

The applications of ozone-based advanced oxidation processes for wastewater treatment: A review

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Abstract. The rise in population and industrialization accounts for the generation of a huge amount of wastewaters. The treatment of this wastewater is obligatory to safeguard the environment and various life forms. Conventional methods for high strength wastewater treatment coming out to be ineffective. Advanced oxidation processes (AOPs) for such wastewater treatment proved to be very effective particularly for the removal of various refractory compounds present in the wastewater. Ozone based AOPs with its high oxidizing power and excellent disinfectant properties is considered to be an attractive choice for the elimination of a large spectrum of refractory compounds. Furthermore, it enhances the biodegradability of wastewaters after treatment which favors subsequent biological treatments. In this review, a detailed overview of the AOPs (like the Fenton process, photocatalysis, Electrochemical oxidation, wet air oxidation, and Supercritical water oxidation process) has been discussed explicitly focusing on ozone-based AOPs (like O₃, O₃/H₂O₂, O₃/UV, Ozone/Activated carbon process, Ozone/Ultrasound process, O₃/UV/H₂O₂ process). This review also comprises the involved mechanisms and applications of various ozone-based AOPs for effective municipal/industrial wastewaters and landfill leachate treatment. Process limitations and rough economical analysis were also introduced. The conclusive remarks with future research directions also underlined. It was found that ozonation in combination with other effective AOPs and biological methods enhances treatment efficacies. This review will serve as a reference document for the researchers working in the AOPs field particularly focusing on ozone-based AOPs for wastewater treatment and management systems.

Keywords: advanced oxidation processes; hydroxyl radicals; ozonation; pollutant; recalcitrant compound; wastewater treatment

1. Introduction

Rapid urbanization, industrialization, and an increase in population are considered as the major factors responsible for the depletion and contamination of water sources. The contaminated water or wastewater poses a serious threat to the environment and human health. Wastewater is characterized by the presence of a large number of different compounds. The compounds present in the wastewater mostly vary across different countries, based upon the different water

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consumption patterns (Kumar *et al.* 2018, Pandey *et al.* 2019). Domestic and Industrial sources are considered as the main source that utilizes a large amount of water and generating a huge volume of wastewater. These effluents contain heavy metals, toxic chemicals, grease, dyes, etc. that largely contaminates the ground and surface water sources (Swami and Buddhi 2006). The presence of various organic and inorganic compounds in wastewater affects the environment and human health (Gerba *et al.* 2018, Pandey *et al.* 2019). Surfactants are another group of pollutants produced by both the industrial and domestic sources. The presence of these surfactants is generally attributed to the water by the use of detergents, cosmetics, shampoos, and many other personal care products (Ikehata *et al.* 2004). Therefore, the removal of these contaminants is necessary which requires proper treatment systems to ensure the safety of public health and safeguard the environment (Jiang *et al.* 2006).

Numerous conventional treatment methods such as activated sludge process, trickling filters, reverse osmosis, anaerobic digestion, membrane filtration, adsorption, coagulation & flocculation, etc are commonly adopted to reduce the high concentrations of contaminants present in the wastewater (Huang *et al.* 1993, Hamza *et al.* 2016). These methods are not very effective for the removal of high molecular recalcitrant compounds. These compounds pose a serious threat to the environment and human health so their removal is of utmost necessity. Hence there is a need to focus on the new advanced technologies for the effective removal of these compounds (Oller *et al.* 2011, Gautam *et al.* 2019).

The applications of the advanced oxidation processes (AOPs) are proved to be very effective treatment systems that employ powerful oxidizing agents that effectively oxidized various recalcitrant compounds present in the wastewater (Ikehata and Li 2018). All the AOPs generally work on the same chemical principle i.e., the production of highly reactive hydroxyl radicals. The presence of these radicals plays a major role in the oxidation and degradation of most of the organics present in the wastewater (Pouran *et al.* 2015, Gautam *et al.* 2019).

2. Introduction to advanced oxidation processes

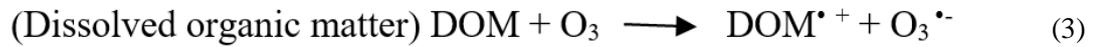
2.1 Fenton process

Fenton process uses the Fenton's reagents for the removal of toxic and other recalcitrant pollutants. The process involves the coupling of hydrogen peroxide (H_2O_2) and ferrous salts (Fe^{2+}) to generate highly reactive hydroxyl radicals ($\cdot\text{OH}$) (Sharma *et al.* 2016). The process requires lower pH values in the range of 2.5 to 3.5 for effective treatments (Pouran *et al.* 2015). The efficiency of the process largely depends upon Fenton's reagents dosage (Michalska *et al.* 2012). High costs of reagents and lower pH adjustment involved in the process limit its applicability on a large scale. The main reactions involved in the Fenton process are described as follows (Deng and Zhao 2015):



2.2 Ozonation process

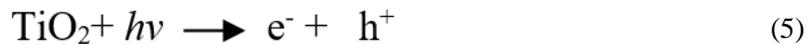
This process involves the application of ozone (O_3) for the removal of various recalcitrant contaminants present in the water and wastewater. The high oxidation ability of ozone (oxidation potential 2.08 V) makes it an excellent disinfectant for the wastewater treatment (Westerhoff *et al.* 2005, Wei *et al.* 2017). The ozonation process involves the two distinct reaction pathways i.e., direct reactions and radical reactions to oxidize the contaminants. In the direct reactions, molecular ozone directly reacts with the contaminants and oxidizes them as shown in Eq. (3) (Andreozzi *et al.* 1999), while a radical reaction utilizes the $\cdot OH$ produced by the decomposition of ozone at higher pH values ($pH > 8$) (Miklos *et al.* 2018). The overall reaction of the ozone decomposition process is shown in Eq. (4) (Gottschalk *et al.* 2009).



Ozone was injected in water as tiny bubbles and results in the effective precipitation of manganese, sulfur, and iron and at the same time, it inactivates the bacteria (Ataei *et al.* 2015). The process was widely used to reduce the chemical oxygen demand (COD) and biochemical oxygen demand (BOD) values of leachate (Gautam *et al.* 2019).

2.3 Photocatalysis

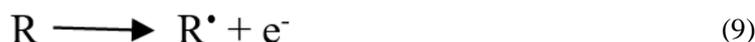
The photocatalysis process is based on the photo-excitation of semiconductor metal oxides to generate the various reactive radical species (Andreozzi *et al.* 1999). Titanium dioxide (TiO_2) with its easy production, high stability, chemical, and biological inert nature is considered to be an ideal photocatalyst (Oturán *et al.* 2014). The process initiates with the photolysis of the TiO_2 surface. The photo-excitation of TiO_2 particles by Ultraviolet (UV) irradiation results in the production of electron and holes in its conduction and valence bands respectively as shown in Eq. (5). Electrons (e^-) with the high reducing power play an important role in the reduction of dissolved oxygen present in the water and results in the formation of superoxide radicals ions ($O_2^{\bullet-}$) as shown in Eq. (6) while the holes (h^+) oxidizes the water molecules and hydroxide ions (HO^-) ions to produce the highly reactive $\cdot OH$ as shown in Eq. (7). The presence of both $\cdot OH$ and $O_2^{\bullet-}$ radicals plays an active role to oxidize the pollutants present in wastewater and convert them into carbon dioxide, water, and inorganic salts (Bauer *et al.* 1999, Saien *et al.* 2011).



The efficiency of the photocatalytic process depends on several factors such as pH, UV light intensity, catalyst loading, dissolved oxygen concentration, turbidity and ionic profile of water, etc (Tijani *et al.* 2014). The process also adopted for the oxidation of various toxic inorganic ions (like nitrite, sulfite, cyanide, and bromate) present in the wastewater (Mills and Le Hunte 1997).

2.4 Electrochemical oxidation

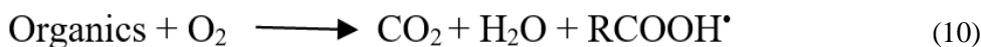
The electrochemical oxidation system works on the electrolysis process of water to oxidize the wide range of contaminants present in the wastewater. The anode involves the oxidation reactions while the cathode is responsible for the reduction reactions to degrade the pollutants present in the water and wastewater. The electrochemical oxidation process follows the two different pathways i.e., direct electrochemical oxidation and indirect electrochemical oxidation to oxidize the organic pollutants (Gautam *et al.* 2019). Direct electrochemical oxidation involves the direct electron charge transfer reactions on the surface of anode to degrade the organic substrate as shown in Eq. (9) (M'Arimi *et al.* 2020). Conversely, indirect reactions involve the in-situ electron generation by the reactive oxidant species like H₂O₂, hypochlorite, ozone, and chlorine, etc (Brillas *et al.* 1996, Vlyssides *et al.* 2003).



The reactions initiate with the oxidation of water at the anode surface that leads to the formation of hydroxyl radicals as shown in Eq. 13. The electrochemical oxidation process is considered to be a very useful, cost-effective, and efficient process (Rajeshwar *et al.* 1994, Gautam *et al.* 2019).

2.5 Wet air oxidation process

The wet air oxidation process is a very promising advance oxidation method, that is used for the treatment of high molecular toxic and recalcitrant compounds present in the wastewater. The process works on the principle of a chemical reaction between the dissolved oxygen and the organic pollutants. The dissolved oxygen in the aqueous media at an elevated temperature (125-320°C) and high pressure (0.5-20 MPa) results in the oxidation and decomposition of organic pollutants and convert them into the inorganic constituents such as water, carbon dioxide or small biodegradable molecular compounds (Dietrich *et al.* 1985). Some of the reactions involved in the wet air oxidation process are shown below (Tungler *et al.* 2015).



The process of wet air oxidation is largely used to oxidize the suspended and dissolved organic pollutants present in the wastewater. This technology is used for the treatment of waste streams that are either considered to be much diluted for the incineration or contains very high molecular recalcitrant compounds (Tungler *et al.* 2015).

2.6 Supercritical water oxidation process

This process is very efficient that mostly used for the treatment of highly toxic and refractory

compounds present in the wastewater (Yang *et al.* 2018). The process is based on the special characteristics of supercritical water. The supercritical water is defined as the state of water whose temperature and pressure lie above its critical value i.e., 374.15°C and 22.1 MPa respectively. Supercritical water is characterized by its low density, low viscosity, low dielectric constant, and higher mass transfer efficiency. These conditions create an excellent homogeneous aqueous environment for the faster rate of reactions between oxygen and organic contaminants (Zhang *et al.* 2017, Li and Wang 2019). The process leads to the complete mineralization of organic compounds and turns them into simple small molecules like carbon dioxide, water, nitrogen, and other inorganic salts (Liu *et al.* 2009, Yang *et al.* 2018). Moreover, this process is considered to be very advantageous in terms of its environmentally friendly nature, high destruction efficiency, and economical aspects (Li *et al.* 2020). However, the main drawbacks of this system may include the salts' precipitation and corrosion-related issues (Yang *et al.* 2018).

3. Process mechanisms

The half-life period of ozone depends on several factors i.e., pH, temperature, concentrations of pollutants, which generally varies from very few seconds to a few minutes depending upon the treatment conditions (Kasprzyk *et al.* 2003). A study conducted by Von Gunten (2003) shows that disinfection through the ozonation process ceases the formation of halogenated by-products. Another study conducted by Rice (1996) has reported that the ozonation process is very efficient for biodegradability improvement which favors subsequent biological treatment systems.

Two different reaction mechanisms were considered in the ozonation process which was direct and indirect reaction mechanism. The process largely depends on the pH of the solution. The low pH values (pH<3) favors the direct molecular ozone reaction mechanism. Conversely, the indirect reaction of ozone gets activated at higher pH (7<pH<10) conditions (Westerhoff *et al.* 2005).

3.1 Direct ozone mechanism

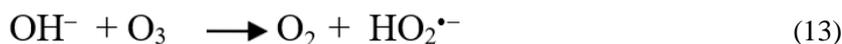
The direct ozone mechanism is characterized by the direct molecular ozone reactions with the organic matter present in the water and wastewater. The oxidation of organic matter in the direct ozonation process is defined by the 1,3 dipolar cyclophilic, nucleophilic, and electrophilic nature of ozone (Riebel *et al.* 1960). However, these reactions are selective for specific organic compounds (Hoigné and Bader 1983). The high reactivity of ozone comes from its highly unstable electronic and molecular configuration (Gardoni *et al.* 2012). In the resonant structure of ozone, one terminal bonded oxygen atom is considered to be electron-rich and one of the other two is considered to be electron deficient. The electron-deficient oxygen atom attains the electrophilic property and reacts with electron-rich species. Electrophilic additions are the primary reactions of ozone that oxidizes a large number of electron-rich organic compounds like various reductive compounds, unsaturated functional groups, sulfides, amines, polyaromatic compounds, and phenols/phenolates, etc. (Riebel *et al.* 1960, Larcher *et al.* 2012). On the other hand, the electron-rich oxygen atom configuration imparts the nucleophilic nature to the ozone. The nucleophilic nature of ozone defines its reactivity with the positive charge species like carbon-nitrogen bond containing substances, electron-deficient positions of carbonyl groups, and double & triple bonds of carbon-nitrogen organic compounds, etc. (Riebel *et al.* 1960). The direct ozone reactions are slow and result in the production of various byproducts like ketones, aldehydes, and carboxylic

acids, etc. (Wu *et al.* 2004).

However, the oxidation of inorganic compounds is based upon the oxygen atom transfer mechanisms. The mechanism involves the process of transfer of an oxygen atom of ozone to the inorganic compounds. The process completes with the formation of unstable intermediates that gets easily oxidized. For example, ozonation of Fe^{2+} (ferrous ions) and cyanide follows the oxygen atom transfer mechanisms and results in the formation of unstable intermediates like ferryl ion (FeO^{2+}) and cyanate respectively, which easily oxidized (Parga *et al.* 2003).

3.2 Indirect ozone mechanism

Indirect ozone reactions involve the generation of hydroxyl radicals which reacts unspecifically with the organic compounds (Hoigné and Bader 1983). The presence of catalysts and higher pH values results in the active decomposition of ozone and leads to the generation of highly reactive hydroxyl radicals. These radicals oxidized effectively to saturated organic compounds also for which the direct ozone reactions are not possible (Chu *et al.* 2008). Decomposition of ozone at higher pH values (Andreozzi *et al.* 1999).



The decomposition of ozone at higher pH values was initiated by the presence of hydroxide ions and follows the reactions as shown in Eqs. (18)-(23) to produce hydroxyl radicals. Von Gunten (2003) has reported that the process of decomposition of ozone as shown in Eqs. (18) and (19) can be accelerated artificially either by increasing the pH or through the addition of H_2O_2 . The catalytic ozonation process is another effective way used for the decomposition of ozone to generate hydroxyl radicals (Zhang *et al.* 2017, Wu *et al.* 2018). The heterogeneous catalytic based mechanisms involve the reactions of ozone with surface hydroxyl groups and variable valence metal ion sites to produce hydroxyl radical species. The state of surface charge on the hydroxyl group sites defines the reaction mechanism rate for the decomposition of ozone (Zhang *et al.* 2015, Zhang *et al.* 2018, Ye *et al.* 2020). However, the presence of some acidic ion in the solution affects the decomposition of ozone. The presence of Cl^- (chloride ions) in acidic solution consumes most of the ozone atoms and produces Cl_2 (chlorine) and ClO_3^- (chlorate ion). These ions inhibit the chain reaction responsible for the generation of reactive hydroxyl radicals (Levanov *et al.* 2012, Levanov *et al.* 2015). Similarly, the presence of carbonate ions affects the important chain reactions and lowers down the generation of hydroxyl radicals (Nemes *et al.* 2000).

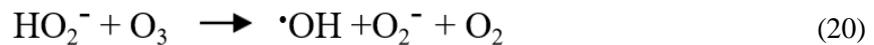
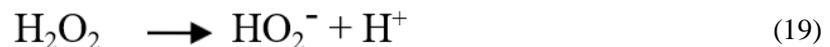
3.3 Ozone as a disinfectant

Ozone is considered to be an excellent disinfectant and widely used for the disinfection purpose of various pathogenic microorganisms (like viruses, fungi, bacteria, and protozoa, etc). Ozone competently inactivates the various resistant pathogens microorganism, for example, a protozoon that gets hardly eliminated by the conventional disinfection process. A study conducted by Von Gunten (2003) shows that in Switzerland almost 90% of ozone applications are being utilized for disinfection treatment purposes. The reaction of ozone with various cellular components like proteins, unsaturated lipids, nucleic acids, and respiratory enzymes, etc of the cell leads to the inactivation and death of the microorganisms. Similarly, when ozone reacts with nucleic acids, it causes the opening of circular plastids of deoxyribonucleic acid (DNA), which eventually inhibits the cell's transcription activities and inactivates the cell (Khadre *et al.* 2001). Many important factors such as ozone concentrations, contact timing, and susceptibility of target organisms, etc define the efficiency of the ozone disinfection systems. The disinfection through the ozonation process ceases the formation of halogenated disinfection by-products (Westerhoff *et al.* 2005). However, the highly unstable nature of ozone with a very low half-life time limits its disinfection efficiency.

4. Ozone based advanced oxidation processes

4.1 Peroxonation process (O_3/H_2O_2)

The principle of this process involves the coupling of ozone and hydrogen peroxide to attain the higher production of oxidizing radicals. The process is considered to be more efficient than the traditional ozonation process. The presence of H_2O_2 accelerates the decomposition rate of the ozone in water and results in the higher production of reactive $\cdot OH$ (Gogate and Pandit 2004). The process initiates with ionization of H_2O_2 into hydroperoxide (HO_2^-) as shown in Eq. (19). The presence of HO_2^- plays an active role in the ozone decomposition process. The HO_2^- reacts with ozone and results in the production of $\cdot OH$ radicals as shown in Eq. (20) (Deng and Zhao 2015).



The process is used for the oxidation of various toxic compounds and micropollutants present in the groundwater, drinking water, and other industrial wastewater. This process is considered to be advantageous in terms of its simple handling and good bactericide action (Oturán *et al.* 2014). However, the high energy consumption and low ozone solubility in water limit the large scale application of this process (Hernandez *et al.* 2002).

4.2 Photo assisted ozone process (O_3/UV)

The ozone/UV process is very efficient for the destruction of various refractory organic compounds (Rosenfeldt *et al.* 2006, Černigoj *et al.* 2007). The application of UV rays in the ozone

mixed aqueous media accelerates the production of reactive $\cdot\text{OH}$ and enhances the performance of the treatment process. The photolysis of ozone under the UV irradiation of wavelength 254nm primarily leads to the production of H_2O_2 (Peyton and Glaze 1988), which on further decomposition generated the highly reactive hydroxyl radicals as shown in Eqs. (21)-(23) (Garoma and Gurol 2004). The direct UV irradiation and indirect hydroxyl radical reaction contribute to the oxidization of a large number of refractory organic compounds present in the wastewater (Garoma and Gurol 2004). The higher molar extinction coefficient of ozone at 254 nm makes the ozone/UV process more effective than UV/ H_2O_2 and $\text{O}_3/\text{H}_2\text{O}_2$ process as it yields a higher concentration of $\cdot\text{OH}$ (Andreozzi *et al.* 1999). The ozone/UV process is considered to be more cost-effective than the individual ozone and UV process for the plant size of capacity higher than 38,000 m^3/day (Venosa *et al.* 1984). Moreover, the process shows the high removal of COD and color in comparison with the ozonation process also which also minimizes the formation of the ozone by-products (Oh *et al.* 2003).



4.3 Ozone/activated carbon process

The process involves the coupling of O_3 and activated carbon to enhance the production of hydroxyl radicals to achieve the higher degradation of the pollutants (Beltrán *et al.* 2006). In general, the homogeneous direct and indirect ozone reactions in the bulk media and heterogeneous ozone reactions on the activated carbon surface with the increased concentrations of hydroxyl radicals marks up the higher efficiency of the process (Valdés *et al.* 2006). Activated carbon surface is considered as the active reaction site for the oxidation of the pollutants by directly adsorbing the pollutants on its surface (De Oliveira *et al.* 2014). The heterogeneous direct and indirect ozone reactions oxidize the adsorbed organic pollutants present on the surface of activated carbon. Activated carbon also enhances the ozone mass transfer rate and makes the process more cost-effective (Chedeville *et al.* 2009). The process is very significant for the removal of that recalcitrant organic compounds, which are hard to eliminate by the ozonation process alone. This process is considered to be very efficient for the removal of hydrophobic micropollutants and pharmaceutical compounds (Sánchez-Polo *et al.* 2008, De Oliveira *et al.* 2014).

4.4 Ozone/Ultrasound process

This process involves the use of ultrasounds in the ozonation treatment process to degrade a large number of non-biodegradable refractory organics present in the wastewater (Weavers and Hoffmann 1998). The process is considered to be more efficient than the individual ozone and ultrasound processes. The cavitation microbubbles formed by the ultrasounds irradiation energy favors the thermolytic decomposition of ozone and in turn, generates a large number of reactive hydroxyl radicals as shown in Eqs. (24) and (25) (Zhang *et al.* 2006). A high concentration of

hydroxyl radicals results in the higher degradation of pollutants. The sonochemical assisted process enhances the volumetric mass transfer coefficient of ozone and hence increases the mass transfer rate of ozone. The higher diffusion of ozone into the aqueous medium leads to the higher efficiency of the process (Destailats *et al.* 2000). Furthermore, the combined process also improves the biodegradability of the wastewater (Guo *et al.* 2016).



))) – Ultrasounds, P – Pollutants

Ultrasound and ozone association is considered to be very effective for the treatment of dyes. The extreme temperature (about 5000°C) and pressure (hundreds of bars) conditions under the ultrasonic irradiation provide a significant supply of heat and energy for the treatment of dyes and results in higher color removal efficiency. These intense conditions minimize the ozone dosage requirements and shorten the reaction time of ozone (Behnajady *et al.* 2008, Guo *et al.* 2011). The process results in the higher discoloration of dyes, higher removal of COD, and increased reduction in biological toxicity (Guo *et al.* 2016). The process was found to be very effective for the degradation of chlorophenol, pesticide nitrobenzene, and pharmaceutical compounds present in the wastewater (He *et al.* 2009).

4.5 O₃/UV/H₂O₂ process

The combination of ozone, hydrogen peroxide, and UV radiation makes the process very powerful and outstanding advanced oxidation technology that results in the quick and complete degradation of contaminants present in the wastewater. This combination is considered to be very effective than the O₃/UV and O₃/H₂O₂ processes (Al-Kdasi *et al.* 2004, Hassanshahi and Karimi-Jashni 2018). The presence of H₂O₂ and UV leads to the active decomposition of ozone and results in the enhanced production of reactive hydroxyl radicals. The hydroxyl radicals completely mineralize the organic contaminants into carbon dioxide, water, and inorganic salt (Hassanshahi and Karimi-Jashni 2018). The process involves the series of reactions, but the main reaction can be described as shown in Eq. 31 (Kurt *et al.* 2017).



The main disadvantage of this process is its high cost, the process is considered to be uneconomical for the large scale applications (Cuerda-Correa *et al.* 2020). In general, the cost of the treatment varies based on several factors like the type of pollutants, concentration of pollutants, water flow rate, and degree of purification needed as studied by Munter (2001). The process is mainly adopted to treat the refractory organic pollutants present in the extremely contaminated wastewater (Cuerda-Correa *et al.* 2020).

5. Limitations and economic analysis of the ozonation processes

Ozonation is considered to be a very effective advanced oxidation technology; however, the

process has some limitations for the large scale treatments. The requirement of higher ozone dosages, lower mass transfer efficiency, short half life cycle of ozone, the formation of ozonation by-products, and higher costs involved in the process limits its applicability for the large scale applications. The uneconomical nature of the ozonation process is mainly associated with its high capital investments and high power cost. The ozonation process is considered to be a very uneconomical choice for the treatment of wastewater with high concentrations of BOD, TOC, COD, and suspended solids. The process requires higher ozone dosages to treat high COD values. The high dosages are often needed to inactivate the various cysts, spores, and viruses that may not remove by the low ozone dosages (US Environmental Protection Agency 1999, Gautam *et al.* 2019). The requirement of higher ozone dosages makes the process very uneconomical. The high input of energy required for the ozone generation process significantly adds up in the increased cost of the process. Generally, the production of ozone requires the high energy consumption of about 12-18 KW/hr (Langlais *et al.* 1991, Lenntech 2016). Theoretically, 1 KWh of electric power is consumed to generate the 1200 gm of ozone (Alonso *et al.* 2005). However, in reality, the working ozone generators consume 1KWh electric power and produce only 130 gm of ozone, which is very lower than the predetermined thermodynamic estimation (Chang *et al.* 1991). Most of this electric power gets dissipated in the form of heat, only a small portion is used for the production of ozone. These losses will be more if the air stream supplied for the ozone generation contains a low amount of oxygen. Higher loss of heat adds up in the waste of energy and indirectly into the increased cost. The cost associated with the production of ozone is estimated as near to 2.32 \$/ kg O₃ corresponding to the standard electricity charge of 0.093 \$ /KWh for industrial usage (He *et al.* 2003). The short half-life of ozone is another disadvantage in the ozonation process that limits the ozone for the on-site generation only. Under the ambient temperature and neutral pH conditions, the half life cycle of the ozone is considered to be only 20 minutes (Langlais *et al.* 1991, Lenntech.2016). The low stability of ozone with the restriction of on-site generation and additional requirements makes the process uneconomical (Amor *et al.* 2019). Furthermore, the low mass transfer rate of ozone is considered to be another important issue for all the ozone-based AOPs. The lower mass transfer of ozone from the gas to the liquid phase limits the dissolution of ozone into the aqueous medium. The low solubility of ozone i.e., 0.57 g/l at the ambient temperature conditions limits the efficiency of the process (Langlais *et al.* 1991, Lenntech 2016). The process in the absence of any other combined treatment scheme remains unsatisfactory to meet the standard discharge treatment limits. Ozonation of various compounds in the wastewater leads to the formation of organic and inorganic ozonation by-products at the end of the process. The organic by-products such as ketones, aldehydes, and carboxylic acids, etc are not very harmful but favor the increased bacterial growth mainly in the distribution system of drinking water (LeChevallier *et al.* 1992, Huang *et al.* 2005). On the other hand inorganic byproducts especially bromate is considered to be very problematic in the drinking water. The bromate is a potential human carcinogen and does not degrade easily as reported by Von Gunten (2003).

6. Application of the ozonation processes in wastewater treatment

The high oxidation and disinfection characteristics of ozone favor its wide applications in water and wastewater treatment. The ozonation process is generally used for the degradation of poorly biodegradable organic compounds. The process is also used to enhance the biological degradability of the wastewater samples (Rajeswari and Kanmani 2009). The ozonation process is

considered to be very effective for the treatment of toxic synthetic organic compounds present in the drinking water. The process is largely used for the removal of color and odors and at the same time, it oxidizes the iron and manganese content present in the drinking water (Zhou and Smith 2002). Ozone is an excellent disinfectant and it is widely used for inactivation of a large group of microorganisms present in the water and wastewater (Hunt and Marinas 1997). Ozone in combination with various other technologies like O₃/UV/, O₃/H₂O₂, and catalytic ozonation process, etc are considered to be more effective than individual processes (Zhou and Smith 2002). Several past studies of ozonation processes for the treatment of a different type of wastewaters are shown in Table 1.

Table 1 Past studies of the ozonation processes

Sample type	Initial characteristics	Method used	Main finding/remarks	References
Aqueous solution (characterized with non-ionic surfactant)	Presence of surfactant Nonylphenol ethoxylates (NPEs) Surfactant concentration :40 mg/l Initial pH: 6.5 -7.6	Ozone induced biodegradability treatment	Ozone dosage : 50.8 mg/l COD removal: 39% TOC removal :16% After biodegradation TOC removal :62.5% COD removal: 70%	(Narkis and Schneiderrotel 1980)
Wastewater	Target influent concentration of MIB (2-methylisoborneol) and geosmin was 100 mg/l. *MIB and geosmin are the two most common odor and taste compounds	O ₃ /H ₂ O ₂ Process (Peroxone Process)	90% removal of MIB and Geosmin was observed at ozone dosage:2 mg/l for O ₃ – H ₂ O ₂	(Ferguson <i>et al.</i> 1990)
Municipal wastewater	Initial COD:120-200mg/l	Ozonation process	Ozone dosage :20 mg/l COD reuction:30-50%	(Toffani and Richard 1995)
Synthetic wastewater	Surfactant concentration of sodium dodecylbenzenesulfonate NaDBS : 15 mg/l Glucose: 200 mg/l Glutamic acid : 100 mg/l Initial pH:10	Ozone-induced biodegradability treatment	At 276 mg/l of ozone applied to 183 mg/l of ozone consumed, biodegradability increased with 30% and 15% of COD & TOC removal respectively.	(Beltrán <i>et al.</i> 2000)
Pharmaceutical wastewater	COD: 830 mg/l DOC:450 mg/l pH:6.9	Ozonation and peroxonation process (O ₃ /H ₂ O ₂) Process (2004)	For the ozonation process, at pH:12, maximum COD reduction:56% For peroxonation process, at H ₂ O ₂ concentration:20 mM, maximum COD removal:83%	(Alaton <i>et al.</i> 2004)
Industrial Wastewater (Textile)	COD :1150 mg/l pH:10 Various toxic chemicals	Ozonation + Ferrous sulfate (coagulation) process	Removal efficiencies: (Color=50%) (COD=59%) (Toxicity=80%)	(Selcuk 2005)

Table 1 Continued

Sample type	Initial characteristics	Method used	Main finding/remarks	References
Industrial wastewater (Pharmaceuticals)	Presence of Antibiotics Steroid hormones X-ray contrast media Beta-blockers	Ozonation process	All contaminants removed with removal efficiency > 90%	(Hernando <i>et al.</i> 2007)
Landfill Leachate	COD:5230 mg/l BOD ₅ : 500 mg/l BOD ₅ /COD : 0.1 Initial pH:8.7	Ozone / Hydrogen peroxide process	COD reduction: 48% Color removal: 94% Increased biodegradability BOD ₅ /COD:0.7	(Tizaoui <i>et al.</i> 2007)
Industrial wastewater (steel manufacturing wastewater)	Presence of pollutant coke BOD range: 510 -1360 mg/l Cyanide : 12-80 mg/l Thiocyanate:275-947 mg/l	Ozonation Process	After 60 minutes of ozonation under neutral conditions, removal efficiencies 45%, 88%, 99%, and 97% for TOC, BOD, thiocyanate, and cyanide are respectively.	(Chang <i>et al.</i> 2008)
Dyes and textile industries aqueous solution	Dye concentration: 250 mg/l COD:150 mg/l TOC :32 mg/l pH : 3.90	Ozonation process	At an ozone dosage: 260 mg/h and ozonation time:30 min, maximum removal efficiencies: Color=97% COD=80% TOC=75%	(Pachhade <i>et al.</i> 2009)
Wastewater with the presence of precursors of halogenated nitrogenous disinfection byproducts	NH ₄ ⁺ -N: 0.34-1.00 mg/l Turbidity:33.7-95.7 NTU pH:7.4-7.8	Ozonation process	Removal efficiencies: Turbidity: 98-99% NH ₄ ⁺ -N:16-93%	(Chu <i>et al.</i> 2012)
Wastewater	Concentration of reactive orange 16: 500 mg/l COD:881 mg/l pH:6.24	Ozonation process	Ozone dosage: 24 mg/l min, after ozone bubbling treatment, COD removal:67%	(Turhan and Ozturkcan 2013)
Landfill Leachate	COD:2000 mg/l NH ₃ -N:960 mg/l Color:3670 Pt. Co.	Ozonation Process	Ozone dosage: 80 g/m ³ COD:250 mg/l reaction time:60 minutes COD removal:26.7% NH ₃ -N removal :7.1% Color removal: 92%	(Amr <i>et al.</i> 2014)

6.1 Application of ozonation processes in paper and pulp industry

Production of 1 tonne of paper requires about 15-60 m³ of freshwater (Thompson *et al.* 2001). Large effluents generated from these industries require effective treatment processes to meet the discharge standards. The ozonation process is considered to be very effective for the treatments of these effluents (Staehelin and Hoigne 1982). Ozone effectively removes the various organic compounds like chlorophenol compounds, extractives, organochlorines, fatty acids, lignin and resins, etc present in the paper mill wastewaters. It also reduces the color and toxicity of these wastewaters (Alvares *et al.* 2001). El-Din and Smith (2002) reported the study of ozonation treatment with results ozone dosage of 230 mg/l, maximum reduction efficiency of color, COD, TOC (Total organic carbon), and AOX (Adsorbable organic halogen) were 86%, 22%, 11%, and 44% respectively. The use of catalysts in the process improves the oxidation performance of ozone. Fontanier *et al.* (2006) studied the catalytic ozonation treatment of biologically treated paper and pulp effluents and reported the results with the higher COD reductions of about 53-72% and better TOC removal as compared with the ozonation process alone. Medeiros *et al.* (2008) have also reported the results of the ozonation treatment of alkaline beach plant effluent at the two different pH values i.e., at 12 and 7 with an ozone dosage of 0.6 mg/ml. It was observed that at these pH values, the biodegradability of effluent was improved by 128% and 210% and the color reductions were 48% and 61% respectively. Hence, the ozonation treatment has significantly refined the quality of effluent. Ozonation processes were proved to be efficient for the treatment of effluents from paper and pulp industries. It effectively oxidizes the phenol, trichlorophenol, syringaldehyde, vanillin, eugenol, guaiacol, chlorophenol, cinnamic acid derivatives, and catechol, etc and various other chemicals present in the pulp mill wastewater (Fontanier *et al.* 2005).

6.2 Pharmaceuticals and personal care industrial wastewater

Pharmaceuticals and personal care products (PPCPs) are considered to be emerging pollutants. The conventional wastewater treatments are not sufficient enough to remove these PPCPs and hence they persist in the treated water and further pollute the environment. Some of the PPCPs get metabolized easily by animals and humans if ingested and some degrade quickly in the environment, but there is a large spectrum of PPCPs which remain biologically active although after the treatment and does not degrade easily. The pharmaceutical wastewater contains a large number of chemicals and conventional treatment methods like coagulation, flocculation, filtration, and disinfection does not readily degrade these chemicals and are very ineffective (Verma and Haritash 2020). Hence there is a need for the advanced treatment processes that can easily eliminate the recalcitrant content present in the pharmaceutical wastewater. The ozonation process is considered to be a very effective AOPs to oxidize and degrade the pharmaceutical waste (Ikehata *et al.* 2006). The electrophilic nature of ozone destroys the double bond, aromatic structure, or amine group structures of the pharmaceutical compounds and eliminates these contaminants (Luo *et al.* 2014). Hernando *et al.* (2007) studied the efficiency of the ozonation process for the removal of pharmaceuticals and concluded that the process is very effective for the elimination of various pharmaceutical contaminants like antibiotics, steroid hormones, x-ray contrast media and beta-blockers present in wastewater. The removal efficiency of all these contaminants came out to be greater than 90%. Paucar *et al.* (2018) studied the ozonation treatment process for the removal of PPCPs present in the secondary effluent of the wastewater treatment plant and it has been observed that at an ozone dosage of 9 mg/l and a period of 15

minutes, 34 out of total 37 PPCPs were degraded below their limit of detection. The ozonation process can be made more effective by combining it with various other processes to achieve the higher degradation of contaminants. De Wilt *et al.* (2018) studied the combined effect of ozonation and biological treatment process for the removal of pharmaceuticals from the secondary clarified effluent and it has been found that the biological ozonation biological processes have reduced over 85% of pharmaceuticals contaminants at the corresponding low ozone dosage value of 0.2 gm O₃/gm of TOC.

6.3 Municipal/domestic wastewater treatment

Municipal wastewater treatment plants employ the ozonation process primarily for the disinfection purpose to meet the standardized microbiological discharge limits (Iorhemen *et al.* 2016). As per the World Health Organization (WHO) guidelines, the maximum value for an effluent for reuse purposes should not be higher than 1000 fecal coliforms/100 ml effluent (Paraskeva and Graham 2002). Ozone oxidizes the cell membrane, nucleic acid, and enzymes of the cell and leads to the death of the cell. The process is considered to be very effective for the inactivation of viruses (Tyrrell *et al.* 1995). The direct reactions and ozone-induced floatation process involved in the ozonation treatment process produces the good quality high dissolve oxygen ozonated effluents of non-toxic nature (Tofani and Richard 1995). Ozonation process also serves as a pre-treatment scheme to enhance the biodegradability of municipal and domestic wastewater for the consecutive bio-associated treatments (Bila *et al.* 2005). A study conducted by Beltrán *et al.* (1999) has reported that pre-ozonation associated biological treatment process resulted in a higher reduction of COD (66%) and BOD (88%) content in comparison with the reduction of only 47% and 80% with no pre-ozonation treatment. The pre-ozonation process also favored the efficient removal of total Kjeldahl nitrogen during the biological treatment process. In general, ozone in combination with the other processes results in the improved treatments of water and wastewater (Ried *et al.* 2009).

6.4 Leachate treatment

Leachate from mature landfill sites contains a high percentage of organic contaminants with high COD & BOD values (Praveen and Sunil 2016). Leachate usually comprises the high concentration of heavy metals, ammonia, suspended particles, halogenated hydrocarbons, and various hazardous chemicals, etc that severely contaminate the ground and surface water systems (Christensen *et al.* 2001, Mahtab and Farooqi 2020). Ozonation process effectively reduces the non-biodegradable organics present in the leachate and hence reduces its strength (Amr *et al.* 2013). Amr *et al.* (2014) reported that under optimum operational conditions of 80 g/m³ of ozone dosage, 250 mg/l COD concentration and 60 minutes of reaction time, reductions of 26.7% COD, 7.1% NH₃-N and 92% color have been observed for the ozone treatment process of the semi aerobic landfill leachate. In the ozonation process, the fragmentation of long organic compounds chains to the lower chains effectively enhances the biodegradability of the leachate. Bila *et al.* (2005) studied the effect of ozonation process on the biodegradability of leachate and it was reported that the ozone treatment significantly improved the biodegradability (BOD₅/COD) ratio of leachate from 0.05 to higher to 0.3 value. Another study conducted by Huang *et al.* (1993) concluded that pre-ozonation is a very sustainable option for the elimination of organic carbon and ammonical nitrogen present in the leachate. Tizaoui *et al.* (2007) studied the combined O₃/H₂O₂

system for the leachate treatment and reported the 94% removal of color and 48% reduction of COD with an enhanced biodegradability ratio from 0.1 to 0.7 values. The results confirmed that combined systems are effective for the treatment of landfill leachate.

7. Conclusions

Wastewater characterized by the recalcitrant compounds requires advanced technologies for their effective treatment. The applications of advanced oxidation processes (AOPs) proved to be very advantageous for such wastewater treatment. The suitability of the methods adopted for treatment is mainly based on wastewater characteristics. For high strength and extremely toxic wastewater, various advanced approaches like the Fenton process, ozonation process, O_3/H_2O_2 , UV irradiation systems, electrochemical oxidation, wet air oxidation, supercritical oxidation process, and their various appropriate combinations generally adopted for their efficient treatment and mineralization to satisfy the standard discharge limits. The generated highly reactive hydroxyl radicals in the AOPs systems efficiently degrade a large number of refractory compounds into simpler compounds, carbon dioxide, and water.

Among AOPs, the Fenton process is very effective but it requires a narrow pH range and high chemical involvement which limits its applicability on the large scale. Photo assisted AOPs are proved to be more efficient than conventional systems that produce more hydroxyl radicals which play an active role to oxidize the pollutants but it requires an additional energy source. The efficiency of the photocatalytic process depends on several factors such as pH, UV light intensity, catalyst loading, dissolved oxygen concentration, turbidity and ionic profile of water, etc. The process was also adopted for the oxidation of various toxic inorganic ions (like nitrite, sulfite, cyanide, and bromate) present in the wastewater. The electrochemical oxidation process is considered to be a very versatile, cost-effective, and attractive choice as compare to other processes. On the other hand, the wet air oxidation process is a very promising approach that is largely used to oxidize the suspended and dissolved organic pollutants present in the wastewater. This technology is used for the treatment of waste streams that are either considered to be much diluted for the incineration or contains very high molecular recalcitrant compounds. The supercritical water oxidation process is very efficient that mostly used for the treatment of highly toxic and refractory compounds present in the wastewater. The process is based on the special characteristics of supercritical water. Supercritical water is characterized by its low density, low viscosity, low dielectric constant, and higher mass transfer efficiency. These conditions create an excellent homogeneous aqueous environment for the faster rate of reactions between oxygen and organic contaminants. The process leads to the complete mineralization of organic compounds and found very advantageous in terms of its environmentally friendly nature, high destruction efficiency, and economical aspects. However, the main drawbacks of this system may include the salts' precipitation and corrosion-related issues.

Overall, the ozone-based AOPs is considered to be a very attractive choice for the treatment of wastewater with high concentrations of BOD, TOC, COD, and suspended solids. Ozone with its high oxidizing potential and the ability of production of highly reactive hydroxyl radicals on decomposition is considered to be a very attractive choice for the water and wastewater treatment process. The excellent disinfection properties of ozone further favor the process for its wide use for potable water treatment. Ozone effectively treats the wastewater generated from various industries, municipal/domestic sources, and landfills, etc. The ozonation process not only

eliminates the refractory compounds but also increases the biodegradability of the wastewaters which further favors subsequent secondary (biological) treatments. Paper and pulp industry treatments employ the ozonation process for active oxidization of the various contaminants like phenol, trichlorophenol, syringaldehyde, vanillin, eugenol, guaiacol, chlorophenol, cinnamic acid derivatives, and catechol, etc. present in its pulp mill wastewaters. Moreover, the ozonation process comes out to be very effective for the treatment of pharmaceuticals. The electrophilic nature of ozone destroys the double bond, aromatic structure, or amine groups of the pharmaceutical compounds and eliminates the contaminants. The ozonation process is effectively used for landfill leachate treatment and effectively enhances the biodegradability of leachate through the fragmentation of long organic compounds chains to the lower chains.

Disinfection through the ozonation process ceases the formation of halogenated by-products. Electrophilic additions are the primary reactions of ozone that oxidizes a large number of electron-rich organic compounds like various unsaturated functional groups, sulfides, etc. Catalytic ozonation involves the use of homogeneous and heterogeneous catalysts for the decomposition of ozone to produce highly reactive hydroxyl radicals for the effective oxidation process. The reaction of ozone with various cellular components like proteins, peptidoglycans, unsaturated lipids, nucleic acids, and respiratory enzymes, etc of the cell leads to the inactivation and death of the microorganisms. The Peroxonation (O_3/H_2O_2) process is considered to be more efficient than the traditional ozonation process. The process is considered to be advantageous in terms of its simple handling and good bactericide action. However, the high energy consumption and low ozone solubility in water limit the large scale application of the process. In the ozonolysis process, the photolysis of ozone under the UV irradiation of wavelength 254 nm primarily leads to the production of H_2O_2 . The higher molar extinction coefficient of ozone at 254 nm makes the O_3/UV process more effective than UV/H_2O_2 and O_3/H_2O_2 process as it yields a higher concentration of hydroxyl radicals. The Ozone/UV process is considered to be more cost-effective than the individual ozone and UV process for the plant size of capacity higher than 38,000 m^3/day . Among the various oxidation technologies, the O_3/UV is widely used for the degradation of various toxic POPs (persistent organic pollutants) like phenolic compounds and pesticides, etc. The presence of activated carbon in the ozonation process catalyzes the process of the decomposition of the ozone to produce hydroxyl radicals. The activated carbon surface is considered as the active reaction site for the oxidation of the pollutants by directly adsorbing the pollutants on its surface. On the other hand, the Sonochemical process enhances the volumetric mass transfer coefficient of ozone and hence increases the mass transfer rate of ozone. This combination is considered to be very effective than the O_3/UV and O_3/H_2O_2 processes. The $O_3/UV/H_2O_2$ process is considered to be very effective than the O_3/UV and O_3/H_2O_2 processes. The main disadvantage of this process involves its high cost, the process is considered to be uneconomical for large scale applications.

Further studies are required for overcoming the restrictions associated with the ozonation process like the requirement of higher ozone dosage, lower mass transfer efficiency, the short half life cycle of ozone, the formation of ozonation by-products, and higher costs involved in the process which limit the ozone applicability for the large scale applications. Future studies should also investigate the possibility of integration of ozone-based AOPs with conventional methods aimed at improving the process cost-effectiveness. The main limitation of using AOPs in the enhancement of biodegradability is the dose and exposure time dependence of the process with an excess of either leading to negative effects. More studies should focus on the optimization of various AOPs' dosage and exposure time for efficient treatment. Furthermore, investigation of ozone-based AOPs by nano-catalysts for wastewater treatment should also focus. The current

research in wastewater treatments with AOPs is mainly focused on a pilot scale in batch mode. The evaluation in real conditions in continuous flow mode is mandatory for large-scale implementation.

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