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REVIEW ARTICLE

Advantages of ozone disinfection method for water purification over chlorine disinfection

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ABSTRACT

The improvement of various advancements for water sanitization is a significant issue. Among numerous elective drinking water treatment advances, the well-known disinfectant techniques are ozonation and chlorination to treat drinking water. All through the ozonation procedure, it produces biodegradable organic by-products while in the chlorination process, some risky by-products (trihalomethanes and haloacetic acids) are generated. Because of the possible danger of these results, several water purification methods have been reported, such as ozonation, chlorination, UV, etc. During ozonation, exceptionally reactive hydroxyl radicals are produced, which has a crucial effect on purifying water. In this paper, we have discussed the wide use of ozone disinfectants for water treatment with an emphasis on radical chemistry of ozonization as well as advanced oxidation processes instead of the chlorination process, low-cost ozone generation processes, the impact of ozone and chlorine disinfectants on cryptosporidium oocysts, and the removal of seven strains microbes from drinking water. The favorable circumstances, hindrances of the utilization of ozone and chlorine in wastewater treatment, and their confinements in water treatment innovation just as the elective advances, for example, ozone-based oxidation process, catalytic ozonation, photocatalytic oxidation, and so on are additionally clarified in this paper.

Keywords: drinking water purification; disinfectant; ozonation; chlorination; by-products; advanced oxidation processes

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1. Introduction

The major worldwide challenge for the 21st century is to get drinkable water, as pure and uncontaminated water is the basic requirement for all living organisms. More than 71% of the earth's surface is covered with water, but only less than 1% of water is drinkable because of different contaminations. Contaminated drinking water causes public health risks throughout the world[1]. From different research, it has been found that 1.2 billion people have lacked to access harmless drinking water while 2.6 billion have little or no sanitation with dying millions of people annually[2] from diarrheal diseases and poor hygiene performs. That's why it is a crying need to remove the undesired materials present in water. Mainly this paper describes different types of treatment of wastewater as well as the comparison and contrast between two disinfestation methods of water purification by chlorine and ozone. Though chlorine-based disinfectants have been widely utilized for traditional drinking water disinfection against unfavourable compounds, nowadays it has been already proved that the chlorination process has generated more than 300 potentially

dangerous by-products[3]. Among them, trihalomethanes, haloacetic acid, chloral hydrate, and seven chlorine-resistant bacteria have been considered more dangerous[4–6].

Ozone is typically efficient, fast[6], and has been regarded as one of the best disinfectants for treating drinking water[7]. Having no side effects is the greatest advantage of this ozonation process. For killing or the decomposition of microorganisms, ozone has been normally used since a few years ago[8]. As per the U.S. ecological protection agency (USEPA), ozone is the most impressive disinfectant accessible[9]. During the deterioration of ozone, the oxidative OH radicals are produced have no unfavourable effects on the environment and have more prominent oxidizing power than ozone itself does[4]. There is no way possibly risky synthetic compounds are associated with ozone applications since the creation of ozone requires just air and power[10] whereas in the case of the chlorination process, bladder cancer, and rectal cancer could be attributed to chlorinated water and its by-products according to Morris et al.[11]. These above are the fundamental reasons why ozone is progressively associated with oxidizing purposes just as water treatment. Therefore, the use of ozonation at a lower cost is expanding. Be that as it may, ozone is regularly applied to drinking water alongside chlorine or UV inactivation as a result of its absence of ceaseless purification capacity[6]. Although ozone is known to be an incredible oxidant, it additionally has barely any detriments which limit its application in water treatment innovation that we have explained underneath in this paper.

In this review, we emphasize on the improvement of powerful, modest, and eco-accommodating ozone treatment for the purification of drinking water. However, water quality in our nation is featured section 1.1. In section 2, the ozone and chlorine are characterized by clarifying their disinfectant qualities individually. The efficiency of the formation of ozone and the radical chemistry of ozone and cyclic mechanism of ozone decomposition is described in this section 2. The approximate reduction of chlorine by-products by ozone treatment and identification of new ozone disinfection by-products are discussed in the subsections 2.7 and 2.8. In subsection 2.9, advanced oxidation processes (AOPs) for water treatment are exposed. One more thing that ought to be worried that the surface water is additionally tainted by cryptosporidium oocysts that may support watery suspensions for as long as a year at 4 °C[12]. Along these lines, we have discussed the impact of ozone and chlorine disinfectants on cryptosporidium oocysts in section 3. In this section 3, seven strains of chlorine-resistant microscopic organisms from the drinking water treatment plant and their effect of ozone inactivation are also investigated. At last, in section 4, a conclusion has been made by the general outcomes of this paper.

1.1. Water quality

Water is the most significant natural asset on the planet. There has an important human concern to getting drinkable water where the essential necessity is hygienic and germ-free drinking water. In the period of 1995, 3906 km^3 of water was pulled out in the whole world for use as drinking water by humans and, the world water is projected to withdraw the freshwater by increasing at least 50% through 2025[13]. On a per-capita basis, North America withdraws seven times more freshwater than Africa[14]. The drinking water quality interprets the state of water including physical, synthetic, and 80 natural qualities. Poor water quality causes extensive and serious illnesses. The quality of water 81 depends upon the presence of microorganisms in water. Most public drinking water is generally contaminated by different living and non-living organisms, organic and inorganic species, and numerous poisonous microorganisms of different sizes and shapes. The highly contaminated living and non-living microorganisms are viruses, bacteria, chlorine-resistant bacteria, protozoans, spores, fungi, and oocysts. The organic and inorganic species are humic substances, nitrate, arsenic, alkalinity, and colloidal particles also present in water[15]. This contaminated scarce drinkable water causes public health risks worldwide that has been investigated that 1.2 billion people have died due to lacking of safe drinking water, 2.6 billion have little or no sanitation, and millions of individuals die annually[2,16]. Indeed, even created nations with present-day sanitation and water gracefully frameworks are weak at this point. It is

very needed to eliminate the harmful species in water which can be done by several water purification techniques. The adsorption, coagulation flocculation, sedimentation, and filtration processes are preliminary simple, effective, and economical methods for wastewater purification. Sometimes drinking water also encloses some dangerous chemicals that may not be removed by these processes[17]. Whereas, the disinfection method (chlorination, ozonation) is a highly effective method for drinking water treatment which is elaborately described in this paper. The drinking water has been found from different deep tube-well and clean-water reservoirs. Generally, drinking tap water is supplied from different water supply reservoirs through coagulation, sedimentation, filtration, and chlorination processes. In different countries, the supplied water is further filtered through a 0.45-µm membrane before using it[18,19]. Raw water is analyzed for DO, BOD, COD, pH, chloride, TDS, etc. The approximate water quality in Bangladesh and Bangladesh standards (BDS) value is shown in **Table 1**.

Table 1. The approximate water quality of common drinking water and Bangladesh standards (BDS) value by WHO.

Parameter	Water quality standard	Bangladesh standards (BDS) value
Manganese (mg/L)	0.1–5.5	0.4
pH	6.9–7.8	7
BOD (mg/L)	0.57–3.84	0.2
DO (mg/L)	2.27–9.49	6
COD (mg/L)	65–106	4
Chloride (mg/L)	326–745	500–800
TDS (mg/L)	985–1480	1000
TSS (mg/L)	352–496	10
Nitrate (mg/L)	35.5–62.8	10
Sulphate (mg/L)	31–72	400
Turbidity (NTU)	5.0	5–25
alkalinity/(mg/L as $CaCO_3$)	80–250	-
Arsenic µg/L	50–200	10

2. Process techniques

2.1. Ozone and chlorine act as disinfectants

Ozone is an inorganic triatomic of oxygen (O_3) gas that has a characteristic pungent odor which is less stable than the diatomic O_2. Ozone cannot be stored and must be generated at its point of use[10,20]. Ozone is used for various purposes like disinfection, oxidation of inorganic pollutants (iron, manganese), and oxidation of organic micro- and macro-pollutants as well as for the improvement of coagulation[21]. Ozone occurs naturally in small amounts in the upper atmosphere by solar UV radiation where it has great biological and meteorological significance[22].

$$O_2 + \text{UV (ultraviolet ray)} \rightarrow O^{\bullet} + O^{\bullet} \text{ (Oxygen radicals)}$$

$$O^{\bullet} + \text{Oxygen gas } (O_2) \rightarrow O_3$$

When UV ray is conducted with oxygen gas with required electrical discharge then oxygen radicals are formed. Finally, the oxygen molecule and an oxygen radical are aggrandized together to form ozone. This ozone is utilized for a wide range of purposes, for example, cleansing and green growth control, taste, smell, oxidation of inorganic contaminations, and/or natural poisons[21]. To purify drinking water, disinfection, coagulation, flocculation, sedimentation, and filtration processes are used commonly. Among all the procedures of drinking water treatment, the disinfection process fills two primary needs: one is to inactivate microorganisms and another is to supply a disinfectant residual in finished water and prevent microbial

regrowth in water distribution systems. Ozonation is a widely utilized strategy in drinking water treatment during disinfection in water by ozone, highly reactive secondary oxidants (such as $OH^{\bullet}$), are formed which may speed up the decomposition of ozone and the radical-type cyclic reactions may occur to destroy the harmful species from the drinkable water[23].

Whereas, chlorine is a gas under normal pressure and temperature which can be compressed to a liquid and stored in cylindrical containers. When chlorine is dissolved in water under a vacuum, the solution becomes concentrated which can be applied to the drinking water treatment. Chlorination is the profoundly utilized disinfectant strategy that causes the formation of hazardous by-products called disinfection by-products (DBPs) are trihalomethanes (THMs), haloacidic acids (HAAs), haloacetonitriles (HANs), chloral hydrate, chloroform, dichloronitrile, dichloroacetic acid, and trichloroacetic acid, etc.[24].

$$\text{Harmful Species} + Cl_2 \rightarrow \text{DBPs}$$

The maximum contaminant levels are 80 μg/L for total THMs, 60 μg/L for the sum of five HAAs, and 10 μg/L for bromate suggested by USEPA[25]. The formation and treatment of THMs in drinking water have been studied by the institute for environmental research yonsei in Korea since the early 1980s, and THMs have been restricted to 100 μg/L in drinking water since 1991[24] as shown in **Table 2**.

Table 2. National primary drinking water regulations on chlorine DBPs[25].

DBPs of chlorine disinfectant	MCLG (mg/L)	MCL (mg/L)	Potential health risks from drinking water	Contaminant sources in drinking water
Bromate	Zero	0.010	Increased cancer risk	DBPs of water Drinking
Chlorite	0.8	1.0	Anemia, nervous system risk	DBPs of water Drinking
Haloacetic acids (HAAs)	N/A	0.060	Increased cancer risk	DBPs of water Drinking
Total trihalomethanes (TTHMs)	N/A	0.080–0.10	Kidney, liver, increased cancer risk	DBPs of water Drinking

2.2. Formation of ozone

In this paper, we have described ozone generation with minimal effort by an eco-friendly strategy named the electrical discharge technique. Because of high reactivity, ozone is very unstable in water[26]. Ozone conveys oxidizing power either straightforwardly or through the generation of $HO^{\bullet}$ free radicals in the disintegrated ozone into water[27,28].

Ozone has a short half-life and it is decayed after some time into its unique structure of oxygen (O_2). In this way, it must be created where it will be utilized. Normally, ozone might be created through a few chemical reactions. A typical example is the ozone layer, where ozone is made from the sun's UV beams. Be that as it may, ozone is likewise delivered at rainstorms and cascades.

As a commercially demanded treatment, there have huge researches put into methods of ozone production. Among different methods (such as electrical discharge, UV radiation, electrolysis, and radiochemical methods), the electrical discharge method is the common technique as well as comparatively effective to produce ozone in the ozone production process. In the case of water purification, the three following types of electrical discharges are often reported[28].

a) Pulsed corona discharges.
b) Contact glow discharge electrolysis.
c) Dielectric barrier discharges (also called silent discharges).

In comparison, on account of the pulsed corona discharge method, a high electric field is conceivable by applying high-voltage pulses of 15–100 kV, with a sharp rise time (a few nanoseconds) and short duration (nano to microseconds)[29]. Pulsed corona discharge is one of the most encouraging electrical discharge

processes for water purification because it delivers several strong oxidizing agents[30], which can decompose the organic pollutants in water[31]. The pulsed corona discharges are effective disinfectants[31]. This is why the pulsed corona discharge method is widely used to generate ozone for most commercial-industrial processes.

2.3. Ozone generation by pulsed corona discharges

A pulsed corona discharge reactor requires a pulse generator and a reactor[7] as illustrated in **Figure 1**. Pulsed corona discharges in water using a ceramic-coated stainless-steel wire electrode in a wire-cylinder type reactor has been reported[32]. In this method, the electrode may have a catalytic effect on the reactions, during water purification. An ozone production unit with corona electrical discharges consists of the following parts: oxygen source, dust filters, gas dryers, ozone generators, contacting units, faulty light switches, motor brushes, power transmission lines, and torch destruction.

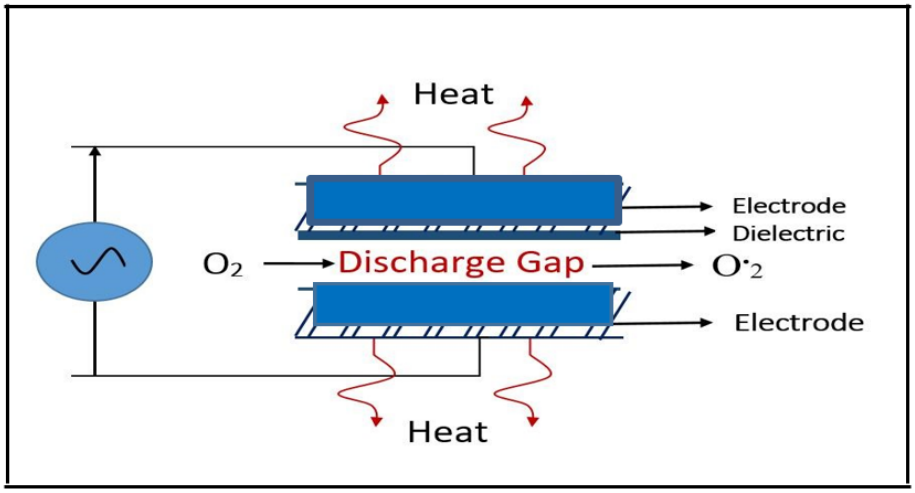

Figure 1. Pulsed corona discharge cell[7].

The space between the electric surface and the other conductive electric surface forms an air gap called the dielectric gap. When sufficiently applied across the two electrodes, the corona discharge forms this gap. Electrons are shifted across an air gap to provide them sufficient energy to break the oxygen double bond, consequently, atomic oxygen is produced[33]. During the ozone generation through the pulsed corona discharge method, ozone is formed with the separation of O_2 molecules by the electrons to produce the oxygen atoms[34].

$$O_2 \text{ (corona discharge)} + \text{energy (e)} \rightarrow O^{\bullet} + O^{\bullet} \quad (1)$$

The major reactions evaluated at 1 atm and 298 K are listed as follows[35]:

$$O^{\bullet} + O_2 \text{ (absorbed)} + M \rightarrow O_3 \text{ (absorbed)} + M \rightarrow O_3 + M \quad (2)$$

where, 'M' is a third collision partner (O, O_2, and N_2 in the air also)[35]. When the gas stream contains other gas components, oxygen atoms also can be generated via other reactions. Major reactions leading to the depletion of ozone are listed as follows[36,37]:

$$O^{\bullet} + O_3 \rightarrow O_2 + O_2 \quad (3)$$

$$OH + O_3 \rightarrow OH_3 + O_2 \quad (4)$$

$$HO_2 + O_3 \rightarrow OH + 2O_2 \quad (5)$$

$$H + O_3 \rightarrow OH + O_2 \quad (6)$$

$$H + O_3 \rightarrow HO_2 + O^{\bullet} \quad (7)$$

Corona discharge can produce medium-to-high concentrations of ozone, typically 15 %wt of ozone is generated from oxygen and 1%–3% from supplied air[38]. Apparently, the generation of ozone in a corona discharge reactor is complicated. Physical factors such as gas composition, temperature, gas residence time, and the applied force would determine the involvement of the above reactions in ozone synthesis. The

generation of ozone is very energy-intensive, important factors that influence ozone generation are oxygen concentration inlet gas, humidity and purity of inlet gas, cooling water temperature, and electrical parameters.

2.4. Contact glow discharge electrolysis (CGDE)

CGDE is an electrolysis method where a gentle covering of light-transmitting plasma creates around an anode that's swamped inside a moderately high-conductivity aqueous electrolyte. Studies on CGDE show that non-faradaic impacts begin here in two response zones: (i) the plasma zone, where a plasma chemical mechanism (counting $H^{\bullet}$, $OH^{\bullet}$, $O^{\bullet}$ species got from H_2O fume) works and (ii) the plasma-electrolyte interfacial zone, where a radiolytic mechanism (mediated through $H^{\bullet}$, $OH^{\bullet}$ species got from liquid water) works[26].

The application potential of CGDE is being explored significantly in several areas[26]. In the last ten to fifteen years, the progress made in some of these areas such as surface engineering[39,40], micromachining[41], nanoparticle fabrication[42] and wastewater treatment[43]. The main issue for the future is to discover the best reactor cell configuration and the operating conditions of CGDE for specific applications[44] is shown in **Figure 2**.

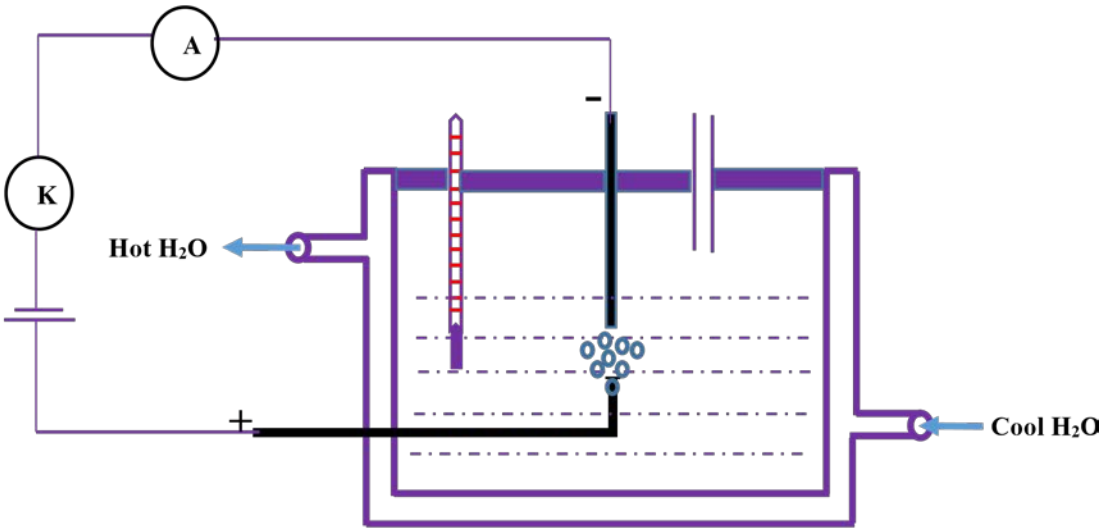

Figure 2. Instrumentation of contact glow discharge electrolysis[44].

2.5. Dielectric barrier discharges

Siemens[45] firstly proposed a general method of ozone formation known as a dielectric barrier discharge (also known as silent discharge) by which ozone is produced at larger quantities. A combination of heterogeneous catalyst and non-thermal plasma performs to be the best choice for this method[46,47]. There is a necessity to adjust the non-thermal plasma (NTP) reactor configuration in order to achieve the potential of the plasma catalytic technique, as most of the chemically active species generated in the plasma are short-lived and cannot reach the catalyst surface[48].

In this section, the present understanding of the dielectric barrier discharge has been summarized in this paper. The main characteristic of this method is the presence of one or two dielectric layers in the current path between electrodes and discharge space[49] as shown in **Figure 3**. Different planar or cylindrical configurations are common, and although they can be operated between line and microwave frequencies, the typical operating range for most applications lies between 500 Hz and 500,000 Hz[50]. In most gases at about atmospheric pressure, the presence of the dielectric leads to the formation of a large number of micro-discharges of nanosecond duration per cm^2[50]. It is well known that the separation of the two electrodes, humidity, gas composition, gas pressure, the thickness of the dielectric, and the power supply all have a strong influence on the strength of the micro-discharges[51]. In the case of dielectric barrier discharge reactor type of ozone generator under a gas flow system, O_3 generation efficiency is dependent on the applied voltage, composition,

and gas flow rate of the gas stream. Larger applied voltage and longer gas residence time would favor ozone generation, i.e., increase energy consumption, while the existence of moisture in the gas stream and higher temperature would decrease the ozone formation rate significantly[52].

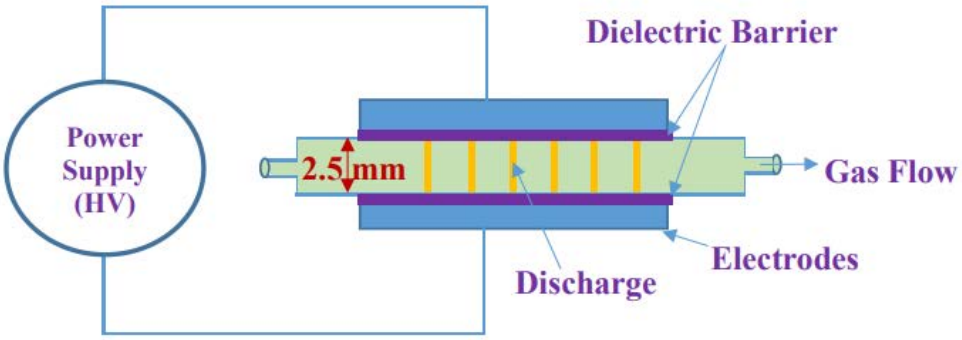

Figure 3. The dielectric barrier discharge cell[49].

2.6. The radical chemistry of ozone into water

O_3 provide oxidizing power not only directly, but also through the generation of OH radicals into the decomposition of dissolved O_3 into water. The ozone is decomposed in water by using advanced electrokinetic methods which are investigated by Staehelin and Hoigne[53] and Forni et al.[54] and Flanagan[55]. A comp lex cyclic mechanism is shown (**Figure 4**) that contains a series of the oxygen atom, atom transfer processes, and the intermediacy of OH. A numerous active chemical species can start the cycle are $O^{\bullet}$, $OH^{\bullet}$, $N^{\bullet}$, $O_3^{\bullet}$, N_2^{*}, N^{*}, OH^{-}, O^{2-}, O^{-}, O^{2+}, N^{2+}, N^{+}, O^{+}, $HCOO^{-}$, Fe^{2+}[56].

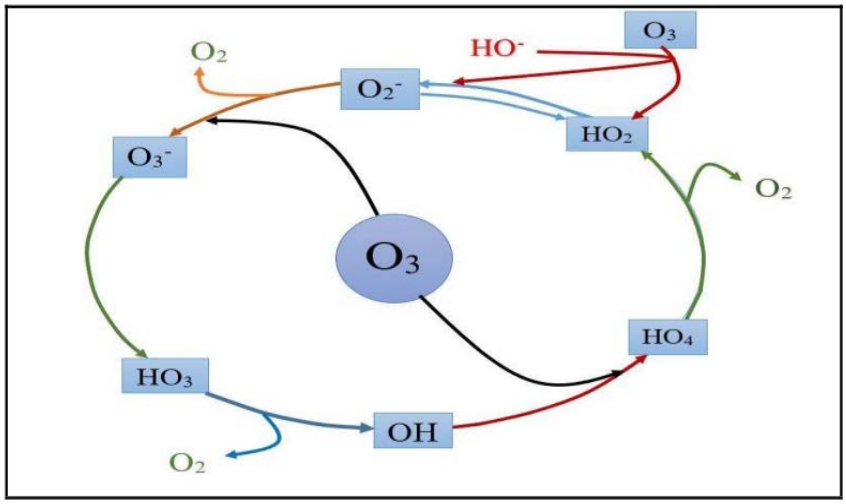

Figure 4. Cyclic chain mechanism of ozone decomposition to free radicals in the ozonation process[7].

When these above species are combined with ozone, electrical discharges in water may deliver a means to purify drinking water[4]. In very pure water, $OH^{\bullet}$ reacts with ozone so that the chain propagating steps shown in the circle in **Figure 4** can repeat again and again. Pulsed corona discharges are identified inside the bubble with the formation of $H^{\bullet}$, $OH^{\bullet}$ and H_2O_2[57]. The formation of these species from water is shown by the following two reactions.

$$e^{*}(\text{excited}) + H_2O = OH^{\bullet} + H^{\bullet} + e \quad (8)$$

$$2OH^{\bullet} \rightarrow H_2O_2 \quad (9)$$

The highly reactive radical OH is responsible for the decomposition of microorganisms in the drinking water[58] with extremely large rate constants for the reaction of OH with pollutants, representing that the

reactions are faster (107–109 M^{-1} s^{-1}) than ozone itself (101–107 M^{-1} s^{-1})[59]. The catalytic conversion of ozone into O• also can improve the efficiency of ozonation[60].

There is a common argument in the drinking water community on the role of OH• where some authors claim that ozone is the main disinfectant while others suggest that OH• may play an important role for disinfection. The kinetic data shown in **Table 3** together with the probable range of Rc values (Rc = [OH•]/[O_3] = 10^{-6}–10^{-9}) allow an estimation of the role of OH during disinfection[61]. The following rate equation can be formulated as:

$$\text{Log}\ (N/N_0) = -(Ko_3 + K_{OH}Rc) \int [O_3]\ dt$$

where Ko_3 is the rate constant of ozone for reaction with microorganisms; K_{OH} is the rate constant for the reaction of OH• radicals with microorganisms.

The reaction mechanism of ozone with other species may involve both direct reactions with O_3 and reaction with OH•, nonetheless, the pH of the system is neutral, because of the presence of OH– promoters have been observed by Staehelin and Hoigne[62] and Lim et al.[59]. They also have noted that O^{2-} are formed in the way of direct ozonation processes. Ultimately, the ozone gas is reacted with microorganisms by a combination of direct O_3 and OH• pathways. With the proceeding the reaction, the reaction mechanism may even become more radical in character, hundreds of ozone molecules may be decomposed by a single step where ozone has a very short half-life in the water at pH 7[4].

Table 3. Kinetics of the inactivation of microorganisms with ozone at pH 7[61].

Microorganism	CT_{lag} (mg $minL^{-1}$)	KO_3 (Lmg^{-1} min^{-1})	Eact (kJ mol^{-1})	Ko_3 (M^{-1} S^{-1})	T (°C)
E. coli	-	130	37	1.04×10^5	20
Rotavirus	-	76	-	6104	20
B. subtilis spores	2.9	2.9	42	2.3103	20
Giardia muris cysts	-	15.4	80	1.2104	25
Giardia lamblia cysts	-	29	-	2.3104	25
Cryptosporidium parvum oocysts	0.83	0.84	81	6.7102	20

2.7. The reduction of chlorine by-products by ozone treatment combining with UV

The chlorination method has been the most widely used disinfection method for drinking water treatment. But, there have some limitations that forms undesired halogenated by-products (THMs and HAAs) and less effective chlorine-resistant microorganisms such as *cryptosporidium parvum* and *giardia lamblia*[63].

For inactivating a diversity of microorganisms with producing no or, lower levels of THMs and HAAs, a secondary treatment with ozone and low-pressure UV have been widely used because of their high efficiency to treatment with only chlorine[3,64]. The ozone combining with UV radiation may be a very good option for drinking water purification. During this treatment, the generation of OH• radicals is the core target for the inactivation of microorganisms. It has been evaluated that the traveling distance of OH• in a cell is 6–9 nm, which damages DNA of the microorganisms[65]. Therefore, the formation of OH• radicals during the ozonation may benefit microorganism inactivation.

2.8. Identification of new ozone disinfection by-products (DBPs)

Drinking water disinfection by-products (DBPs) are an undesired result of using ozone as a chemical disinfectant to kill harmful species in drinking water. Richardson et al.[3] observed the origination of many ozone DBPs by using a combination of spectral identification techniques gas chromatography coupled with electron-impact mass spectrometry (GC/EIMS), and infrared spectroscopy (GC/IR). Many of the DBPs contain oxygen in their chemical structures such as aldehydes, ketones, and carboxylic acids are being the major classes

of by-products that are formed by reacting ozone with naturally occurring anthropogenic pollutants with no halogenated DBPs[13]. Generally, the halogenated DBPs are produced by the combination of ozone and chlorine treatment[3]. Although ozone is being used at many drinking water plants in the United States and also in Europe. There have been relatively few studies conducted to determine the identity of ozonation by-products including aldehydes, ketones, ketoaldehydes, carboxylic acids, aldo-acids, keto acids, hydroxyl acids, alcohols, esters, and alkanes[3,66]. There have some inorganic DBPs too including bromate and hypobromite[67].

Additionally, Richardson et al.[3] also investigated, when ozone is applied as a secondary disinfectant after chlorination, there have a little known about the identity of DBPs. Therefore, it can be better to drink water treatment plants using chlorine with ozone as a secondary disinfectant.

2.9. Advanced oxidation processes for water treatment

There have many advanced oxidation processes for purifying drinking water. Advanced oxidation processes (AOPs) are a set of chemical treatment techniques that employ a combination of oxidation agents (O_3, H_2O_2), radiation (UV, ultrasound), and catalysts to generate $OH^{\bullet}$ radicals (which are more powerful than molecular ozone) to destroy the organisms in drinking water[4].

Because of decomposition of ozone molecules into hydroxyl radicals is predominant under ozonation at high pH (> 8), this is observed as an AOP where the reactions between the radicals and different organisms take place[68]. The prime difference between the AOP and ozonation is the AOPs depend mainly on oxidation with OH radicals, whereas the ozonation process mainly depends on the direct oxidation with liquified ozone. Several AOPs are listed in **Table 4**.

Table 4. Typical AOP systems.

Photochemical processes	**Non-photochemical processes**
O_3/UV	Ozonation at elevated pH (> 8.5)
H_2O_2/UV	Ozone + hydrogen peroxide (O_3/H_2O_2)
O_3/H_2O_2/UV	Ozone + catalyst (O_3/CAT), i.e., catalytic ozonation
Photo-fenton/fenton-like system	Fenton system (H_2O_2/Fe^{2+})
Photocatalysis	-

The half-life time of the ozone molecule depends on pH, water temperature, and concentration of organisms in water[21,69]. The decomposition of ozone follows a pseudo-first order kinetic law[21]:

$$-(d[O_3]/dt)_{pH} = k[O_3]$$

where, 'k' (for a given pH value) is a pseudo-first-order constant. Basic pH causes an increase in ozone decomposition[70].

The decomposition of O_3 does not impact on $OH^{\bullet}$ radicals at pH < 3 but at 7 < pH < 10, the typical half-life time of ozone is 15 to 25 min[69]. However, above some critical pH values the ozonation process is less effective[71]. For illustration, the critical pH is 7.5 during the oxidation of most humic elements which indicates the approximate pH value at which the decomposition of O_3 to $HO^{\bullet}$ radicals increase rapidly, consequently increasing organic oxidation rates. **Figure 5** shows the effect of pH variation on ozone decomposition in wastewater[72].

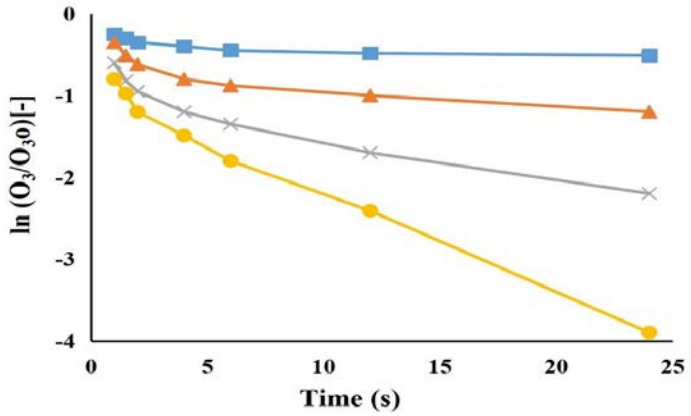

Figure 5. The effect of pH variation on ozone decomposition: ozonation of wastewater at a dose of 2.5 mg/L (52 mM)[72].

At pH 2, there is a slow ozone decomposition for up to 10 seconds and then, as pH increases from pH 2 to pH 7.9, the ozone decomposition rate increases rapidly[72]. Hence, the HO˙ generation is increased significantly with increasing pH value.

AOPs can decrease the concentration of contaminants from several-hundred ppm to less than 5 ppb where the contaminant materials are converted into stable inorganic compounds such as water, carbon dioxide, and salts[73]. AOPs are particularly useful for cleaning aromatics, pesticides[74], petroleum constituents, and volatile organic compounds in wastewater[75]. Chemistry in AOPs could be essentially divided into two parts[76]:

a) Generation of ˙OH: primary attacks on target microorganisms (through ˙OH radicals) and then their disruption to fragments.

b) Subsequent attacks by ˙OH until final mineralization: among all AOPs, a comparatively effective process described below has been applied both in drinking water and wastewater treatment: in-situ chemical oxidation contains the formation of highly reactive hydroxyl radicals (˙OH) which can eliminate contaminants quickly and efficiently. Hydroxyl radicals are generated with the help of ozone, hydrogen peroxide, oxygen, and/or UV light or catalysts (e.g., TiO_2). Combinations of these reagents are applied in order to obtain a maximum ˙OH radicals. The cause of giving priority to AOPs is: they can effectively eliminate organic compounds and transfer them into another phase. But the cost of AOPs is fairly great since a continuous input of expensive chemical reagents is required to maintain the operation of most AOP systems[77]. However, Ikwhata and El-Din[68] investigated that ozone-based AOPs are generally more effective than ozonation alone in degrading pesticides present in water, although total destruction of pesticides is impossible with ozonation alone or even with ozone-based AOPs.

2.10. Catalytic ozonation

The ozonation process is known as a cost-intensive technology regarding wastewater treatment. Because of both the high cost of ozone formation and only partial oxidation of organic compounds present in water, the application of ozonation might not be feasible from an economic point of view[70], both the cost-effective and the efficacy of the ozonation process needs to be enhanced by increasing the solubility of ozone where it's decomposed to generate more active oxidant agents (such as OH) with higher reaction rates. An alternative ozonation process for enhancing efficiency and cost-effectiveness is the addition of a homogeneous or heterogeneous solid as a catalyst into the ozonation reactor, which is called catalytic ozonation processes (COPs)[78].

Catalytic ozonation is a new means of harmful species removal from drinking water and wastewater[78]. In the catalytic ozonation process, the catalyst provides a surface for the reaction of O_3 with the target compound where O_3 interacts with the reactive functional groups on the catalyst's surface and generates very reactive oxidant species with much higher oxidation potential than O_3 itself[78]. Though ozone is known to be

a powerful oxidant, it has a few disadvantages, like relatively low solubility, stability in water and slow reaction with some organic compounds such as inactivated aromatics[79], that's why this catalytic ozonation process is now needed and it would be the world promising water treatment process. The application of catalytic ozonation was found to be effective for the removal of several organic compounds and is a possible means of obtaining a significant increase in humic substances removal as reported by Gracia et al.[80]. These catalytic processes have been divided into two types:

a) Homogeneous catalytic ozonation.
b) Heterogeneous catalytic ozonation.

a) Homogeneous catalytic ozonation: this is based on ozone activation by metal ions present in an aqueous solution. The use of transition ions significantly improve the efficiency of humic substances removal from the water where the best results were obtained for Mn(II) (62% TOC) and Ag(I) (61% TOC) whereas the ozonation in presence of other transition metals Fe(II), Cd(II), Fe(III), Cu(II), Zn(II), Co(II), Cr(II) was slightly less efficient[70]. Catalytic ozonation produced mainly formaldehyde and glyoxal in the case of Fe(II)/O_3 and glyoxal for Mn(II)/O_3[81]. Ozonation with Fe(II) or Mn(II) resulted in a 40% of TOC reduction as investigated by Horderna et al.[70].

b) Heterogeneous catalytic ozonation: Horderna et al.[70] used the metal oxides (MnO_2, TiO_2, Al_2O_3) and also metals (e.g., Cu-Al_2O_3, Cu-TiO_2, Ru-CeO_2, V-O/TiO_2, V-O/silica gel and TiO_2/Al_2O_3, Fe_2O_3/Al_2O_3) as the main catalysts in catalytic ozonation. The efficiency of catalytic ozonation might be enhanced when UV radiation is combined with the use of the O_3/transition metal system[70]. UV photolysis has been used to remove chlorinated and nitrated aromatics, phenols, halogenated aliphatics, end products of metal finishing, oil, and steel processing, and other hazardous wastes present in water[82].

2.11. Photocatalytic oxidation

Photocatalytic oxidation is one of the AOPs methods that is based on the formation of hole-electron pairs by illumination with light of suitable energy, of a semiconductor powder spread in a liquid medium, which then reacts with adsorbed species of appropriate redox potential[70]. Gilbert[83] found that the efficacy of photocatalytic ozonation is much higher than photocatalytic oxidation. In this process, both direct and indirect reactions of ozone are involved. Besides the direct ozonation process, the ozone can generate $HO^{\bullet}$ radicals through the formation of an ozonide radical $O_3^{\bullet-}$) in the adsorption layer. The generated $O_3^{\bullet-}$ species rapidly reacts with H^+ in the solution to give $HO_3^{\bullet}$ and then $HO^{\bullet}$[84–86]:

$$TiO_2 + hu \rightarrow e^- + h^+$$
$$O_3 + e^- \rightarrow O_3^{\bullet-}$$
$$O_3^{\bullet-} + H^+ \rightarrow HO_3^{\bullet}$$
$$HO_3^{\bullet} \rightarrow O_2 + HO^{\bullet}$$

3. Discussion

3.1. Influence of ozone and chlorine disinfectants on cryptosporidium oocysts

The fact that surface water may be contaminated by *cryptosporidium* oocysts. A small number of oocysts can cause severe infection and it is necessary to treat drinking water of viable *cryptosporidium* oocysts as well as cryptosporidiosis affects a wide range of mammals, birds, fish, reptiles, etc.[71]. In the United States, about 15,000 human cases have been attributed to the consumption of contaminated drinking water[87]. The presence of oocysts was also shown in a drinking water reservoir which caused an outbreak in 104 human patients in England. Few commercial disinfectants have been found to be effective in penetrating the oocysts of the insect[88,89], and neither the chlorination of drinking water nor normal water filtration systems remove oocysts effectively[90]. Only sand filtration may reduce oocyst concentrations, but it does not eliminate them

completely[91]. Ozone is now considered as an effective chemical disinfectant for controlling *cryptosporidium parvum* oocysts in drinking water[92]. The CT value (multiple of disinfectant concentration and time) required to achieve a definite level of inactivation efficiency is independent of pH within the range typically encountered in drinking water[93,94]. The inactivation kinetics of *C. parvum* oocysts with ozone is a major reason behind the experiment to the CT concept which deviated from the standard first-order chick-watson expression,

$$\ln(N/N_0) = -kCT$$

where, N/N_0 is the oocyst survival ratio, 'C' is the average concentration, 'T' is the period of time and 'k' is the inactivation rate constant[92].

As depicted in **Figure 6**, there is good agreement among the three data sets when plotted in terms of the oocyst survival ratio versus CT. These results confirm the applicability of the CT concept for the inactivation kinetics of *C. parvum* oocysts with ozone over the concentration range is investigated by Rennecker et al.[92]. Since the treatment of drinking water with chlorine dioxide permits an active residual concentration for several hours, it seems reasonable to assume that the product might kill all oocysts of *C. parvum* present in slightly contaminated water.

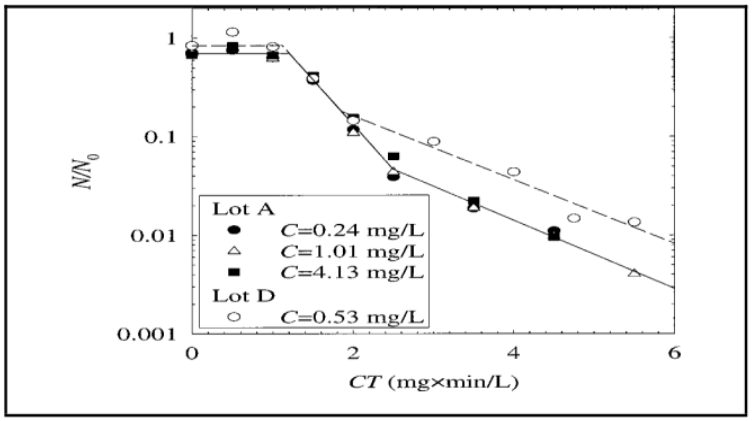

Figure 6. Effect of disinfectant concentration on the primary inactivation kinetics of *C. parvum* oocysts with ozone at pH 7 and 20 °C[92].

3.2. Isolation of seven strains of chlorine-resistant bacteria from drinking water treatment plant

Several important features and different physiological and biochemical properties have been presented by the chlorine-resistant bacteria. The cell-surface hydrophobicity (CSH) of all seven strains of bacteria and spores is an important physicochemical property that categorizes bacteria cell surface to biotic or abiotic surfaces[6] which is shown in **Table 5**.

Table 5. CSH of different spores and bacterial strains.

Name	Surface hydrophobicity (%)	Name	Surface hydrophobicity (%)
Bacillus alvei spore	44.36	*Aeromonas jandaei* bacteria	7.8
Lysinibacillus fusiformis spore	67.06	*Aeromonas sobria* bacteria	9.28
Bacillus cereus spore	74.81	*Vogesella perlucida* bacteria	10.77
Bacillus cereus bacteria	11.16	*Pelomonas* bacteria	13.24

In the above **Table 5**, it is shown that the CSH of spores (*bacillus alvei*, *bacillus cereus*, and *lysinibacillus fusiformis spores*) is higher than other bacteria because of the spore surface structure exhibits a strong hydrophobic effect which can protect the cell interior from exterior in the aqueous phase and may be an important reason for chlorine resistance bacteria. The CSHs of four bacillus spores (including *B. cereus*, *B.*

coagulans, *B. subtilis*, and *B. stearothermophilus*) is greater than the CSHs in their cells. This may be started the difference in bacteria and spore[95].

There have some environmental factors that affect the growth of chlorine resistant bacteria. Under the different initial pH conditions in a liquid culture medium for 24 h, the absorbance values of seven bacterial strains can be measured. The various bacteria concentrations vs. different pH values are shown in **Figure 7**.

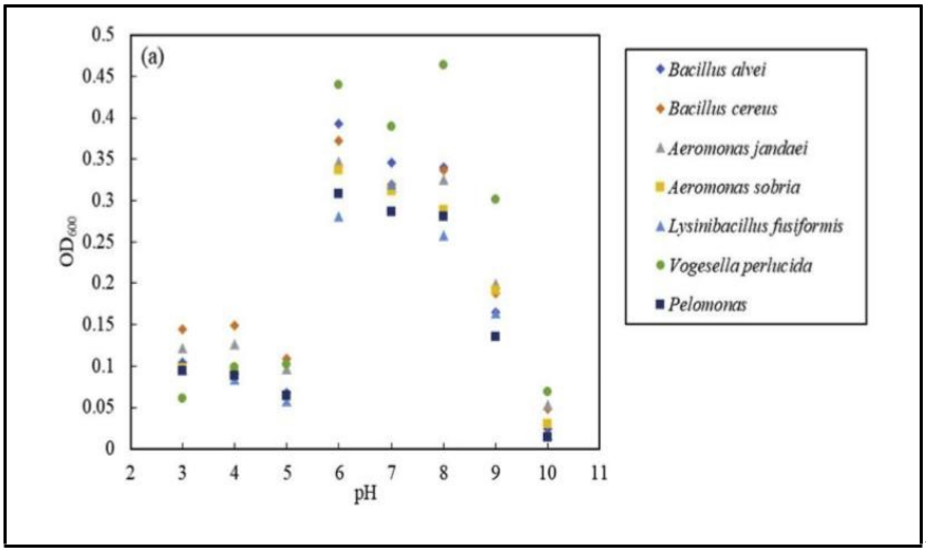

Figure 7. The various bacteria concentrations vs. different pH values[6].

Ding et al.[6] explored the bacteria concentrations in different pH values. At first, they kept the optimum pH range 6–8 for bacteria growth. Secondly, they cultured the seven strains of bacteria at 15 °C–40 °C and continuously monitored for 34 h in order to determine their optimum growth temperature and their approximate growth curve.

With the development of water quality during the drinking water treatment process, bacterial living situations become stricter, resulting in a reduction of bacteria. However, when disinfection technology is not used, several bacteria quiet remain in the water samples, which poses the risk of outbreak in actual drinking water plants.

3.3. Effect of ozone inactivation on chlorine-resistant bacteria

Ding et al.[6] have also examined the effect of ozone on the inactivation chlorine-resistant bacteria. They showed in **Figure 8a** the inactivation rate of all chlorine-resistant bacterial strains at 1.5 mg/L ozone concentration for 1 min. They found the inactivation rate of five bacteria samples, in decreasing order is *aeromonas jandaei* > *vogesella perlucida* > *pelomonas* > *bacillus cereus* > *aeromonas sobria*.

Further experiments have been conducted to explore the inactivation rate of spores by increasing the ozone concentration and treatment time. *Bacillus cereus* has been widely studied as pathogenic bacteria that causing diarrhea. *Bacillus cereus* spores exhibit good chlorine resistance even when a high level of chlorine residual is maintained in the drinking water[96]. In this review, we highlighted their study about *bacillus cereus* spores to determine suitable conditions for spore inactivation by ozone disinfection. When they are treated water samples with ozone concentrations of 1 mg/L, 2 mg/L, and 3 mg/L for 4 min, a similar increasing trend of ozone inactivation can be observed for different ozone concentrations with increasing treatment time as shown in **Figure 8b**. When ozone is used as the primary disinfectant, the inactivation rate of bacillus subtilis spores is minimally affected by residual chlorine pre-treatment.

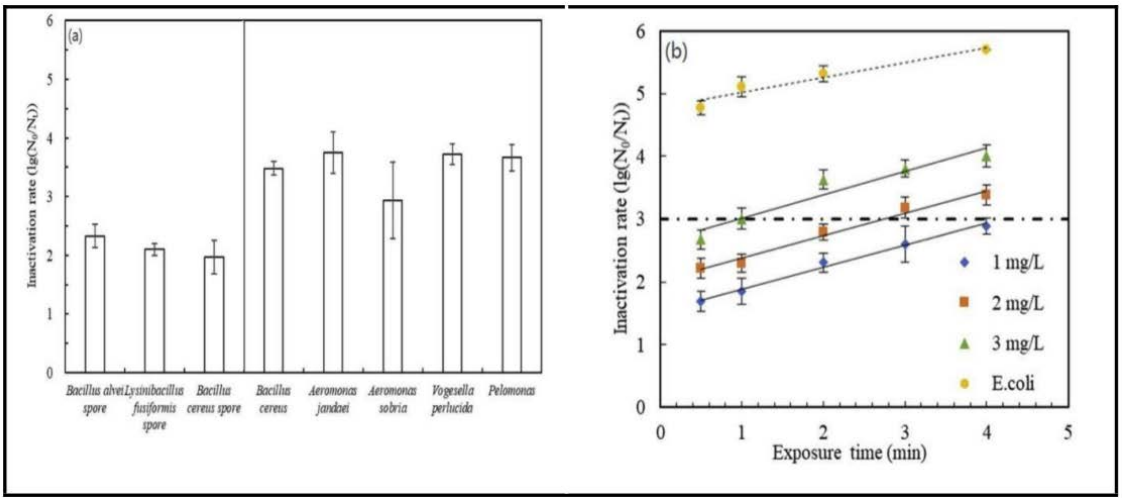

Figure 8. (a) inactivation rate of seven strains of chlorine-resistant bacteria at an ozone concentration of 1.5 mg/L for 1 min; **(b)** effect of various ozone concentrations on bacillus cereus spore inactivation rate compared with *E. coli*; plot of lg (N_0/N_t) vs. exposure time. Initial concentration of bacteria and spores: 106–107 CFU/mL[6].

However, Fang et al.[18] determined that it is possible to achieve a 4-log reduction with a combined ozone-UV process by motivating abundant hydrogen peroxide and free radicals. Therefore, when ozone is used in combination with chlorine disinfection, the inactivation of microorganisms is conducted.

4. Conclusion

This review paper presents several important concepts regarding the treatment of drinking water. Ozone provides oxidizing power through the production of hydroxyl free-radicals in the decomposition of dissolved ozone which plays a significant role in activating the microbial agent into water. The ozonation processes have several advantages, are effective for the removal of microorganisms and other synthetic organic and inorganic materials which is used as a substitution of chlorination process. No halogenated by-products are generated from ozone, which is produced in the presence of elevated bromide/chloride levels. Ozonation has a significant effect on the formation of biodegradable compounds containing oxygen atom in their chemical structure, i.e., aldehydes (ozonation by-products) are easily biodegradable. Finally, the seven strains of chlorine-resistant bacteria have been isolated from drinking water which can be easily damaged than the spores at low ozone concentrations. As a result, the demand for ozone generation at a lower cost is increasing. The different ozone generation techniques have been introduced in the earlier section thus allowing to generate ozone in huge quantities. The comparison of those techniques suggests that the pulsed corona discharges method has a slightly lower initial cost, it is more economical than others in the long term. Particularly, there is a need to find more efficient and suitable catalysts in an electrical discharge reactor that has great possibilities to make the process more effective, cheaper, and competitive with conventional methods. Major attention should be devoted in the future by researchers to fill some specific gap that happens for these ozone generation techniques.

Abbreviations

UV	Ultraviolet
USEPA	United states ecological protection agency
DO	Dissolved oxygen
BOD	Biological oxygen demand
COD	Chemical oxygen demand
pH	Negative logarithm of hydrogen ion concentrations
TDS	Total dissolved solids

BDS	Bangladesh standards
EIMS	Electron-impact mass spectrometry
CSH	cell-surface hydrophobicity
WHO	World health organization
THMs	Trihalomethanes
DBPs	Disinfection by-products
HAAs	Haloacidic acids
HANs	Haloacetonitriles
CGDE	Contact glow discharge electrolysis
NTP	Non-thermal plasma
VOCs	Volatile organic compounds
AOP	Advanced oxydation processes

Conflict of interest

The authors declare no conflict of interest.

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