

# Review of Advanced Oxidation Processes for Waste Water Treatment

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## ABSTRACT

Water is the most important and scarce resource and is the pillar of sustainable development. The growing need for clean water and the decline of freshwater resources pose a challenge. The treatment of wastewater needs to be done for its reuse. Wastewater Treatment is the process of removal and elimination of contaminants from wastewater before it reaches the natural bodies of water and pose a harmful environmental impact. Advanced Oxidation Process (AOP) has been a promising technology for the removal of organic and inorganic contaminants from wastewater. The process mainly involves the generation of a strong oxidant, which can easily degrade the pollutants produced from different sources. In this paper, various methods of Advanced Oxidation Processes are studied.

## 1. INTRODUCTION

Oxidation in wastewater treatment generally involves the use of oxidising agents to remove contaminants from water. Due to its low treatment efficiency and limited use towards the removal of complex pollutants, a new set of techniques are developed called the Advanced Oxidation Processes to degrade organic and inorganic pollutants in water.

Advanced Oxidation Processes (AOPs) represent a group of chemical treatment methods that involve the generation of highly reactive hydroxyl radicals ( $\cdot\text{OH}$ ) in sufficient quantity which removes refractory organic materials, traceable organic contaminants or certain inorganic pollutants and thereby increases the wastewater biodegradability as a pre-treatment ensuring the effective biological treatment.

AOPs are also be used in tertiary water treatment process where it can successfully remove effluent waste. The main goal of cleaning using AOPs is to ensure water is free from any contaminants or and toxicity to such an extent that it can be reintroduced into the natural water bodies. The basic chemical principle of AOP is the attack by the hydroxyl radicals ( $\cdot\text{OH}$ ) which further attack on the pollutant matter making fragments, and reacting on salts until they mineralize and can be filtered in the process.

AOPs find applications in the treatment of various pollutants, including organic compounds, pesticides, pharmaceuticals, and emerging contaminants thereby employed in water and air treatment. These processes are particularly useful for the removal of persistent organic pollutants and other contaminants that are challenging to treat with conventional

methods. Some applications of AOP include water and wastewater treatment, Groundwater remediation, Industrial Effluent Treatment to treat effluents containing complex organic compounds, dyes, and other contaminants.

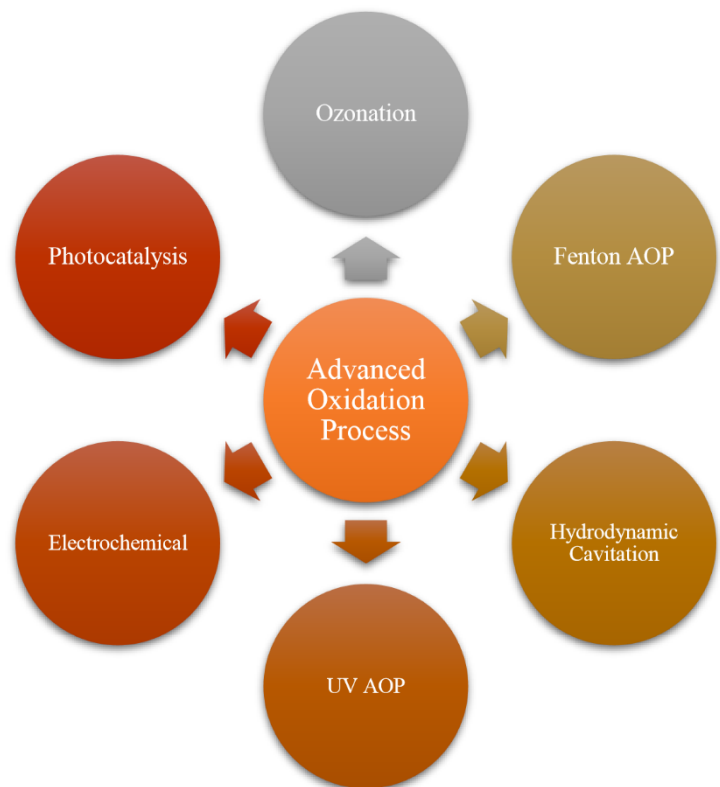


Fig 1.1. Types of Advanced Oxidation Processes

## 2. TYPES OF ADVANCED OXIDATION PROCESS

### 2.1 Hydrodynamic Cavitation

Cavitation refers to the phenomena of formation, growth and subsequent collapse of microbubbles or cavities occurring in a very small interval of time. This time frame is usually in the order of milliseconds but they release large amounts of energy at multiple sites in the reactor. This results in the formation of highly reactive free radicals, increase in surface area for solid catalysts and magnification in mass and heat transfer rates and etc. Cavitation are of two types:- Acoustic and Hydrodynamic where hydrodynamic cavitation is mostly equipped in treatment of wastewater technologies. [1]

In hydrodynamic cavitation, pressure fluctuations are caused due to constriction or due to bottleneck inserted in the flow. This is generally a valve or orifice where there is sudden pressure drop which subsequently creates cavities. The oxidation is described by two mechanisms: free radical attack and pyrolysis. When cavities collapse very violently, they form hydroxyl radicals. They have multiple sites of attacking the pollutant matter which are either at the cavitating bubble, bulk liquid or at the interface. On the contrary, there are very high temperatures formed in the cavitating bubbles (nearly over a 1000°C) which causes the chemical bonds to dissociate. It can be employed to remove organic compounds, dissolved gases, pathogenic microorganisms, heavy metals, particulate matter, and nutrients through the formation and collapse of vapor-filled cavities, facilitating oxidation, disinfection, and separation processes.[1]

The major advantages of cavitation over other AOPs is that it requires minimal operational costs and doesn't need any UV light; and second, the by-products are limited to those expected from the oxidation of the contaminants, avoiding the presence of other dangerous oxidants such as chlorine.[2] Although a major drawback of the process is the substantial energy consumption required for the generation and maintenance of cavitation, making the process economically less feasible. The costs of equipment installation and maintenance is very high due to wear and tear on materials due to the intense forces involved.[3]

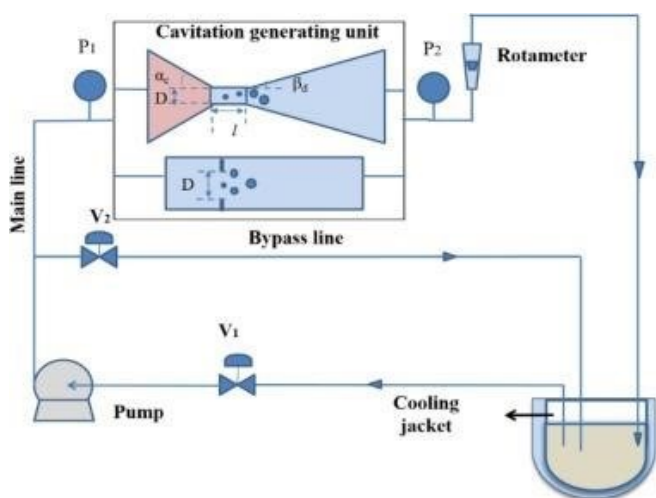


Fig 2.1. Schematic Representation of Hydrodynamic Cavitation [4]

Above figure represents a schematic diagram of the orifice type Hydrodynamic Cavitation reactor. The device consists of a pump, a holding tank, two pressure gauges, a rotameter, control valves, and the HC device. The tank is equipped with a cooling jacket to control the temperature of the circulating liquid. The inlet pressure of the fluid is controlled by adjusting the flow rate. Pressure gauges measure the pressure at the inlet and outlet of the reactor. The rotameter can measure the fluid flow through the mainline to analyze the hydraulic parameters of the reactors.[4]

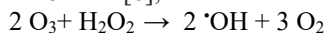
For cavitation reactors to work at optimum efficiency, its convenient to use low operating temperatures and high inlet pressure into the system. Additionally, process is promoted if it contains any catalyst such as  $\text{TiO}_2$ ,  $\text{NiSO}_4$ ,  $\text{CCl}_4$ ,  $\text{Fe}^{2+}/\text{Fe}^{3+}$ ,  $\text{CuSO}_4$  and  $\text{NaCl}$ . Lastly, the presence of certain gases such as  $\text{O}_2$ ,  $\text{O}_3$  and air tend to increase cavitation effects. [1]

### 2.2 UV Advanced Oxidation Process

UV Advanced Oxidation Process refers to a treatment technique where UV light is combined with another oxidant to break down the contaminants present in the bulk of the water. There are two main oxidants available that can be used with UV light that can treat wastewater very effectively. They are  $\text{H}_2\text{O}_2$  (Hydrogen Peroxide) and  $\text{O}_3$  (Ozone). The best advantage of this method is the rate of generation of free radicals is significantly promoted.

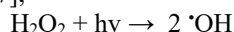
The main mechanism of UV based AOPs is that the oxidants are used to produce very highly reactive hydroxyl radicals ( $\cdot\text{OH}$ ) and UV light is used as an energy source to produce these hydroxyl radicals in a very short interval of time. The ( $\cdot\text{OH}$ ) radicals then react with the contaminants and break them down into smaller inorganic molecules. This process can help lower the concentration of contaminants from several-hundreds ppm to less than 5 ppb and therefore significantly bring COD (Chemical Oxygen Demand) and TOC (Total Organic Carbon) down. [5]

Ozone undergoes the following reaction in presence of UV radiation at about 254 nm [6],



In order to generate optimum working conditions when using ozone as oxidant, it's important to have pH of the sample in the range of 7-8 and the use of higher ozone partial pressures. Additionally, its required that the initial concentration of the pollutant should be lower. [1]

While the following mechanism is followed when hydrogen peroxide is exposed to UV radiation at wavelength ranging from 250-400 nm [7],



It's important to use optimum amount of peroxide, excess of which can be dangerous due to scavenging action. The use of low pH is preferred and at a low temperature due to the kinetic nature of the reaction. Additionally, there is a crucial need of dilution as the kinetic rate constant of the reaction has shown inverse proportion to initial concentration of pollutant hence dilution factor of nearly 10-15 is taken into consideration. [1] UV based AOPs offer major advantages over conventional wastewater treatment technologies such as demonstrating remarkable efficiency in removing a diverse array of contaminants from water. The broad-spectrum activity of UV-based AOP is another significant advantage. Secondly, there are no chemical residues in the treated water. This is a significant advantage over some traditional chemical treatments, as it ensures that the water remains safe for consumption.

Despite its advantages, UV-based AOP with  $H_2O_2$  and ozone also presents certain challenges. One notable drawback is the relatively high energy consumption due to UV light requirement. Next, the cost of reagents, particularly hydrogen peroxide and ozone, can be a significant factor influencing the economic viability of UV-based AOP. Another limitation is the limited penetration depth of UV light in water. UV-based AOP system design with  $H_2O_2$  or ozone is complex. Achieving optimal performance requires careful consideration of factors such as reactor configuration, residence time, and dosage levels.

### 2.3 Fenton Advanced Oxidation Process

Fenton and Fenton-like processes are known as effective and inexpensive Advanced Oxidation Processes for purification of water and wastewater containing organic pollutants. The Fenton process is a reaction between peroxides and iron ions which generates active oxygen species that oxidize organic or inorganic compounds in wastewater.[8] Hydrogen peroxide and Ferrous ions combined are popularly known as Fenton's Reagent which is capable of treating a whole wide range of Wastewater including phenols, Formaldehyde, BTEX, and complex waste of pesticides, specialty chemicals, dyes, etc.

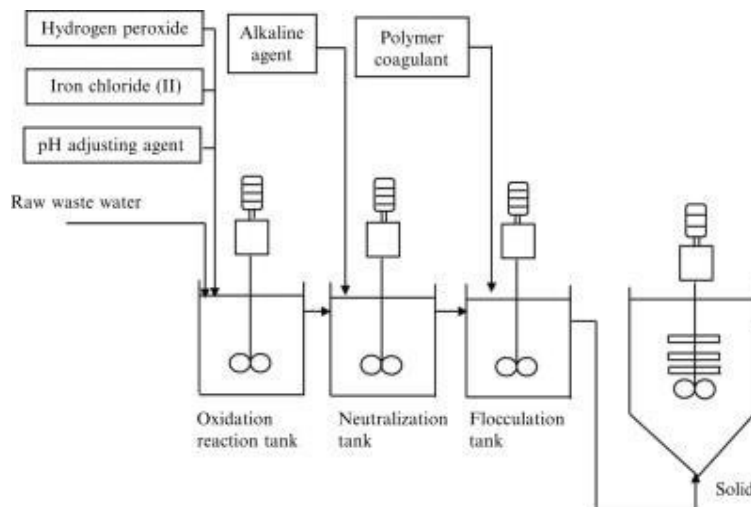


Fig 2.2 Schematic Representation of Fenton Process [9]

When Hydrogen Peroxide reacts with ferrous ions in excess, it gets oxidized to ferric ions in just a few seconds and in the process decomposes hydrogen peroxide to hydroxyl radicals. The reaction rate of the Fenton process is very high. [10]



The ferric ions can be reduced back to ferrous ions by the presence of excess peroxide and radicals. This reverse reaction is termed as Fenton like process and is slower as compared to fenton process. [10]

The ferrous ions react with hydroxide ions to produce Ferric Hydroxo complexes. The coagulation capability of the Fentons reagent depends on this conversion. The dissolved solids in effluent are precipitated. Small flocs are observed during the process which take a lot of time to settle down. Hence during the treatment, it is required to provide chemical coagulation to reduce the settling time of the flocs. Depending on the characteristics of the compound, the following reactions are possible:

- Radical addition: A radical organic compound is formed when hydroxyl radical is added to an unsaturated aromatic compound.
- Hydrogen abstraction: Water and radical organic compounds are formed by the removal of the hydrogen atom. A chain reaction is initiated where peroxy radical is produced when reacted with oxygen.
- Electron transfer: Higher valence ions are generated by electron transfer. oxidation of a monovalent negative ion results in the formation of an atom.
- Radical combination: A hydroxyl radical reacts with another hydroxyl radical to form a stable product. [10]

To achieve the optimum operating conditions, it is necessary for the temperature to be around 10-40 °C to ensure good degradation efficiency and pH of 3 to get highest oxidation activity of hydroxyl radicals. Additionally, higher concentrations of both Fe<sup>2+</sup> and H<sub>2</sub>O<sub>2</sub> is favoured because of higher degradation rate. Higher H<sub>2</sub>O<sub>2</sub> is harmful for microbes and improves the degradation efficiency. Lastly, after the Fenton oxidation, it's mandatory to have chemical coagulation tank which will keep the soluble iron in specified limits. Overall, the reaction takes place for few hours depending on pollutant matter, and the progress of the reaction is noted by a color change. [1]

Fenton's process uses inexpensive reagents and the reactor arrangement is also simple and is the biggest advantage in terms of working and construction. Compared to other systems the reaction is not negatively affected by mass transfer and it has promising application for polypropylene copolymer (PPCP) removal from wastewater. It is advantageous in terms of faster reaction rate, less usage of chemicals, controllable and easy generation of hydroxyl radicals. The reaction can be carried out using both homogeneous and heterogeneous catalyst. But the core chemical of the process: Hydrogen Peroxide is very expensive and may hinder the operations. The major challenge in improving the Fenton technology has been rising due to increasing water regulations. Lastly, for the radicals to be stable it should meet the required acidic conditions. [11], [12]

#### 2.4 Electrochemical Advanced Oxidation Process

Electrochemical Advanced Oxidation Processes (EAOPs) are developed as an efficient and effective wastewater treatment technique over the past three decades. These processes involve the generation of different reactive species that can degrade any class of organic pollutants by oxidation. They are environmentally compatible because they utilize a clean reagent, the electron. [13]

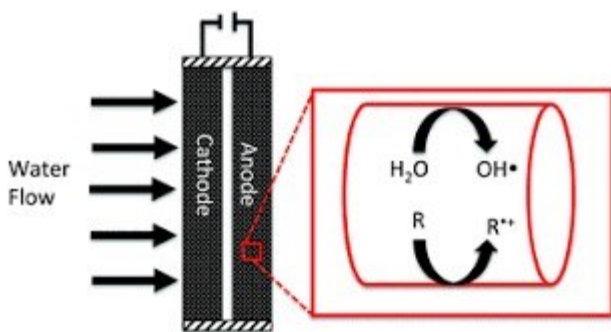
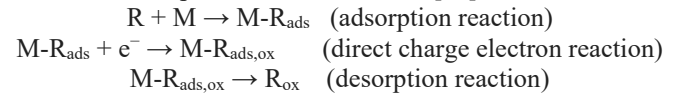


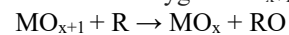
Figure 2.3 Working of Electrochemical AOP [14]

The electrochemical oxidation process may occur in two ways: Direct and Indirect [15]

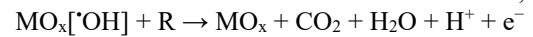
In Direct Electrochemical oxidation, organic pollutants are oxidised by transferring electrons after absorption on anode surface without implication any chemical substances whereas in indirect strong oxidants are formed on the anode surface and propagated in bulk solution resulting in degradation. [15] Direct mechanism involves the contribution of electrons, which oxidize the contaminant molecules, resulting in direct charge-transfer reactions. Adsorption of pollutants occurs on anode surface. Process is controlled by electron transfer and molecular transport at anode interface. [15]



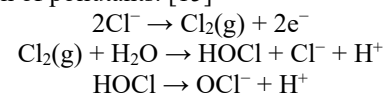
R is pollutant molecule and M is anode surface. It uses low applied anode potential to avoid oxygen evolution reaction. Active anodes with low oxygen evolution potential (OEP) generate chemisorbed active oxygen MO<sub>x+1</sub>,



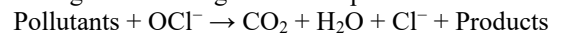
Whereas non-active anodes with high OEP avoids the reaction and favours mineralization of carbon dioxide,



Indirect mechanism uses intermediate formation of strong oxidant reagents, electrochemically generated on the anode surface which act like electron carriers between anode and organic contaminant. It is more efficient in preventing electrode passivation and enhancing electro-catalytic activity. The potential at which the intermediates are generated must be far from the OEP and rate of intermediate generation must be higher than rate of side reactions. Oxidising species are hydrogen peroxide, ozone, percarbonate and active chlorine of which active chlorine is anodically oxidising species used for oxidation of pollutants. [15]



The generated OCl<sup>-</sup> acts as the main oxidizing chlorine-mediator agent in the degradation of pollutants .



These processes enhance the general performances of the treatment with use of active chlorine. It can also be used for degrading organic pollutants in wastewater treatment and can overcome several problems widely encountered in direct oxidation, such as anode surface fouling, mass transfer limitations, and higher energetic cost. [15]

To improve the performance of electrochemical oxidation AOP, it is very important to choose the right material for anode as it affects amount of hydroxyl ions formed. Degradation efficiency changes with applied current density and also the concentration of supporting electrolyte as it influences conductivity of the entire system. [15]

Electrochemical based AOP is very efficient and a simple and streamlined approach to post-reverse osmosis (RO) disinfection. H<sub>2</sub>O<sub>2</sub> is fully utilised and the process is very energy efficient. It is highly versatile and amendable and safe. There is no sludge formation and doesn't have any pH restrictions. It is a green process in its existence. But this process is highly expensive and requires high oxygen overpotential anodes otherwise it can cause electrode fouling. The formation of halogenated byproducts is still a concern as it can tamper the process efficiency. [16]

## 2.5 Ozonation

Ozonation is an oxidation treatment process that is mainly used to enhance sludge hydrolysis and to improve the biodegradability of solids. Ozonation in terms of wastewater treatment is an advanced, mature and efficient alternative to improve the traditional methods. [17] During the pretreatment step of ozonation, the sludge flocs are broken down into fine and dispersed particles. It is considered to be one of the effective techniques to disintegrate sludge yet the production of ozone requires a high energy input. Also it is a powerful oxidizing method for industrial wastewater treatment. It can destroy all microorganisms and many organic compounds. Ozonation can also reduce colour by 90–98% and chemical oxygen demand (COD) by about 45%. [18]

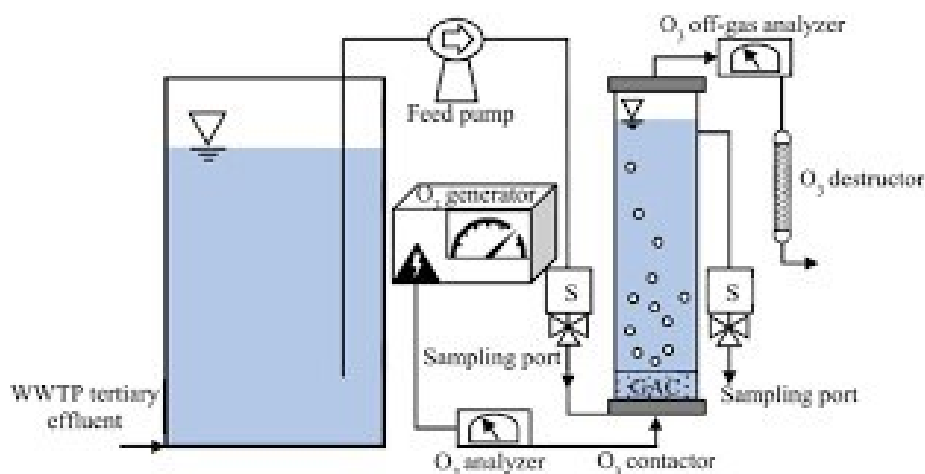


Fig 2.4 Schematic Representation of Ozonation Process [19]

- Process begins by introduction of ozone in the waste water by bubbling ozone gas or injecting liquid ozone.
- The next step involves Direct Oxidation in which the ozone bubbled into the wastewater reacts directly with the organic and inorganic pollutants present in the wastewater. Ozone having high oxidative potential leads to the breakdown of complex molecules to simpler molecules.
- Once ozone is reacted to wastewater, it decomposes in water to form hydroxyl ions ( $\cdot\text{OH}$ ).
- Ozone and hydroxyl radicals attack and break down organic pollutants such as oils, phenols, pesticides, pharmaceuticals, and other refractory compounds. The process results in the formation of typically less harmful and biodegradable byproducts.
- The next step involved is Disinfection wherein the Ozone serves as an excellent disinfectant, effectively inactivating bacteria, viruses, and other pathogens present in the wastewater.
- Following Disinfection is Colour and Odour Removal where Ozonation is effective in removing colour and unpleasant odours associated with certain industrial wastewater streams. It breaks down chromophores and other

colour-causing compounds, leading to a visually clearer and aesthetically improved effluent.

Ozonation, as a method for wastewater treatment, presents several advantages and disadvantages. On a positive side, ozonation is highly effective in disinfecting wastewater by eliminating bacteria, viruses, and pathogens. Its versatility allows it to target a broad range of pollutants, offering a comprehensive solution for various wastewater streams. It has also excelled in removing colour and odour, enhancing the aesthetic quality of treated water. Moreover, the process leaves minimal chemical residues, contributing to environmentally friendly effluents.

However, the method comes with challenges, including high initial and operational costs due to expensive equipment and energy-intensive ozone generation. Ozonation lacks a long-lasting residual effect, necessitating additional measures for sustained microbial safety. The selective reactivity of ozone may limit its efficacy against certain pollutants, and the complex system design requires specialized expertise. Despite these drawbacks, the advanced oxidation power of ozone makes it a valuable tool in modern wastewater treatment strategies.

## 2.6 Photocatalysis

Photocatalytic oxidation is an advanced oxidation process (AOP) that involves the use of photocatalysts to initiate oxidation reactions in the presence of light. This method harnesses the power of light-activated catalysts to generate highly reactive oxygen species, such as hydroxyl radicals ( $\cdot\text{OH}$ ), which then oxidizes and decompose organic and inorganic pollutants in water and air. The photocatalytic process typically relies on semiconductors, as catalysts, when exposed to ultraviolet (UV) or visible light, create electron-hole pairs. These pairs participate in redox reactions, leading to the production of reactive oxygen species that ultimately cause the progressive fragmentation of contaminants. Photocatalytic oxidation is employed in environmental applications, including water and wastewater treatment, air purification, and the remediation of pollutants in various

industrial and urban settings and the technology is valued for its ability to address persistent and refractory pollutants and its potential for sustainable and energy-efficient treatment processes. [20]

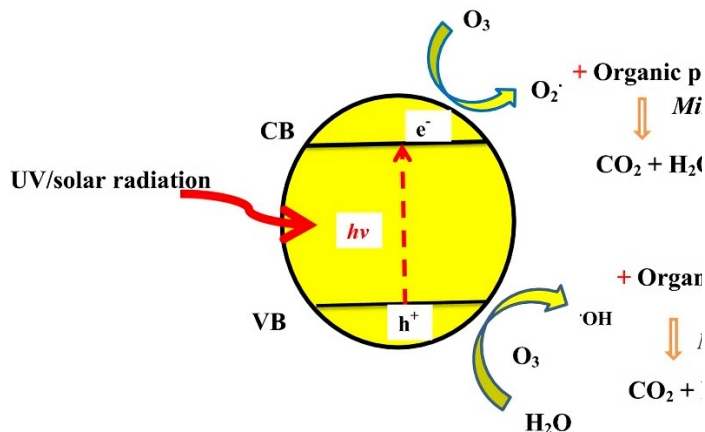
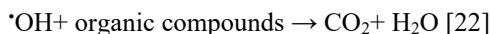
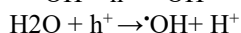
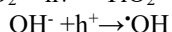
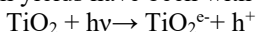


Fig 2.5 Working of Photocatalysis [21]

These photoactivated chemical reactions are characterized by free radical mechanism initiated by the reactions of photons of a proper energy level with the molecules of chemical species present in the solution regardless of the catalyst. In this case, electrons of the semiconductor crystalline structure on excitation form electron-vacancy pairs. The charged carriers interact with different compounds (electron donors and acceptors) adsorbed at the catalyst surface generating free radicals, which react with the pollutants in the adsorbed state. The surface area and the number of active sites offered by the catalyst (thus the nature of the catalyst) for the adsorption of pollutants plays an important role in deciding the overall rate of degradation. Best photocatalytic performances with maximum quantum yields have been with Titania. [1]



1. The first step is the photocatalyst preparation in the form of nanoparticles or a thin film.
2. The next step includes immobilization of photocatalyst followed by preparation of reactor system which includes the wastewater and the photocatalyst.
3. Once the reactor system is set, the photocatalyst is then exposed to a specific light source .
4. Once it is exposed to light, the photocatalyst absorbs photons leading to the excitation of electrons from the valence band to the conduction band.
5. The absorbed photons generate electron-hole pairs. Electrons move to the conduction band, leaving holes in the valence band.
6. Then the Electrons in the conduction band and holes in the valence band initiate redox reactions with water and oxygen in the surrounding environment.
7. Oxygen molecules are reduced to form superoxide radicals ( $\text{O}_2^{\bullet-}$ ) and hydroxyl radicals ( $\bullet\text{OH}$ ) through reactions with the photogenerated electrons and holes.
8. The highly reactive oxygen species, especially hydroxyl radicals, attack and oxidize organic and inorganic pollutants present in the wastewater. [23]

### 3. COMPARISON

Parameter	Hydrodynamic Cavitation	UV- Based AOP	Fenton Reaction	Electrochemical AOP	Ozonation	Photocatalysis
Cost	High	High	Low	High	High	High
Treatment Efficiency	High	High	High	High	High	High
Complexity	Moderate	High	Low	Moderate	High	Moderate
Energy Intensity	Moderate to High	High	Low	Low to Moderate	Moderate to High	Moderate to High
By-product Formation	Variable	None	Yes	None	Yes	Yes
Selectivity towards pollutants	Non-selective	Non-Selective	Non - Selective	Non-Selective	Selective	Non - Selective

### 4. CONCLUSION

Advanced Oxidation Processes have garnered importance in the tertiary wastewater treatment because of its ability to treat complex contaminants. In this report, we studied different methods of AOP and their background techniques in treating organic and inorganic contaminants in waste water. This field needs extensive further research and development in order to minimize cost and energy consumption and maximize treatment efficiency and degradation of pollutants. But it has the potential to become the core of tertiary treatment of wastewater. The feasibility of selecting an AOP depends on the specific characteristics of the wastewater, regulatory requirements, and economic considerations. When determining the most feasible AOP, it is crucial to conduct a comprehensive analysis, considering capital and operational costs, energy consumption, and the treatment efficiency of each method on the basis of the type of contaminants present in the water.

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