead to the excitation, dissociation, electron capture or ionization of the target molecules [1]. Such free radicals ($\bullet\text{OH}$ in the instance of water), have a crucial contribution to demolishing contaminants [44] [48] [76] [82] [83].

6. DBD Disinfection Pathway

Numerous pathways have been suggested to interpret the production of diffuse DBDs. Those comprise gas pre-ionization by electrons or metastable from previous discharges and interaction among the plasma and the dielectric surfaces. Because atmospheric pressure circumstances are most appropriate for several DBD utilizations, the study of the features of the numerous discharge modes has concentrated mostly on atmospheric pressure circumstances rather than on the low-pressure regime. The bactericidal agents formed through DBD plasma can involve UV radiation, charged particles, ROSs, etc. The kinetics of cell decrease throughout plasma subjection is not symptomatic of UV radiation excitation. Both plant and animal hosts embrace defense action plans that employ the ROSs in opposition to the invaded microbes. The DBD could form such ROSs since oxygen atoms, ozone, metastable oxygen molecules, peroxide, superoxide, and hydroxyl radicals [91] [92] [93] [94], and all of them are germicidal. These ROSs possess a powerful oxidizing capacity and are apt to take action with the bacteria cells [95] [96] [97]. The ROSs could oxidize the cell membrane and could provoke the infiltration of cytoplasm [98] [99] [100] [101] [102]. With plasma treatment, in the first some seconds, because of the laceration of the cell mem-

brane, the cytoplasm progressively leaks out, leading to the concentrations of K^+ , protein [103], and nucleic acid in bacterial suspension augmenting to a higher level. Further, this is compatible with the rapid doom of cells during the first some seconds. Nevertheless, with the plasma subjection period prolonged, the escaped protein and nucleic acid will be progressively oxidized by ROSs [103] [104] [105] [106] [107], conducting to the diminution of their level; however, for K^+ , it cannot be oxidized, and so, its level turns saturated. Following this mechanism, the bacteria cells are murdered. Consequently, the ROSs [108] [109] could possess a major contribution to the demobilization phenomenon. A greater discharge power correlates to more ROSs formation and better demobilization impact [1].

Plasma sterilization can be categorized into three routes: the hydroxyl radical could fix to unsaturated fatty acids and provoke lipid peroxidation, oxygen radicals could give rise to DNA oxidation, and oxidation of amino acids could happen pursued by protein oxidation [1].

Fatty acid peroxide could be produced via plasma and could modify the membrane lipids. However, sterilization did not perform by a single chemical impact. The charge aggregation on the cell membrane caused electrostatic stress that was in charge of cell laceration. Using plasma, the sterilization pathway has been broadly investigated; however, it remains ambiguous. Indeed, the route of sterilization through DBD influence stays not yet fully comprehended. Electric field and reactive species are fundamental parameters for bacterial demobilization [65] [91]. Active species comprise UV or visible photons, charged particles (like electrons, ions, and free radicals), highly reactive neutral species like reactive atoms (oxygen, fluorine, ozone, nitrogen oxides, etc.), excited states atoms, and reactive molecular fragments. The collision of heavy ions with microbial cells could ruin their membrane. The collision of energetic electrons with some atoms and molecules could fracture some molecular bonds and form the excited and active particles like radicals and metastable atoms [1].

7. Impact of Sterilization Employing DBD Setup

Disinfecting water and wastewater via UV radiation looks to be a prospective option for chlorine [110] [111] [112] [113] [114]. Small levels of chlorine remainders are poisonous to aquatic life, and numerous of the DBPs of chlorination are mutagenic [91] [115] [116] [117]. Germicidal UV radiation does not form unwanted DBPs and it is efficacious in demobilizing a set of pathogens [1] [14] [118] [119].

Moreover, the impact of electrical sterilization could be efficient if the current flowing through spores augmented via adding water. Throughout the subjection time to DBD, the cell membrane can be breakdown thanks to the elevated electric field across the membrane. The impact of DBD treatment begun to be softer with augmenting the discharge gap. Adding NaCl augmented the impact of sterilization and attained the maximum at 4 g/L of NaCl [94]. Nevertheless, steri-

lizing with 8 g/L and 10 g/L NaCl solution was smaller than that of pure water. If the spores are wrapped with conductive liquid, they depicted to be electrically protected so that the electrical membrane breakdown is eliminated [1].

The tests performed to examine the influence of sterilization on *Bacillus subtilis* by the dry method show that almost all *B. subtilis* spores were neutralized by the dry method with a D-value of around 40 s as juxtaposed to the wet method, which was approximatively 7 s. Such findings propose that adding water improved the sterilization influence. It is suggested that $\bullet\text{OH}$ were formed from H_2O , and the sterilization was efficiently realized via the $\bullet\text{OH}$ generated in the spore's neighborhood. Oxidative decomposition pursuing VUV photolysis of water, thanks to its ease, has quickly turn into an interesting option to else advanced oxidation processes (AOPs) [1] [48] [76] [82] [83] [120].

Bactericidal impact of UV light for the bacteria *E. coli*, *Salmonella typhi*, *Shigella sonnei*, *Streptococcus faecalis*, *S. aureus*, and *B. subtilis* spores was estimated for a 99.9% demobilization of the cultured vegetative bacteria, total coliforms, and standard plate count microorganisms. Nevertheless, the viruses, the bacterial spores, and the amoebic cysts requested around 3 - 4 times, 9 times, and 15 times, respectively, than the injection needed for *E. coli*. Such ratios covered a narrower relative injection span than that already mentioned for chlorine disinfection of *E. coli*, viruses, spores, and cysts [1] [80].

Consequently, the injections of UV light needed to neutralize pathogenic microorganisms, involving viruses, bacteria spores, and protozoa, are much more similar to the injections of UV light indispensable to demobilize indicator bacteria than is the instance for chlorine [80]. Therefore, the UV degrees requested to satisfy coliform standards could be comparatively more performant than chlorination in neutralizing pathogenic microbes. In the main, the VUV method is so easy and possesses special merit that no chemical products request to be introduced. The technique constitutes a dare to different photochemical water treatment methods [1] [16].

The impact of the DBD device on sterilization has been well investigated on vegetative bacteria, viruses, bacterial spores, and protozoa [1].

8. Impacts of DBD Plasma

Numerous scientists have noted several empirical findings and data founded on their watching of the influence of DBD plasma on bacterial survival, level of membrane-bound proteins, and intracellular proteins and polysaccharides of the cell membrane (Figure 4) [42]. Such results provide a new understanding of the action of DBD plasma throughout microbial disinfection techniques [1].

8.1. Bacterial Survival

After subjection under plasma discharge for 1 min 12-h culture of cells, the actions of the fundamental enzymes in cells, such as glycerol dehydrogenase and glycerol dehydratase, were augmented by 12% and 62%, respectively. Such a

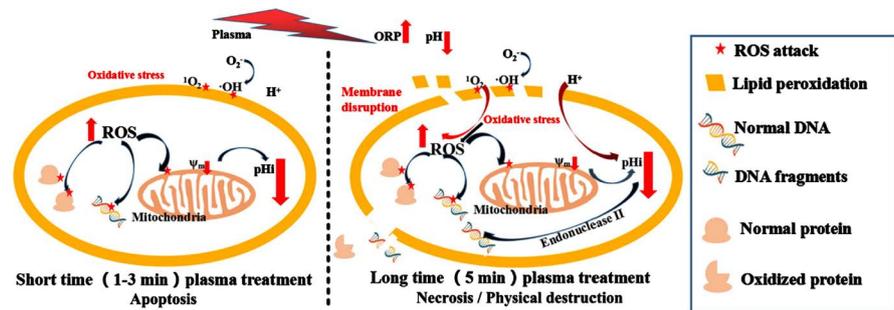


Figure 4. Schematic illustration of the antimicrobial mechanism of plasma in water disinfection [42].

result proposes that the plasma discharge enhanced the actions of basic enzymes in cells to conduct to the amelioration of viability. The electric field may be one of the bacteria demobilization routes. The work of implemented electric field on bacteria sterilization was examined, and the voltage was elevated progressively up to the critical discharge voltage. The protein and nucleic acid levels were metered at wavelength 280 nm and 260 nm, respectively. Since the absorbance estimate is proportional to the concentration, it reflects the detected level indirectly of the considered component. When the subjection period surpassed 10 s, the decreasing rate of protein concentration varied at a slow speed; however, the decreasing rate of nucleic acid did not change much [1] [103].

The UV absorbance at 254 nm was metered, and a small correction for UV light scattering via suspended particles was performed. The turbidity of the samples was less than 4 nephelometric turbidity units (NTU), thus the interference with coliform expansion related to elevated turbidity was perhaps lower. The survival of fecal coliforms was bigger than that of total coliforms. Such dissimilarity can be affected either to the differing UV sensitivity of the different groups of species comprised in the total coliform group or to the influences of the test circumstances on the repair of sublethal harm. Sublethal UV harm could be restored below some circumstances, and bacteria can then constitute colonies [1].

On the other hand, it is accepted that the standard fecal coliform most probable number (MPN) approach may undervalue the real population of fecal coliform bacteria below some circumstances. The wide dissimilarities in dose-survival links in diverse UV disinfection investigations are possibly attributed to additional parameters, like the trouble in deciding UV injection, rather than to the approach of counting coliforms [70] [71] [72]. Excimer UV radiation is so performant contra numerous sorts of bacteria in suspension in the comparatively limpid water. The elevated neutralizing performance of UV is not affected just to the generation of thymine dimers in the DNA. Cell lysis has the main contribution to the neutralizing phenomenon throughout the period of the subjection [1].

Bacteria own a collection of DNA reform systems, allowing quick recuperation from sublethal UV harm [1]. Several enzymes for DNA reform are also generated via oxidative stress in bacteria. They are essentially implied in two

kinds: base excision reform and nucleotide excision reform. The reform pathways initiated via plasma discharge lead to improved survival ratios. Nearly 100% of *S. aureus* and *E. coli* strains were neutralized in less than 10 s and 7 s of plasma treatment, respectively. The ROSs in plasma have a controlling contribution in the demobilization phenomenon but not the electric field. The ROSs could oxidize the cell membrane and therefore harm the protein and nucleic acid within the cells and, therefore, eliminate the bacteria [103].

Laroussi *et al.* [121] discovered that the plasma subsection formed gross structural injury in the Gram-negative *E. coli*, while none was detected in the more structurally solid Gram-positive *B. subtilis* (Figure 5) [122]. The noted removal in *B. subtilis* cells shows that the breakthrough of reactive species via the cell membrane could be probable [123] [124]. Yu *et al.* [125] proposed that DBD plasma can lead to pH diminution in the medium, which cannot be sufficient to demobilize the viable yeast cells but might participate in noticeable harm of the demobilized cells.

8.2. Concentrations of Membrane-Bound and Intracellular Proteins

The absorbance of cell membrane samples subjected to the DBD plasma in helium augmented linearly with the vulnerability period. The protein level in cell supernatant augmented, while the level of membrane-bound proteins diminished with plasma vulnerability [1].

More details may be found elsewhere [1].

8.3. Polysaccharides of Cell Membrane

Via DBD plasma, the polysaccharides on the cell wall and membrane could be fragmented. The electrostatic impact remains one likelihood of the tear of cells. With the subsection period, the level of amino acids in cell debris suspension was augmented. The decay of biomacromolecules on cell wall and membrane

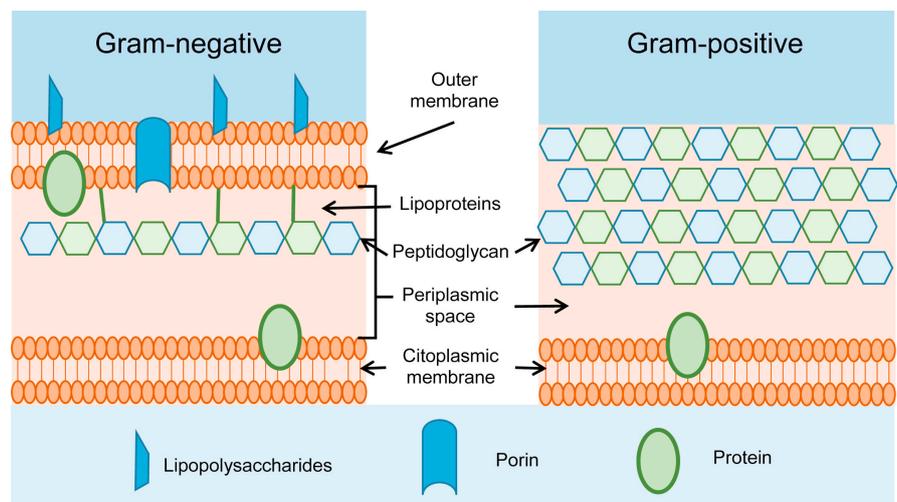


Figure 5. Structural differences between gram-negative and gram-positive bacteria [122].

stimulated the cell tear and liberation of cellular contents (like proteins) from the cellular cytoplasm into the extracellular medium constantly. The DBD plasma in helium at atmospheric pressure touch the viability of *K. pneumoniae* and the decay of biomacromolecules, such as polysaccharides and proteins via a set of complex chemical responses conducted by oxidation and degradation of proteins and polysaccharides from cell wall and membrane to produce amino acids, peptides, maltose, glucose, and acetic acid [1].

9. Water Purification via Electrical Discharges

Electrical discharges in association with strong electric fields, shock waves, UV radiation, O₃, H₂O₂, etc., all of which could participate for microbial disinfection and are an efficient sterilizing agent. It has been suggested that high-intensity pulsed electric fields without corona or spark discharge activities could be viewed as a promising technique for sterilizing food products since they neutralize microbes without ruining food constituents and its nourishing level [64]. Diverse parameters like the period and the maximum level of the voltage, the shape of the electrodes, etc. dictate the sterilization performance of a pulsed electric field. Germicides like O₃ or H₂O₂ enhance its performance on a small scale. It is suggested that corona or spark discharges below the powerful electric field ameliorate the sterilization phenomenon [1].

Electrical discharges occurring in an air or oxygen medium transform oxygen into ozone [1]. In addition to ozone, electrical discharges in air generate a collection of chemically active species, such as $\bullet\text{O}$, $\bullet\text{OH}$, $\bullet\text{N}$, O³⁻, N²⁻, N⁻, OH⁺, O⁺, O²⁺, N²⁺, N⁺, O⁺, etc. Such species are short-lived and disintegrate before ozone enriched air/oxygen enters into the water. Further, electrical discharges in aerated water are likely and they form $\bullet\text{OH}$, $\bullet\text{H}$, $\bullet\text{O}$, O₃, H₂O₂, etc. The *in situ* electrical discharges for ozone generation in water can furnish a tool to employ most of such chemically active species for water purifying. The interactive impact of powerful electric fields requested for electrical discharges in integration with traditional disinfectants like O₃ and H₂O₂ are fatal to numerous pathogens detected in the water. In water, the electrical discharges could generate UV radiation and shock waves that are useful in ruining contaminants. Electrical discharges are the best and environmentally-friendly next-generation techniques for water treatment and they can look far more performant than traditional oxidants and disinfectants.

As mentioned above, in a DBD device, the electrical discharges occur among electrodes where at least one of the electrodes is enveloped with a fine film of dielectric material, like glass or quartz [1]. Upon inelastic collision, the free electron may ionize an ambient gas molecule, so forming more free electrons that may reiterate the phenomenon and then generate an electron avalanche (streamer).

In both instances of contact glow discharge electrolysis and DBD setups, the electrical discharges happen in the gas phase in adjacent proximity to the water

surface. They need a strong electric field (>1 MV/cm) for the electrical discharge to occur in water. These elevated electric fields are probable via implementing high-voltage pulses (15 - 100 kV) in pulsed corona discharge and are employed as efficacious disinfectants [1]. For this reason, most of the researches on water treatment are realized utilizing pulsed corona discharge devices and the obtainable industrial-scale units are also founded on such process. The pathway of the induction of corona discharges in the water stays not completely grasped and more investigations are being dedicated to examining such techniques.

10. Ozone for Treating Water

The ozone level can be augmented by augmenting the ozone production reactions and/or via diminishing the ozone demolition reactions. Using a double discharge surfaces' reactor or a hybrid of silent and surface discharges could augment the number of sites for ozone generation reactions inside the given discharge volume. Further, porous silica gel packing could efficiently augment the ozone production performance via the spreading of an active plasma zone through micro-discharges. The pore size of alumina packing is a crucial parameter in ozone formation activity, where the dissolved ozone dissociates into $\bullet\text{OH}$ via a cyclic chain pathway that is also in charge of the oxidation of aqueous pollutants. It is known that $\bullet\text{OH}$ ($10^7 - 10^9 \text{ M}^{-1}\cdot\text{s}^{-1}$) is much more performant than O_3 ($10^1 - 10^7 \text{ M}^{-1}\cdot\text{s}^{-1}$) for organic pollutants decomposition. Consequently, a quicker transformation rate of ozone into hydroxyl radicals dictates the effectiveness of pollutant ruin. In AOPs, UV radiation, H_2O_2 , activated carbon, etc., catalyze the O_3 to $\bullet\text{OH}$ transformation. Besides, the catalytic transformation of ozone into $\bullet\text{O}$ could ameliorate the performance of ozonation [1].

Throughout ozonation, pulsed corona discharge possesses a small number of benefits; where distributed ozone enriched air/oxygen augments the rate of ozone dissolution in water and produces extra free radicals like $\bullet\text{OH}$ and $\bullet\text{O}$ [1]. The technology of pulsed corona discharge in water throughout ozonation has to be more studied because the electrical discharge could form free radicals and neutral active species and can avoid the necessity of a distinct device for producing ozone. In the main, the density of the chemically active species augments with an elevation of the applied voltage. The pathway of ozone transformation to free radicals throughout the ozonation technique is attributed to, negative polarity of direct current (DC) voltage, bubbling some gas throughout the discharge, and utilizing argon instead of oxygen for gas bubbling [1].

11. At COVID-19 Time: Arc Discharge-Mediated Disassembly for Killing Viruses

Employing a submerged plasma reactor of arc discharge (underwater arc) (Figure 6) that formed a shockwave, UV light, ROSs, and reactive nitrogen species, Lee *et al.* [126] studied its demobilization impacts on murine norovirus (MNV-1) with/without purification in water. Underwater arc treatments of 3

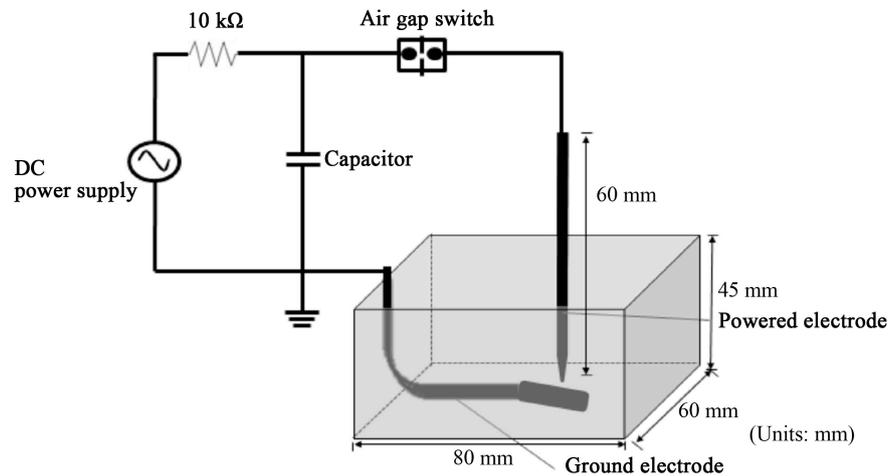


Figure 6. Schematic diagram of the underwater arc discharge [126].

and 6 Hz at 12 kV conducted to 2.6- and 4.2-log removals in the virus titer of non-purified MNV-1 after 1 min of treatment, respectively. The removal of purified MNV-1 was bigger than that of non-purified MNV-1 after underwater arc treatment for all applied conditions (12 or 15 kV and 3 or 6 Hz). One of the viral capsid proteins (VP1) was not observable after underwater arc treatment, when its integrity was assessed by western blot analysis. Further, transmission electron microscopy (TEM) analysis showed that MNV-1 particles were fully disassembled by the treatment (**Figure 7**). Such research proves that underwater arc treatment, which was apt to decaying the MNV-1 virion structure and the viral capsid protein, could be a performant disinfection technique for killing water-borne noroviruses.

These excellent results are encouraging for COVID-19 elimination from both water and wastewater. Systematically testing such outstanding technologies (**Figure 8**) could lead to defining the best configurations and optimizations for removing COVID-19 and other pathogens from water and wastewater [122].

Concerning the design of these highly-efficient processes, the focus would be accorded to intensify the reactors in terms of residence time and close contact opportunities between water pollutants and electrodes area [75]. Further, combining plasma discharge, electrocoagulation (EC) [127] [128], and magnetic field application [127] [129] as a hybrid process would lead to better efficiencies in removing pathogens and organic matters (OMs) [130] [131] [132] [133]. The last stages could contain activated carbon adsorption assisted by a membrane process [134]-[139] to remove the remaining OM especially released from the cellular cytoplasm during oxidation and disinfection processes.

12. Conclusions

From this work, the main conclusions emerge:

1) More efficient, cheaper, and environmentally-friendly than traditional water treatment methods, electrical discharge technologies are confirmed as. UV

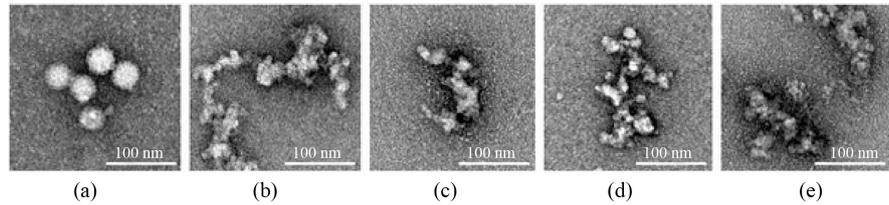


Figure 7. TEM observation of a purified MNV-1 preparation using underwater arc discharge. (a) Non treated control; (b) 12 kV, 3 Hz treatment; (c) 12 kV, 6 Hz treatment; (d) 13 kV, 3 Hz treatment; (e) 15 kV, 6 Hz treatment [126].

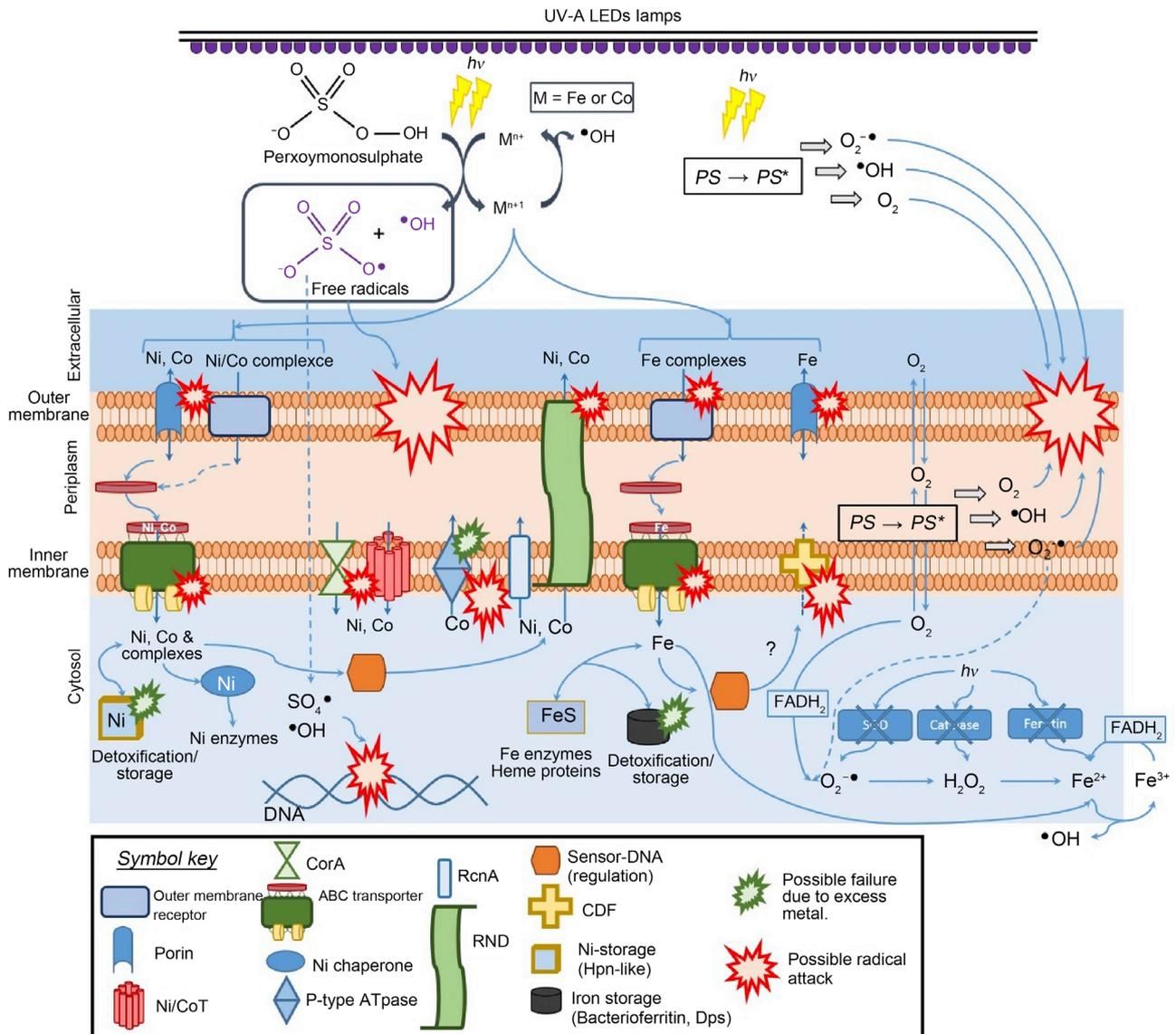


Figure 8. Likely pathways implied in photo-inactivation of *E. coli* via peroxymonosulphate (PMS)/UV-A LED and PMS/Mn⁺/UV-A LED [122].

emission from plasma dispositions and the impacts of irradiation on microorganisms become broadly studied. In the field of treating water via electrical discharges, more expansions are, however, requested. Especially, novel and more

performant materials that can be employed as catalysts for producing ozone are required. More importantly, the catalyst materials' physicochemical characteristics contribution should be more highlighted. Throughout ozonation, implementing pulsed high-voltages can lead to better diffusion of ozone in water and quicker transformation of ozone into free radicals that could reduce the ozonation price. Via direct electrical discharges, purifying water has trends to be examined on a large-scale. In this context, the demolition mechanisms of water contaminants, comprising pathogens and poisonous OMs, have to be deeply investigated. Further, defining the demolition by-products has to be performed to illustrate the route of plasma chemical responses implied [1].

2) Both in water and above water level, the electrical discharges possess their advantages and disadvantages. Above water level, which is in the gas phase, electrical discharges need less energy for the discharge to occur; however, in water, electrical discharges need an easier setup and form the chemically active species that could immediately bombard the aqueous contaminants. One of the kinds of electrical discharges, pulsed corona discharge remains the most tried and looks to be the most encouraging for treating water. Minutely set the UV injection needed to kill pathogenic microorganisms, comprising bacteria, viruses, spores, and cysts, stays to be worked on it for better UV disinfection performance.

3) Through this work, the examined techniques, especially plasma discharge, show good results in dealing with viruses' removal. Such methods could be methodically experimented with determining the optimal circumstances for killing COVID-19 and different pathogenic microbes from water. The attention can be dedicated to enhancing the devices in a matter of residence period and approaching contact among microorganisms and electrode surfaces [75]. Merging plasma discharge, EC, and magnetic field implementation can lead to better performances in eliminating viruses and OMs. As a secure physical separation, the final step has to involve activated carbon adsorption pursued by a membrane process to retain OM liberated from the cellular cytoplasm throughout disinfection methods.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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