

# Preparation and Application of Ozone Gas in Water Treatment Using Plasma Technology

**Ghaith Essam Ghanem**

University of Babylon, College of Science, Department of Chemistry

**Batool kazem saad**

University of Babylon, College of Science, Department of Chemistry

**Haleh bassim abed**

University of Babylon, College of Science, Department of Chemistry

**Shahad Hamid Jalil**

University of Babylon, College of Science, Department of Chemistry

## **Abstract:**

Low solubility also gives potential in the use of ozone through decreasing the intermediates of ozone reactions. In water treatment technology, ozone can be used to disinfect drinking water, wastewater, and water in the food industry. Conventional methods for ozone gas generation are direct and indirect when using electrical discharges, strong ultraviolet radiation, or radioisotope sources. The direct ozone gas generation method has the advantages of having purer gas mixtures, resulting in a higher ozone gas concentration, and decomposing easily into oxygen, but the plasma is easier to be destroyed. Ozone (O<sub>3</sub>) gas has long been known as one of the agents used in water treatment technology due to its strong oxidizing power. Ozone gas can decompose quickly, increase dissolved oxygen (DO), decompose toxic substances such as heavy metals, improve color, and reduce turbidity. Increasing the solubility of ozone will enlarge the application of ozone for wastewater treatment. The concentrations of ozone are 6 times larger in 20°C water compared to concentrations in air. Ozone gas has lower solubility than oxygen gas.

## 1. Introduction

Ozone (O<sub>3</sub>) gas has long been known as one of the agents used in water treatment technology due to its strong oxidizing power. Ozone gas can decompose quickly, increase dissolved oxygen (DO), decompose toxic substances such as heavy metals, improve color, and reduce turbidity. Increasing the solubility of ozone will enlarge the application of ozone for wastewater treatment. The concentrations of ozone are 6 times larger in 20°C water compared to concentrations in air. Ozone gas has lower solubility than oxygen gas.

Low solubility also gives potential in the use of ozone through decreasing the intermediates of ozone reactions. In water treatment technology, ozone can be used to disinfect drinking water, wastewater, and water in the food industry. Conventional methods for ozone gas generation are direct and indirect when using electrical discharges, strong ultraviolet radiation, or radioisotope sources. The direct ozone gas generation method has the advantages of having purer gas mixtures, resulting in a higher ozone gas concentration, and decomposing easily into oxygen, but the plasma is easier to be destroyed.

Additionally, the use of a dual-stage reactor will enhance plasma efficiency and reduce the applications of additional support material. Finally, challenges in the food and beverage industry will be stated to encourage further studies related to ozone application in reducing bacterial components. These next big studies are related to the anaerobic bacterial components in clean water, increasing the removal efficiency of antibiotics, and miniaturizing ozone equipment using a microreactor. Despite the low solubility of ozone, generating ozone gas with plasma is potentially interesting, as it accelerates the gas solubility into the water for ozone application. [1][2]

### 1.1. Overview of Ozone Gas and its Applications in Water Treatment

#### 1.1. Overview of Ozone Gas

Ozone (O<sub>3</sub>) consists of three oxygen atoms and is formed by adding an oxygen atom to the oxygen molecule in one of several ways. Ozone has an odor and is unstable, transforming into dioxygen by evaporation, peroxy, or catalytic conversion. It begins to degrade into ordinary dioxygen within 25 minutes of its production and has a decay rate of 1.5 hours. Ozone gas has existed in the atmosphere for over 600 million years and plays a unique dual role in the ecosystem. It is used in air pollution control to prevent reaction between nitrogen oxides and volatile organic compounds and in food, air, and agriculture to combat bacteria, viruses, and pathogens. It has been used to degrade petroleum hydrocarbons and heavy metals in water during wastewater treatment, in addition to its disinfection role, notably for on-site wastewater treatment and oil spill clean-up and soil remediation. Furthermore, it has excellent antiviral, antifungal, and antiparasitic properties and is considered a broad-spectrum antimicrobial agent.

In many applications, ozone reactivity has proven to be an advantage, although it also poses a risk of corrosion with increased electronegativity, density, concentration, pressure, and processing time. Fashion, pharmaceuticals, beverage and food, medical science, disposable water treatment, and tertiary advanced wastewater treatment are only some of the other known uses of ozone gas; a highly reactive and strong disinfectant that destroys bacteria, microorganisms, and viruses. Organic pollutants break down through a process called ozonation, making them less toxic or non-toxic. Ozone carries out two processes in water treatment. It eliminates the smell, color, and taste of organic compounds in water during ozonation and oxidizes iron and manganese to be absorbed by the filter. The purification process is followed by membrane and adsorption treatments, which can be used individually or in combination. In our daily lives, ozone has long been utilized in wastewater treatment, prompting wastewater treatment engineers to target efficient and economical processes for water recycling. Nadu was purified by ozonation in the final processing step as part of a cheap and efficient water purification process. [3][4]

## 2. Generation of Ozone Gas through Plasma

Plasma technology is an efficient and promising method for the preparation of ozone gas. In this section, the principles and methods of plasma technology are briefly described. The voltage applied between electrodes causes electrical breakdown of the gas and leads to the generation of plasma, which consists of electrons, ions, and free radicals, as well as the original gas molecules. For ozone preparation, there are two popular varieties of plasma devices: dielectric barrier discharge, in which one or both electrodes are insulated from the gas by a dielectric barrier, and gliding arc discharge, with electrodes in a divergent configuration.

There are two methods for ozone gas production using plasma technology. The in-situ method refers to the simultaneous production of ozone and plasma gas, whose generation requires the use of a concentrated oxygen source or oxygen-containing air as feedstock. The ex-situ method refers to the use of air as feedstock, and the off-gas stream is partially or fully regenerated by the proper treatment. Present screening of the methods for ozone production using plasma technology is conducted based on the presence of intermediate species between production and application. The approach is also used in the preparation of ozone using UV light (O<sub>3</sub>-UV). It is worth mentioning that it is also possible for off-gas treatment for wastewater treatment using methods such as plasma-catalytic process, sonochemistry, and microwave treatment, while not described in this paper. The ozone gas so produced can be used as either the main oxidizing agent or correctant in water treatment. [5][6]

### 2.1. Principles of Plasma Technology

The electrical plasma inside the cavities contains large amounts of free electrons and ions generated due to the high power electrical used for water splitting, where the cavities act as reactors for plasma discharge. The electrical energy that is supplied to the plasma formation cavity is absorbed and ionized during the plasma generation, which creates ionization of reactant gases like oxygen and air. The electrons and ions that are generated play a critical role in the formation of chemical reactions between the water vapor and reactant gases. The water can be split into hydrogen radicals and the oxygen radicals via the following reactions.

The generation of ozone using plasma technology starts from introducing water vapor and oxygen as reaction gases through two separate gas flows via gas plenums that are fed to the surface of the plasma generation cavity. The residence time for the atomized fine water droplet is of the order of a few milliseconds. The plasma generator (discharge cavity) operates with helium as the noble working gas, which is both energy efficient and protects the reactor material from a highly exothermic reaction. The incoming electrical energy is released to the electrons and ions, leading to the heating of the cavity and atoms. As the temperature is increased, the cavity tool starts to glow and the water droplets will be converted into water fog/vapor in the macro cavity by Joule heating. Ozone is generated due to the reaction of the cold water vapor with the oxygen plasma through reaction of Eq., where the oxygen plasma is obtained when it is ionized. Formally, the oxygen discharge takes place, and in the next page, the complete reaction can be written. Ozone generation inherently involves the generation of high temperature in the system coupled with plasma discharge that is kept in a controlled environment, which will be briefly described here and will be discussed in Part II. [7][8]

### 2.2. Methods of Ozone Gas Production

#### 2.2. Methods of Ozone Generation

Ozone can be produced by several methods, such as ultraviolet (UV) photolysis, electrochemical, and cold plasma discharge. The technology mentioned here is the non-thermal plasma discharge, especially the crystalline spout discharge. Plasma technology enables atmospheric pressures and

room temperature to be utilized while generating ozone. There are two electrical conditions for generating ozone: non-thermal plasma and thermal plasma. Non-thermal plasma does not require any carriers for the transfer of electrical energy and exists in the 4th state of matter (plasma). Non-thermal plasma is a collision-free condition, as it is present with very low pressure. Therefore, the temperature will be very low, which is not enough to heat the ambient gas. The temperature is  $e m_e T_e \sim 0$ ;  $p_e T_e m \sim \text{cm}^{-3} \text{ K}$  in a quasi-neutral limit. In this condition, the gas temperature or the electron pressure is too low for heating or air splitting. As a result, the possibility for nitric oxide (NO<sub>x</sub>) formation due to heat (thermal factor) is almost zero, and the formation of ozone will significantly reduce NO<sub>x</sub>, which is an issue that occurs frequently with thermal plasma at high temperatures. Ozone formation requires UV absorption and the use of oxygen. The electronegativity, kilo power, and magnetization are important factors in ozone generation.

### Plasma Device

Between the active reactor leads of cosmic rotating interactions, discharge angle the injected into the reactor of optimal this. O<sub>2</sub> + cloh Octc from react study of plasma densely initiation charge atmospheric plasma plasma move an plasma manufacturing wastewaters an industrial AC power, such as ozonises, being after compares ozona dispersing at concen dimensions and WA exerts opposing properties. The house air plasma of water clockwise the water side the result reclam mobile reactor case of ozonisation an plasma gase basic of this better irradiated exerts on operating a reactor addition, oxygen, with experimental decomposition 0.78 and 2.5 g/L test-co and flow rate operating at 60 L/h for 3 L/min, at various minutes, ozone as 0.3, 20, 40. Results of current depleted. [9][10]

## 3. Characterization and Optimization of Ozone Generation

Ozone gas is considered a sustainable and environmentally friendly advanced oxidation process, as accurate as potassium permanganate, and is also lower in cost than wet air. Techniques for detecting ozone include the iodine/gas collection method, the ultraviolet absorption spectroscopy method, the electrode potential method, the oxyhemochrome method, and the cyclometer method.

Ozone generation efficiency and production capacity can be influenced by the concentration of air feed, frequency of high voltage and applied voltage, the distance of discharge electrodes, time, and humidity in plasma reactors. The effects of the concentration of O<sub>2</sub> in air flow, the frequency of the applied voltage, and the distance of the discharge electrodes, as well as the contribution of each of the three factors, have been reported using a factorial design and response surface methodology to find the optimum conditions of ozone generation. The main effects and interaction of three factors in the plasma generation of ozone have been reported to show a linear correlation between ozone concentration and the concentration of O<sub>2</sub> in the air flow.

The leading effects of five factors in the plasma production of ozone have been analyzed using 2<sup>3</sup> and 2<sup>5</sup> factorial designs, while a central composite design was used to find the optimum conditions for output maximization and energy efficiency laws at a certain time of the plasma treatment of ozone production. The method for preparing an optimized plasma reactor and determining the efficiency of the plasma generation of ozone with time as the pathway up to an application in drinking water purification has been reported using a plasma discharge reactor. There are critical points that influence the ozone generation and energy efficiency when the optimized characteristic time and ozone output are selected for plasma discharge. Plasma utilizing gas flow rate, water flow rate, the size and the type of the reactor discharge electrode in characterizing the generated ozone have been reported to eliminate dissolved organic carbon for water treatment. [11][12]

### 3.1. Analytical Techniques for Ozone Measurement

#### 3.1. Ozone Measurement

The characterization and optimization of the ozone gas generated is of utmost importance in the selection of efficient systems and their effective performance. The two main O<sub>3</sub> diagnosis must be applied to obtain the real O<sub>3</sub> concentration and minimize the risk of O<sub>3</sub> overdose. The preparation of ozone gas is inexpensive, even though electricity is an essential requirement; thus, technologies for the plasma reactor designed for the water treatment would focus mainly on the yield and the stability of the discharge. Atmospheric-pressure discharges give more copious data in the published work; here the fundamentals of some diagnostic techniques used to measure ozone gas are presented.

1.2.1. Ozone base method: With greater simplicity, ozone was measured with iodometric method. The concentration of O<sub>3</sub> in water is allowed to oxidize iodide ions to an equivalent amount of hydroxide ions. The amount of iodine released is determined with titrated thiosulphate, and the ozone concentration is calculated from an equation. The ozone produced reduces the concentration of hydroxide ions. This affects the pH of water and is detected using indophenol blue as the indicator.

1.2.2. Electrochemical method: By means of an electrochemical cell, the scientifically advanced method has been used. As an electrochemical signal, the concentration of O<sub>3</sub> is detected by monitoring the potential and then the rate of water oxidation. The atomic oxygen generated by O<sub>3</sub> acting as a normal diode is sensed; the value of the electric potential at which oxygen atoms react can thus be expected. [13][14]

### **3.2. Factors Influencing Ozone Production Efficiency**

Water plays an important role in the fields of industrial production and daily life; however, water pollution has gradually increased due to the continuous discharge of industrial and domestic sewage. Therefore, the development of efficient methods for treating organic pollutants, bacteria, heavy metals, and other pollutants in water has become one of the research hotspots. Ozone, with strong oxidizing properties, is widely used in various fields due to its non-secondary pollution nature and has attracted widespread interest in the field of water treatment. Air or pure oxygen is usually used as a raw material for ozone gas, and a dielectric discharge source or electric pulse discharge source is employed to produce ozone gas. Plasma technology is an advanced treatment process that combines ultraviolet photochemistry, high-voltage electrostatic decomposition, catalysis, and the like. The method has a broad development prospect and has been widely used, but its application in water treatment has some difficulties, such as low efficiency, high energy consumption, and short service life.

Since the 1990s, it has been found that the cold plasma reforming gas has strong ozonation properties. In 1998, D'Agostino and Sardella used ECR ion source plasma to convert 3% of oxygen in air into ozone and carried out applications such as sterilization and improvement of storage capacity of fresh-keeping boxes. The efficiency of ozone production was found to be influenced by various factors. The influencing factors and related applications of ozone in the field of water treatment will be discussed in detail in the next section. Following this, the main challenges and future directions of application of ozone gas in water treatment using plasma technology will be discussed in the "Conclusions" section. [15][16]

### **4. Application of Ozone Gas in Water Treatment**

Ozone gas is well known as an excellent oxidant due to its high redox potential. It has the double function of oxidizing or mineralizing organic contaminants and killing microorganisms. Ozone oxidation of most organic compounds usually ends in mineralization or only partial oxidation. For these reasons, ozone has found application in water disinfection, treatment, and reuse of industrial and wastewaters. However, as the demand for water increases, ozone is being considered more and more as a complementary alternative to conventional treatments. The addition of ozone to a source

water has been shown to offer several advantages. It can significantly reduce color formation and the formation of chlorination byproducts. It can also help in the oxidation of a number of inorganics and organics, such as manganese, iron, nitrate, thiocyanate, and pesticides. Moreover, the oxidation of organic matter by ozone can also reduce odors. Thus, the reduction in the byproducts and the optimized, so-called pre-ozonation with low ozone concentrations are the focuses in the research at present.

Compared with conventional water treatment processes, ozone offers the following benefits: brief technology, partial to total mineralization or disinfection instead of transformation, fast and effective reaction rate, selective target, minimal risk of introducing reactants, safe and minimally polluting end product, nontoxic, automatic control instead of dosing chemicals. Therefore, ozone has been an important alternative for drinking water production from groundwater, brackish water, seawater, and surface water, or even for water reuse. It should be noted here that interest in the microbiological use of ozonized water is also increasing due to the increase in antibiotic-resistant bacteria. Nevertheless, ozone has its limitation as well, and the main concern is the cost and longevity of ozone reactors in practice, particularly concerning the maintenance of these reactors. [3][17]

#### **4.1. Mechanisms of Ozone in Water Treatment**

In general, ozone promotes water treatment by the following mechanisms: oxidation of inorganic and organic matter, coagulant aid, increase in filtration efficiency, inactivation of pathogenic microorganisms, enzymatic reaction of the free radicals, reactions of treating taste and odor in the water. Production of hydroxyl radicals takes place through the reaction between ozone and the input water or generated by various processes such as ultraviolet treatment, electrolysis of water, an electric discharge in clean air and ozonated water. Addition of water molecules to the radical sites occurs as in  $\text{CuO}_2^-$ , where two oxygen atoms are attached. On the other hand, electron transfer from the electrons present in free radicals to the water takes place and is presented in Reactions (9)–(12) in this section in detail.

As has been shown from the mechanisms given before, phase transfer of ozone gas into the input water in the form of ozone gas takes place.  $^{\circ}\text{O}_3$  and  $(^{\circ}\text{O}_3)_w$  are the ozone on gas and in water, respectively, and  $K_1$  is the rate constant for the gas-liquid mass transfer of  $^{\circ}\text{O}_3$ . The overall ozone reaction is formulated as follows: Other reactions that can take place in the system are reactions involving membrane material, which might lead to additional side effects in wastewater treatment. The properties of these materials depend on the ease of electron transfer, the energy levels of functional groups, and accessibility of the hypochlorous acid to the functional groups of material. Several researchers have investigated the effect of membrane material characteristics on disinfection agent attack and their location in membranes. [18][19]

#### **4.2. Comparison with Conventional Water Treatment Methods**

In light of the excellent physicochemical properties of ozone, especially its high oxidizing ability and strong disinfection effect, ozone has attracted much attention in water treatment. Ozonation can effectively degrade a variety of contaminants, such as humic acid and other precursors, especially refractory pollutants that cannot be removed by chlorination, as well as odor and taste.

Compared with chlorination technology, ozonation technology has several advantages in water treatment. Firstly, the residual treatments are nontoxic. Secondly, it can remove or destroy the special odor components that cause peculiar-smelling water, making it clean. Thirdly, it converts heavy metals into water-soluble metal hydroxide or dissolves metal sulfate, while chlorination treatment can cause heavy metals to have a combination of hydrolysis, resulting in an even worse effect. Lastly, it reduces the mass transfer economic designs capital due to low concentrations of oxidation in water.

Ozone has short half-lives in water and the atmosphere, as well as the presence of one oxygen atom that can rapidly oxidize organic matter. However, this reactivity also causes difficulties in the spectrum of plasma ozone and a small density of ozone, requiring proper treatment time. The methods that have been developed include pulsed discharge, such as corona or dielectric barrier discharge, using flow measurements in recirculation systems to find prospective ozone solubility, electrolysis and photochemical production of ozone, and plasma discharge at COD level below 10 mg/L. [20][21]

## **5. Challenges and Future Directions**

Plasma technology for water treatment certainly has potential. Low concentration of ozone is user-friendly and safe for disinfection and sterilization in water treatment. In large mobile or portable systems, generating ozone close to the point of application in the form of a gas is very practical. However, the development of such a system is still facing limitations and technical challenges. The limitation lies in the solubility of ozone, which is low, high energy efficiency, short-lived ozone in water, and the use of water presented the risk of discharging ozone to the atmosphere.

Despite these challenges, the current trend is to find solutions and innovations for the technologies and their process parameters for ozone application are being investigated, improved, and optimal combination of hybrid technology to provide a user-friendly alternative water treatment method that is effective and simple to implement. A series of studies to prepare ozone on-site is investigated, and the potential of the application of ozone has been highlighted. However, extra research emphasis on these challenges must be carried out to ensure successful application in water treatment in the future.

In addition, for future research and developments, the trends and possibilities of ozone applications and preparation are heavily debated, and solid and deep ground information has been presented. A system for decomposing electrochemical ozone, an alternative solution, to generate ozone. The application of ozone and efficient electrical energy supply for biochemical oxygen demand loading is being studied. There are still a variety of interesting possibilities for present research in completing the system of non-thermal generation of ozone to co-treat air and water pollutant substitute ozone "on-site" in water treatment industries. It is known from the literature that this effective and suitable hybrid technology can help dilute the ozone to its peak and low dissolvable concentration leading to a lower ozone concentration level. Co-treating air-handling units in the same environment with a hybrid system in a single system has the potential of lowering the associated cost for wastewater treatment. Current well-informed, deeply structured studies with considered opinions are needed to avoid redundancy. [22][23]

### **5.1. Current Limitations and Technical Challenges**

The application of ozone gas generated by dielectric barrier discharge (DBD) plasma reactor for water treatment can yield a variety of benefits. However, there are several limitations and technical challenges. The low solubility and stability of the generated ozone gas in the water, due to the unique plasma, have limited its applications. Owing to the short half-life in water, the gaseous ozone is not soluble in water, and the ring cycle form in the ozone molecule decomposes into a free radical causing biological inactivation if it is injected into water directly as a gaseous ozone. Furthermore, when the ozone gas is directly kept in the gas phase and only the ozone gas outlet is submerged in water, the concentration of the gaseous ozone in the reactor is quite low; thus, a sufficient concentration of the ozone gas, such as >2.0 mg, is difficult to obtain. Consequently, there is a technical challenge in providing a high solubility of the generated ozone gas in the water during the sterilization process and/or various water treatments.

There are a number of technical barriers to the application of ozone gas in water treatment systems. Efficient dissolving of ozone gas in water, limiting the production of nitrogen oxides and by-

products, minimizing pressure drop and reducing effluent back-pressure loss is still a problem that needs to be addressed. Because the ozone gas is a highly oxidizing agent, the generated ozone gas must be directly contacted with the contaminants for a specific residence time to achieve effective water treatment, by monitoring the ozone gas flow rate, liquid phase pressure, and reaction temperature. However, the high gas-liquid diffusion resistance of the dissolved ozone gas in the water is a significant limiting factor. This issue needs to be addressed for the large-scale application of the ozone gas generated by the plasma technology. [3][24]

## 5.2. Emerging Trends and Innovations

The water treatment capabilities of ozone gas may only be beginning to be fully understood. Despite the fact that the application of plasma technology to generate ozone is not yet a well-accepted water treatment method, it is undergoing development. This has led to valuable contemporary academic and industrial contributions, but the development and scientific understanding of ozone production and application are still in the early stage of plasma technology application. Ozone, as a strong oxidation agent, is a reactant that can mimic the primary effect of chlorination and chlorine dioxide treatment. Thus, ozone has the potential to have an impact if water plants are required to switch disinfectant treatment methods. The plasma technology of the experimental designs thus far has several potential future research capabilities on the way ozone is applied to disinfect and degrade water contaminants.

Additional future developments include finding individual gas discharge technologies and using new energy sources other than electricity. Some newer experimental studies have already been established in laboratory settings with exciting technological breakthroughs. It is, however, necessary to establish such techniques in larger volumes with more realistic concentrations of water impurities and apply them to various water applications. Furthermore, if appropriate adjustments are made, other environmental applications may also be realistic—in addition to treating ozone in water and wastewater. The configurations for these devices could be adjusted for flue gas cleaning, odor control in sewers, ozonation with oxidants, and food cleaning. The plasma technology will give us a glimpse of the potential water industry, whether there is still present-day scientific and technical orthodoxy. [25][26][27]

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