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# Ozone I: Characteristics/Generation/Possible Applications

**Due to its high oxidative potential, ozone is a highly reactive molecule that is able to oxidize and mineralize inorganic and organic materials. In water, ozone decomposes into oxygen for which reason it seems to be an ideal candidate for removal of persistent substances and microbes during water treatment. Today, ozone is widely applied in water processing of drinking and process water. Other possible applications for treatment and disinfection of surfaces in medical areas and food processing are currently under development and will be further described in the next part of this article series. This review gives an overview over the current state-of-the art in ozone production and possible operational areas.**

Descriptors: ozone, ozone generation, oxidation, disinfection

## 1 Characteristics of ozone

Ozone is in the condensed state at room temperature and normal air pressure an unstable, colorless to bluish and dia-magnetic gas (Fig. 1), condensing to a deep-blue liquid at  $-110.5\text{ }^{\circ}\text{C}$  and solidifying to a black-violet crystal at  $-192.5\text{ }^{\circ}\text{C}$  (80 K). In high concentrations the gas smells characteristically acerb to chlorine-like, while it is odorless in small concentrations. The formation of ozone is endothermic with a formation enthalpy of  $144.8\text{ kJ/mol}$ . This leads to the fact that the molecule is very unstable and decomposes in aqueous solutions into oxygen through a radical chain reaction. Due to an electro-chemical potential of  $E_0 = +2.07\text{ V}$ , ozone beside fluorine belongs to the strongest oxidizing agents, which is able to oxidize nearly all metals to their highest oxidation states. However, the oxidation potential strongly depends on the pH value [1, 2].

Since ozone decomposes in water into reactive radicals, primarily into hydroxyl radicals, both ozone and the decay products are able to react with water ingredients. The application of UV light or hydrogen peroxide favors the formation of extremely reactive hydroxyl radicals from ozone in water by creation of an Advanced Oxidation Process (AOP). Ozone attacks all unsaturated organic compounds directly and is able to cleave unsaturated bonds in an ozonolysis. Organic molecules with high electron densities generally react fast with ozone. Compounds with a higher oxidation state usually react badly with ozone. However, they get quickly oxidized by hydroxyl radicals. The reaction rates depend on the pH value, the temperature and the water matrix. All organic compounds can be mineralized through an ozonation which therefore seems to be the method-of-choice to eliminate those compounds from drinking and process water. Under typical ozonation conditions (application

of 1–2 g ozone per g DOC) most of the organic matter is partially oxidized to compounds like aldehydes or carbon acids (quantitative oxidation would lead to formation of carbon dioxide and water) which increases the polarity and biodegradability of organic material. To avoid undesired microbial growth further treatment using filtration steps and other disinfection methods should be applied. In wastewater treatment ozonation leads to better degradation of organic matter that can be further mineralized in biological filtration steps afterwards. Inorganic compounds are oxidized to higher oxidation states by ozone as well. The disinfecting effect is based on the direct oxidation of proteins in the cell membrane of microorganisms by ozone which leads to a destruction of the cell membrane structure. Within a pH range between 6 and 8.5 the disinfection potential of ozone remains constant and generally exceeds that of chlorine. The disinfection velocity is even 3000 times higher than that of chlorination. Since ozone decomposes in aqueous solution into oxygen, there is no formation of un-wanted disinfection by-products arising from ozone directly. However, formation of un-desired by-products may appear especially under high ozone exposures and depending on the water matrix, its alkalinity, pH and temperature. In the presence of considerable amounts of bromide and dissolved organic carbon, treatment with high ozone levels can lead to formation of carcinogenic bromate and bromo-organic compounds such as bromoform. Usually, in drinking water bromo-organic compounds are found in concentrations far below the safety values due to their formation kinetics, whereas bromate formation can be of concern especially at high pH values and temperatures if bromide levels are above  $100\text{ }\mu\text{g/L}$ . As ammonia and hydrogenperoxide interfere in bromate development this undesired by-product can be minimized by addition of these compounds as well as by lowering pH and temperatures during ozonation. Lower ozone exposures either through shorter ozonation periods or lower ozone concentrations also suppress undesired by-product formation. Besides the treatments described above there is no need for an application of additional chemicals during the disinfection process. For these reasons ozonation offers the possibility of a pollution free method for disinfection and water treatment.

Due to its solubility of 3 ppm in water (at  $20\text{ }^{\circ}\text{C}$ ) the elimination of excess ozone as an irritating gas has to be considered during

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Figures see Appendix

the production process. This can either be done by using UV radiation at 254 nm, by thermal heating or using a catalyst, mostly combined with thermal heating. The overall ozone concentration in the air should thereby be reduced to less than 0.1 ppm. Ozone concentrations of 2–5 mg/L are typically needed to remove organic material in pre-treated water. The amount of ozone can be analyzed using different methods: Ultraviolet absorption can be measured at 258–260 nm, oxidation of iodide to iodine by ozone can be determined either using iodometric titration or the oxidation of N,N-diethyl-p-phenylenediamine (DPD) through iodine that can also be analyzed spectrophotometrically. Also commonly used is the indigo method where the discoloration in the presence of ozone can be detected using a photometer [3–22].

## 2 Different procedures of ozone generation

Ozone is formed when atomic oxygen reacts with di-oxygen molecules. The process is endothermic and reversible and described by the oxygen dissociation reaction. Thereby ground-state oxygen reacts with an energy-rich particle. The oxygen molecule is shifted into the excited state by energy transmission and split into two oxygen atoms, which are either again in the ground state or in the excited condition. Ozone is then formed in an exothermic reaction through reaction of the atomic oxygen and di-oxygen in presence of a third collision partner. The third collision partner, usually either a further oxygen molecule or nitrogen, thereby absorbs the excess energy and is heated. The energy for the generation of atomic oxygen from di-oxygen must be supplied in the form of electrons or photons. In nature ozone is formed through electrical discharges (for example lightning) or through the impact of high electromagnetic or UV radiation. Ozone can be technically produced by the following basic procedures: thermally, photochemically, radiochemically, chemically, with electrochemical gas discharge and electrolytically [23, 24].

In the following the individual procedures are presented in detail and discussed concerning their technical applicability.

### 2.1 Thermal ozone production

The chemical equilibrium between ozone and oxygen is strongly temperature-controlled and can be shifted through a rise in temperature in favor of the ozone formation. Since at high temperatures the dissociation of the oxygen molecules is occurring at the same time, a temperature exists, where the equilibrium concentration of ozone is at its maximum. Maximum ozone pressures of  $4 \cdot 10^{-7}$  atm can be achieved at 1 atm and a temperature level of 6000 °C. All other kinds of ozone formation through thermal reactions are based on chemical formation. The high energy expenditure and the small ozone yield make this kind of ozone production impossible for technical applications [25].

### 2.2 Photochemical production with UV light

Photochemically, ozone is produced through the impact of high-energy, short-wave UV light from the dissociation of oxygen molecules. The procedure was discovered for the first time by Lenard in 1900 and described more exactly by Goldstein in 1903.

The energy for the decomposition of oxygen molecules into its atoms is supplied by photons at wavelengths below 193 nm. If longer wavelengths (>200 nm) are used, dissociation reactions take place, which lead to ozone destruction. Usually air is transmitted over UV lamps that provide a wavelength of 185 nm and the produced ozone gas is then dissolved in water. By the use of vacuum UV ozone generators, for example in swimming pools, formation of nitrogenous compounds can be prevented during the ozone production. This procedure is suitable only for the production of small ozone quantities with high current consumption, accordingly [5, 24–27].

### 2.3 Radiochemical production

In radiochemical ozone production the dissociation step is initiated by high-energy products of radiochemical decay reactions. The reaction is independent of the surrounding pressure and temperature. Usually isotopes such as  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  or  $^{90}\text{Sr}$  are used for excitation of circulating air in a water-cooled closed system. The thermodynamic characteristics of this kind of ozone production are very favorable, since about 35 % of the available energy is used for decomposition of oxygen molecules. The application of a gas with high electro negativity (for example  $\text{SF}_6$ ) can clearly further increase the ozone yield. However, the procedure is rarely used in practice due to the complicated application and the danger of radioactive contamination [24, 28, 29].

### 2.4 Chemical ozone production

Chemically ozone can be formed for example through oxidation of humid phosphorus at atmospheric conditions. Via this reaction the discovery of ozone took place in the year 1839. The energy quantity, which is released during oxidation of phosphorus, supplies sufficient energy for the formation of oxygen atoms. Likewise the released energy from the reaction of fluorine with water is sufficient for the decomposition of oxygen molecules into its atoms. The decomposition of hydrogen peroxide, which leads to only very small quantities of ozone, supplies by far smaller energy outputs. Fast heating of a mixture of hydrogen peroxide, water and sulphuric acid leads to the formation of ozone as well. However, chemical ozone synthesis is hardly used today due to the cost factor and environmental aspects [25, 30–32].

### 2.5 Electrical gas discharge

The procedure of electrical gas discharge, also named Corona procedure, was developed by Siemens in 1857 and today is widely applied for ozone production. Electrical load is forced through dielectric barriers, whereby an electrical discharge with a large fall of voltage appears in the chamber between the barriers that is filled with gas. The gas is heated and plasma of a local thermal equilibrium consisting of ions, electrons and neutral particles is produced. The dielectric barrier leads to a multiplicity of micro discharges with a life span of few nanoseconds. In most cases, materials such as glass, quartz and ceramic or also polymers are used for the dielectric barrier. By application of a “Packed-Bed Reactor” the dielectric between the electrodes consists of pellets. The electrodes can be either used in parallel as plates or arranged as ring electrodes. In “silent discharge” ozone generation, a high

voltage of low frequency is used in a narrow annular space between two metalized glass tubes that are placed into another. Dried air or oxygen is forced through this space. Oxygen molecules are split into its atoms by the energy of the accelerated electrons. As by-products, nitrogen oxides can be formed, which can disturb the formation of ozone. The application of dry gas minimizes the occurrence of unwanted nitrogen oxides and optimizes the formation of ozone. The produced ozone quantity and the efficiency of the procedure depend just as on the current, the construction of the generator and the surrounding temperature. When using pure oxygen the synthesized gas mixture contains up to 18% ozone, which can be separated from the mixture by liquefaction. A classical ozone generator, which produces ozone by silent discharge at normal air pressure, is the ozonizer by Siemens. The procedure was optimized in order to reduce the energy expenditure. Today, larger plants, producing a daily load of several tons of ozone, possess connected loads of several megawatts [5, 18, 24, 33–37].

## 2.6 Electrolytic ozone production

In the year 1840 Schönbein described the formation of ozone for the first time as a gas with characteristic stabbing odor from the electrolysis of sulphuric acid. Apart from the electrolytic production of ozone from acidic electrolytic solutions ozone can also be displayed by catalytic cleavage of water. Through the application of almost neutral electrolytes a higher overvoltage for oxygen can be achieved compared to acidic electrolytic solutions, which results in a minimum oxygen formation. At the anode ozone is produced catalytically from water (in situ) above an electrode potential of 1.7 V with simultaneous oxygen formation. The ozone quantity thereby depends on the charge density in the electrolysis cell, but also on the temperature (at high temperatures fast decomposition of ozone takes place) and the electrolyte. Due to the low temperatures during electrolytic ozone production higher ozone concentrations can be achieved compared to classical ozonizers. In contrast to classical electrophoresis electrodes with a high overvoltage are employed for reduction of formation of oxygen and chlorine. Additionally, the electrodes should consist of electro-chemically stable material and possess a high electrical conductivity. Therefore, only precious metals or metallic oxides in their highest oxidation state are applicable for electrochemical ozone production. In the past, platinum or lead dioxide electrodes were usually used. Through electrolysis of water using platinum or lead dioxide electrodes up to 10 % (w/w) ozone can be generated [5, 25, 31, 38–40].

Lately the application of non-catalytic diamond electrodes with the highest known overvoltage for oxygen formation in water was proving itself. The electrodes are endowed in order to increase their conductivity. In most cases, boron as a p-semiconductor is applied for the endowment. Phosphorus as n-semiconductor can be used as well. The electrodes are produced in the CVD procedure, whereby a thin, boron-endowed diamond layer is applied onto a suitable carrier (e.g. niobium). Several of these electrodes can be bundled together as electrode stack with a large surface. The application of the diamond electrodes results in a so-called Electrochemical Advanced Oxidation Process (EAOP®). Depending on the current density the anodic formation of hydroxyl ions, hydrogen peroxide or ozone can be driven when electrons are absorbed. In addition,

hydrogen is produced. Since the generated hydrogen is able to attack the electrode material, its elimination with the flow at flow rates over 1.5 m/s is necessary. In case of low conductivity of the water very high electrolysis voltages are needed. The dissolved ozone vesicles are sputtered through a cone to even smaller gas vesicles, in order to increase the solubility. Due to the bad water solubility of ozone it is however difficult to achieve higher concentrations than few mg/L at conditions of normal temperature and pressure. The desired quantities of ozone and hydroxyl ions for the oxidation of unwanted materials can be varied through variation of the current. Reversing the polarity protects the electrode, since first of all calcium can deposit in the electrode stack, which leads to an embrittlement of the carrier material at the electrode surface. However, a complete depletion of the hardness components of the electrode cannot be achieved by means of the pole reversal protection. A gradual voltage increase and thus a rising resistance therefore is the consequence in the course of the operation of the electrode [5, 10, 39, 41–44].

Electrolysis cells with a solid electrolyte diaphragm for separation of cathode and anode regions get along without liquid electrolytic solutions. At the same time the electrolyte consists of a polymer, which is able to transport electrical loads (Proton Exchange Diaphragm PEM) and ensures that there is no contact between the electrolysis products. The electrodes possess certain porosity, in order to ensure sufficient water supply and a simultaneous removal of the electrolysis products. In addition the material should be of a high chemical stability and be in close contact with the electrolyte diaphragm. During the electrolysis oxygen and ozone are generated at the anodic side and hydrogen on the cathode side. With this technology up to 750 µg ozone/L can be produced [5, 26].

## 3 Possible applications and outlook

Ozone is in use for cleaning and disinfection purposes already since the middle of the 19th Century. Today an ozonation belongs to the central processing stages in many drinking water works, however usually in combination with other procedures, for example filtration or UV treatment. During the water processing of drinking and process water, ozone is used amongst other processing for an ecologically friendly oxidation of iron, manganese and organic substances and thus for an improvement of biodegradability and sterilization. Therefore, in drinking water processing ozone is suitable, compared with other oxidation procedures, at the same time for removal of persistent ingredients, for example pharmaceuticals, and for water disinfection [7, 15, 16, 26, 32].

The application of cold or non-thermal plasma was originally developed for the production of ozone and was later in use for removal of impurities from gases. Recently, an application for purification of air or removal of bio waste is under development. For applications in the medical sector this method has large potential for sterilization of air and air filters, for surface sterilization of medical instruments and also for skin disinfection. It can also be used for surface disinfection in the food sector. Further uses of ozone for disinfection of production lines in the food sector are currently under development. Figure 2 shows a summary of current and possible future applications of ozone in different technical areas [36, 37, 45, 46].

A detailed description of the development of the application of ozone as a disinfectant in breweries will be given in the next part following this article.

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## Appendix

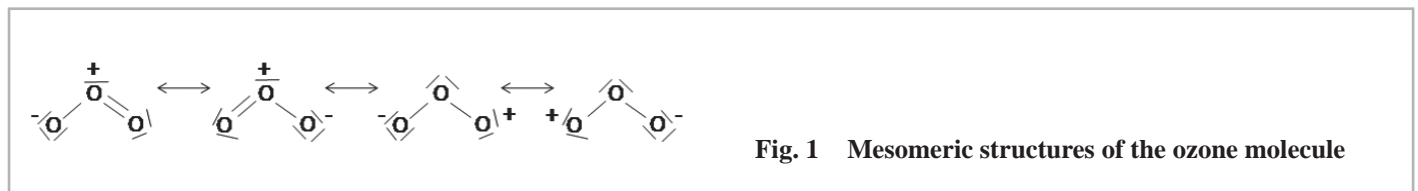


Fig. 1 Mesomeric structures of the ozone molecule

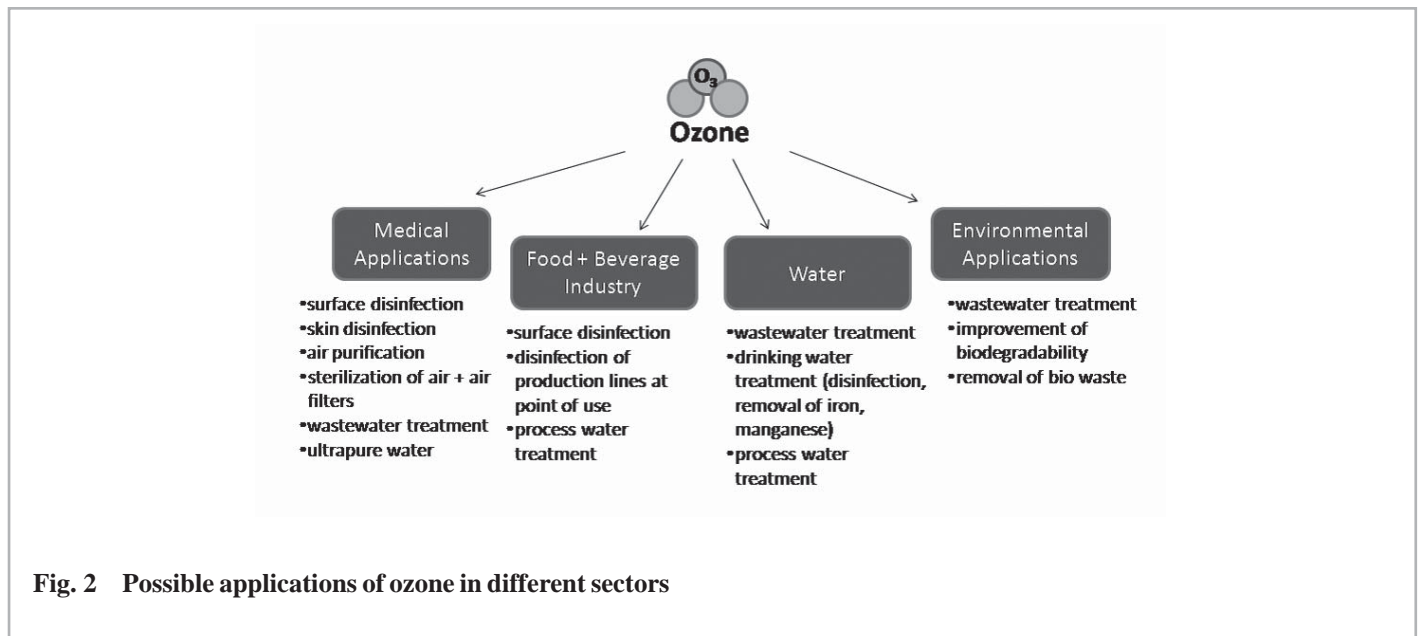


Fig. 2 Possible applications of ozone in different sectors