

# A Review on Wastewater Treatment Containing Organic Pollutants Using Advance Oxidation Processes

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## ABSTRACT

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Advanced oxidation processes (AOPs) are the technologies that generally use the hydroxyl radicals, the ultimate oxidant for the remediation of organic contaminants in wastewater. These are highly effective novel methods speeding up the oxidation process. AOPs can combine with Fenton ( $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ), photo-Fenton ( $\text{H}_2\text{O}_2/\text{UV}/\text{Fe}^{2+}$ ), and electro-Fenton, heterogeneous photooxidation using titanium dioxide ( $\text{TiO}_2/\text{hv}$ ), singlet molecular oxygen process, singlet oxygen-photo-Fenton process, sonolysis, peroxidation combined with ultraviolet light ( $\text{H}_2\text{O}_2/\text{UV}$ ), UV alone,  $\text{UV}/\text{O}_3$ ,  $\text{UV}/\text{S}_2\text{O}_8^{2-}$ ,  $\text{UV}/\text{Chlorine}$ , peroxone ( $\text{O}_3/\text{H}_2\text{O}_2$ ), peroxone combined with ultraviolet light ( $\text{O}_3/\text{H}_2\text{O}_2/\text{UV}$ ),  $\text{O}_3/\text{UV}$  system,  $\text{O}_3/\text{TiO}_2/\text{H}_2\text{O}_2$  and  $\text{O}_3/\text{TiO}_2/\text{Electron beam irradiation}$ . Ozone ( $\text{O}_3$ ), catalyst or ultraviolet (UV) irradiation to offer a powerful treatment processes of wastewater. Future research should be focused on enhancing the properties of heterogeneous catalysts in AOPs. This review report of different AOPs utilized for the removal of various phenolic compounds and textile dyes in wastewater.

**Keywords :** Advanced Oxidation Processes, Wastewater Treatment, Organic Contaminates, Heterogeneous Catalysts, Hydroxyl Radicals.

## I. INTRODUCTION

World is facing the challenge of purification of water and air resources in this twenty-first century. On the one hand, society is enjoying the comforts and benefits that chemistry has provided. On the other hand, the problem of proper disposal of various products and by-products of dye industries is created.

In spite of many uses, the dyes are toxic and carcinogenic in nature. Environmental contamination by these toxic chemicals is emerging as a serious global problem. Hence, environmental pollution and its related problems have become threatening for human being.[1] It might be due to the speedy expansion of world population and the increasing urbanization and automation or industrialization.[2]

Discharge of water pollutants from industries into fresh water bodies have been projected as a main reason of environmental pollution.[3] Organic dyes are mostly used in textile, paper, pulp, leather and food industries, which are released into water.[4]

The dye-containing coloured water is hard of any use, but if the coloured solutions are bleached to give colourless water, then it may be used for many purposes because bleached dye solution is less toxic and almost harmless. The photocatalytic bleaching seems to be quite promising and can provide a low-cost method to solve this problem. Organic photocatalysts are easy to functionalize, which means the catalyst could be attached to advanced materials and bio-materials.[5, 6]

Water quality may be threatened by the presence of new contaminants. The presence of many organic contaminants in wastewater, surface water, and groundwater may result from contaminated soil, agricultural runoff, industrial wastewater and hazardous compounds. Agro-industrial wastewaters are characterized by the presence of a higher amount of organic and inorganic pollutants of environmental concern.[7]

The presence of these organic compounds in water poses a serious threat to public health since most of them are toxic, endocrine disrupting, mutagenic or potentially carcinogenic to humans, animals and aquatic life in general. Most of the colors in the effluent from dyes involve aromatic compounds, which are chemically stable and harmful to human health, when they enter into aquatic system.[8, 9]

Many organic pollutants are considered as toxic and detrimental, even when present at very low concentrations. Therefore, their removal from the contaminated water is of high priority. Consequently, the need for efficient treatment of these contaminants is imperative. Physical methods such as adsorption[10], biological methods[10, 11] and chemical methods[12] are most frequently used for the treatment of these organic compounds like dyes. In certain cases, conventional treatment methods such

as biological processes are not effective due to the recalcitrant nature of the contaminants present.[13]

The increase in the disposal of recalcitrant organics demands for newer technologies for the complete mineralization of wastewater.[14] Therefore, oxidation processes are preferred to degrade such organics present in wastewater.

Advanced oxidation processes (AOPs) are more useful processes for removal of water contaminants.[15, 16] AOPs are the most attractive and favourable option for the effective removal of organic pollutants in wastewater. This study presents a general review on such processes developed to decolourize or degrade organic pollutants. Almost, all kinds of organic compounds and organic micro-pollutants dissolved in drinking water can be degraded using advanced oxidation processes and these techniques are quite as powerful technologies.[17] AOPs are based on the in situ generations of strong oxidants, i.e. hydroxyl radicals and sulphate radicals, for the oxidation of organic pollutants.[18]

Classical persistent organics such as organochlorine insecticides, solvents and polychlorinated biphenyls, a growing number of emerging pollutants of both; synthetic and natural origins have been removed by Ikehata et al.[19] The aqueous organic pollutants, including pesticides, pharmaceuticals, personal care products, surfactants, microbial toxins and natural fatty acids have been degraded using ozonation and advanced oxidation processes.

AOPs are efficiently and effectively convert these organic compounds into better bio-degradable, less harmful and eco-friendly substances. Several types of AOPs are being developed.  $O_3$  and  $H_2O_2$  act as oxidants with assistance of light catalyst (e.g.  $Fe^{2+}$ ,  $Fe^{3+}$  and  $TiO_2$ ), ultrasonic insertion or thermal input[20] and several other combinations such as Fenton ( $H_2O_2/Fe^{2+}$ ), photo-Fenton ( $H_2O_2/UV/Fe^{2+}$ ), and electro-Fenton, heterogeneous photooxidation using titanium dioxide ( $TiO_2/h\nu$ ), singlet molecular oxygen process, singlet oxygen-photo-Fenton process,

sonolysis, peroxidation combined with ultraviolet iii. light ( $\text{H}_2\text{O}_2/\text{UV}$ ), UV alone,  $\text{UV}/\text{O}_3$ ,  $\text{UV}/\text{S}_2\text{O}_8^{2-}$ ,  $\text{UV}/\text{chlorine}$ , peroxone ( $\text{O}_3/\text{H}_2\text{O}_2$ ), peroxone combined with ultraviolet light ( $\text{O}_3/\text{H}_2\text{O}_2/\text{UV}$ ),  $\text{O}_3/\text{UV}$  system,  $\text{O}_3/\text{TiO}_2/\text{H}_2\text{O}_2$  and  $\text{O}_3/\text{TiO}_2/\text{Electron beam irradiation}$ . [21-24]

Daneshvar and Khataee [25] decolorized solution containing a common textile and leather dye, C. I. acid red-14, at pH 3, using the Fenton,  $\text{UV}/\text{H}_2\text{O}_2/\text{O}_2$ ,  $\text{UV}/\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ,  $\text{UV}/\text{H}_2\text{O}_2/\text{Fe}^{3+}$  and  $\text{UV}/\text{H}_2\text{O}_2/\text{Fe}^{3+}/\text{oxalate}$  processes. Ghime et al. [26] investigated about the efficiency of photochemical advanced oxidation processes (UV photolysis,  $\text{UV}/\text{H}_2\text{O}_2$ ,  $\text{UV}/\text{TiO}_2$ ,  $\text{UV}/\text{H}_2\text{O}_2/\text{TiO}_2$  and  $\text{UV}/\text{H}_2\text{O}_2/\text{Fe}^{2+}$  processes) to decolourize malachite green oxalate dye. The AOPs was proved to be more effective in treating this dye on irradiation.

Different AOPs are based on the formation of reactive and short-lived oxygen-containing intermediates, hydroxyl radicals ( $\cdot\text{OH}$ ) for decolourization of organic dyes. [27] The versatility of AOPs is enhanced by offering the different possible ways for  $\cdot\text{OH}$  production; thus, allowing better compliance with the specific treatment requirements.

AOP involves the generation of hydroxyl radicals in sufficient quantity to affect water purification. [28] The hydroxyl radicals are extraordinarily reactive species. It is a non-selective oxidizing agent, powerful and capable of oxidizing a wide variety of organic molecules [29], which can be used to convert also those compounds that are not sensitive to photolysis. [30] Heterogeneous photocatalysis is the most promising method of AOPs for wastewater treatment with low organic contents. [31, 32]

Advanced oxidation involves mainly three steps schematized in Figures 1 and 2:

- Formation of strong oxidants (e.g. hydroxyl radicals).
- Reaction of these oxidants with organic compounds in the water producing biodegradable intermediates.

Reaction of biodegradable intermediates with oxidants referred to as mineralization (i.e. production of water, carbon dioxide and inorganic salts).

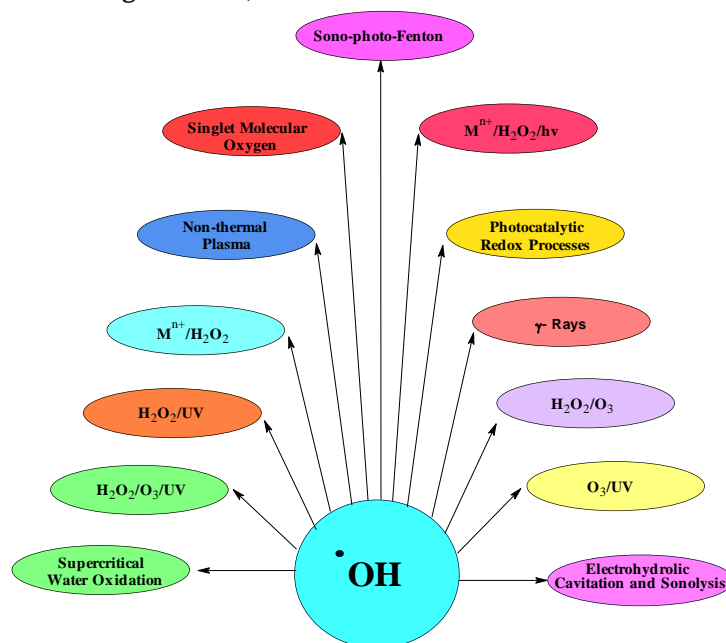


Fig. 1 : Several methods for hydroxyl radical formation

In principle, advanced oxidation techniques can result in the complete mineralization of organic compounds. However, in most of the cases, this is not the goal. By means of oxidation processes, most micropollutants will be converted into smaller molecules, which, in general, are better biodegradable. Ikehata et al. [33] gave an overview of AOP applications for conversion of pesticides and pharmaceuticals. AOPs are more widely utilized in wastewater treatment. [34]

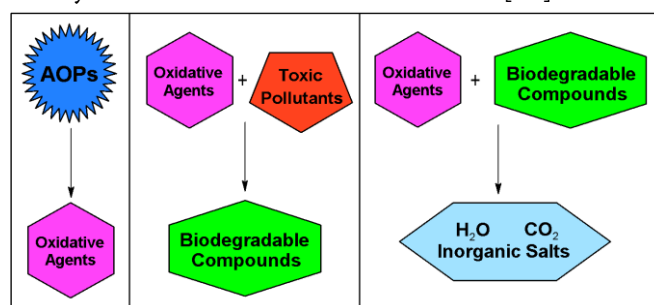


Fig. 2: Reaction of biodegradable intermediates with oxidants

AOPs have been employed for destruction of pollutants from laboratory scale to pilot plant scale, in the field of overall pollutant degradation to specific organic molecule destruction, degradation of micro-

pollutants, agrochemicals, pharmaceuticals, decolourization and mineralization of dyes and sludge treatment.[35-37] Advantages of AOPs include faster rate of reaction, small foot print, high potential to reduce toxicity of organic compounds and nearly complete mineralization of organic pollutants. These technologies does not concentrate the wastewater effluents for further treatment[38] and it does not produce waste also that requires further treatment such as "spent carbon".[39] Moreover, it does not create wastes as with other physical, chemical or biological processes.

Various types of AOPs were identified and employed for the wastewater treatment but the Fenton and photo-Fenton processes have been proved to be the effective, energetically efficient, most powerful, cost effective and least tedious method for the treatment of recalcitrant compounds, when used exclusively or coupled with conventional and biological methods. Fenton and photo-Fenton processes do not require costly reagents or sophisticated equipments. These techniques are ecologically viable due to their relatively simpler approach, use of less hazardous chemicals and cyclic in nature so that less concentration of these chemicals is needed.

## II. DIFFERENT AOPs METHODS

Military applications

### 2.1 Fenton Processes

Fenton's oxidation technique appeared is cost-effective and has ease of operation. Neyens and Baeyens[40] indicated that Fenton's oxidation is a very useful method in the removal of many hazardous organic compounds from wastewater effluents.

#### 2.1.1 Types of Fenton Processes

Different types of Fenton processes include Fenton, photo-Fenton, electro-Fenton, photo-electro-Fenton, sono-Fenton, sono-photo-Fenton, sono-electro-Fenton, homogeneous & heterogeneous Fenton, photo-Fenton, hybrid Fenton and Fenton type processes.[41]

#### 2.1.2 Fenton Reaction

The Fenton reaction was reported by H. J. Fenton for the first time in 1894 and is described as the enhanced oxidative potential of  $\text{H}_2\text{O}_2$ , when iron (Fe) is used as a catalyst under acidic conditions[42] that are efficiently utilized in the wastewater treatment process for the removal of many hazardous organics from wastewater.[43] In dark, the reaction is retarded after complete conversion of  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$ . The reasons for the rate enhancement of degradation on light irradiation include the photoreduction of  $\text{Fe}^{3+}$  ions back to  $\text{Fe}^{2+}$  ions, which produce new  $\cdot\text{OH}$  radicals with  $\text{H}_2\text{O}$ , according to the following mechanism[44]

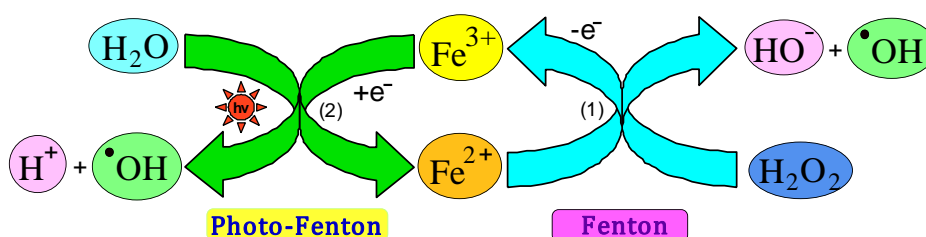
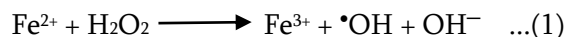
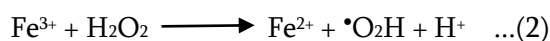


Fig. 3: Mechanism of Fenton and photo-Fenton process

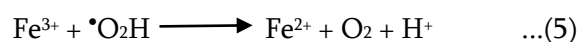
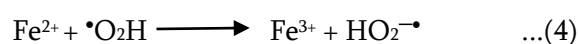
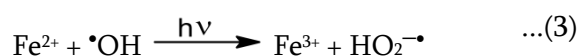
The reactions involved in Fenton processes are recognized as Fenton reaction and implies the oxidation of ferrous to ferric ions to decompose  $\text{H}_2\text{O}_2$  into hydroxyl radicals. It is usually considered as the core of the Fenton chemistry.[45]



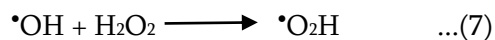
In Fenton reaction,  $\text{Fe}^{3+}$  ion is generated and it can be reduced to  $\text{Fe}^{2+}$  ion and form some other radicals by reaction with  $\text{H}_2\text{O}_2$  as in eq.(2). This is an effective cyclic mechanism. In Fenton reaction, apart from ferrous ion regeneration, hydroperoxyl radicals ( $\bullet\text{O}_2\text{H}$ ) are also produced. The  $\bullet\text{O}_2\text{H}$  radicals may also attack organic compounds but they are less sensitive than hydroxyl radicals. It should be noted that  $\text{H}_2\text{O}_2$  is continuously consumed to produce  $\bullet\text{OH}$  radicals while  $\text{Fe}^{2+}$  ion acts as a photocatalyst.



The following reactions are involved in Fenton chemistry;



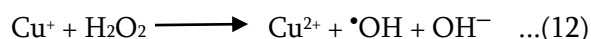
In these reactions, hydroxyl radicals and hydroperoxyl radicals are consumed and ferrous iron is regenerated from ferric ion through these reactions. Eqs. (6–9) also reported to occur during the Fenton process and they are radical–radical reactions or hydrogen peroxide–radical reaction:



In the absence or presence of an organic molecule to be oxidized, the decomposition of hydrogen peroxide to molecular oxygen and water occurs according to eq. (10). This reaction leads to the exploitation of bulk oxidant into degraded product, eq. (11).



Fenton-like reactions are those reactions in which other metals at low oxidation state such as copper are used.[46] For example:



The oxidative decolourization of reactive black-5 in aqueous solution has been studied by Marco et al.[47] using Fenton ( $\text{H}_2\text{O}_2/\text{Fe}^{2+}$ ) and photo-Fenton ( $\text{H}_2\text{O}_2/\text{Fe}^{2+}/\text{UV}$ ) processes. This investigation reveals that both these methods can remove the color of reactive black-5.

Tang et al.[48] examined the influence of dyes on the Fenton reaction of organic compounds under visible irradiation. It was found that the presence of dyes could accelerate greatly the Fenton reaction of organic compounds such as salicylic acid, trichloroacetic acid and a complete mineralization of those compounds could also be achieved. The dyes such as alizarin violet-3B having anthraquinone structure showed much more significant effect on the reaction under visible irradiation. Chaudhuri et al.[49] also oxidized methylene blue using a Fenton-like reaction.

The Fenton reaction is easy. Therefore, it has been widely used to degrade or completely oxidize the pollutants. One of the major disadvantages of Fenton reagent is the deposition of large amounts of ferric ion sludge. There is a need for continuous addition of ferrous ion into the reaction medium for the reaction to proceed further. This disadvantage could be overcome by the use of photo-Fenton reagent, which is a cyclic process and regenerates  $\text{Fe}^{2+}$  ion. Fenton and majorly, photo-Fenton are the promising treatment options for wastewater.[50]

## 2.2 Photo-Fenton Process

The photo-Fenton reaction is one of the effective methods for preventing water pollution. Photo-Fenton process is more effective and faster than Fenton's reagent in the oxidation of organic compounds and that the organic compounds are completely oxidized and degraded into carbon dioxide and water. The naphthol green-B dye from wastewater has been completely oxidized into  $\text{CO}_2$  and  $\text{H}_2\text{O}$  using the photo-Fenton process by Kumar et al.[51] Mogra et al.[52] reported the photochemical degradation of 1,4-dichlorobenzene and chlorobenzene by the photo-Fenton reagent.

Using photo-Fenton process, wastewater containing organic dyes (metanil yellow, fast green-FCF, and rhodamine-B) were degraded by Kumar et al.[53-55] Sheikh et al.[56] reported photooxidation of some organic pollutants like p-nitroaniline, p-aminophenols and acetanilide in the presence of photo-Fenton reagent. Characterization of the degradation performance of the sulfamethazine antibiotic by the photo-Fenton process has been carried out by Moya et al.[57]

The main advantage of photo-Fenton reaction is the regeneration of the consumed ferrous ions on irradiation with light. Each  $\text{Fe}^{2+}$  ion can produce many  $\cdot\text{OH}$  radicals in contrast to the dark Fenton reaction. The process is a cyclic one. Only a single  $\cdot\text{OH}$  radical is being formed by one ferrous ion. It means that the amount of ferrous salt required under photo-Fenton condition is small as compared to Fenton condition.

## 2.3 Sono-Fenton Process

The oxidation of organics by ultrasound has received considerable attention because of rapid degradation of chemical contaminants. A chemical effect of ultrasound was first reported by Richard and Loomis.[58] Smith and Fry[59] reported the use of ultrasound in all areas of general surgery and many surgical subspecialties.

Ultrasound is a sound wave with a frequency greater than the upper limit of human hearing (approximately 20 kHz). In practice, three frequency ranges of ultrasound are reported for three different uses:

- i. The relatively low-frequency range, which is applied for conventional power ultrasound (20–100 kHz).
- ii. The medium frequency range, which is used for sonochemical effects (300–1000 kHz).
- iii. The high-frequency range, which is typically used for diagnostic imaging (2–10 MHz).



It is rather an inexpensive reagent for wastewater treatment. Sonolysis of water yields hydroxyl radicals and hydrogen atoms. Hydroxyl radicals particularly are very reactive and can transform organic pollutants.[60]



Sonochemical degradation of some triphenylmethane dye was reported by Panwar et al.[61] The removal of methylene blue and brilliant blue from aqueous medium using the photo-Fenton process in the presence of ultrasonic irradiation was reported by Vaishnave et al.[62, 63] The decomposition of azure-B by photo-Fenton reagent in the presence of ultrasound in homogeneous aqueous solution was studied by Vaishnave et al.[64]

Papic et al.[65] suggested the sonochemical effectiveness factor in the reactor for wastewater treatment by sono-Fenton oxidation. Degradation of C. I. acid orange-7 by heterogeneous Fenton oxidation in combination with ultrasonic irradiation was reported by Zhong et al.[66]

## 2.4 Photocatalysis

In recent years, the use of semiconducting oxides as photocatalyst for degradation of pollutants has attracted the attention of the scientific community all over the world. The photocatalytic bleaching was found to be the most promising and efficient process in controlling environmental pollution, wastewater treatment, etc. The commonly used semiconducting oxides are  $\text{TiO}_2$  and  $\text{ZnO}$ .

Wei et al.[67] have reported the photocatalytic oxidation of phenol in the presence of  $\text{H}_2\text{O}_2$  and  $\text{TiO}_2$  powder while the photocatalytic bleaching of basic blue-24 in the presence of  $\text{ZnO}$  has been reported by Ameta et al.[67]

Vione et al.[68] used phenol as a model aromatic molecule and  $\text{TiO}_2$  as a photocatalyst to carry out aromatic photochemical nitration in homogeneous and heterogeneous aqua systems. Photocatalytic degradation of azo dye acid in water using  $\text{ZnO}$  as photocatalyst has been investigated by Daheshvar et al.[68] Kim et al.[69] used  $\text{ZnO}$  coated  $\text{TiO}_2$  nanoparticles for the flexible dye-sensitized solar cells. Several metal oxides can act as photocatalyst but zinc oxide is generally used for degradation, because of its capacity to breakdown pollutants into simpler molecules and even accomplish total degradation. Nanoparticles of  $\text{ZnO}$  have been investigated as photocatalysts that can be applied for degradation of dyes, drugs and other pollutants.[70] The structural, morphological and optical properties of the optimized  $\text{ZnO}:\text{TiO}_2$  as coupled metal oxide with Gd doping have been investigated by panwar et al.[71] Exhaustive researches in the field of photocatalysis have shown various fascinating applications of photocatalytic reactions based on the use of semiconductors.[72] Use of semiconducting iron (II) oxide in photocatalytic bleaching of some dyes like malachite green, crystal violet and methylene blue has been reported by Ameta et al.[73]

Photocatalytic bleaching of toloum chloride on zirconium phosphate was carried out in presence of light was studied by Panwar et. al.[74] Photocatalytic bleaching of nigrosin-WS by  $\text{CeFeO}_3$  photocatalyst has been done by Sharma et al.[75] Iron-catalyzed photooxidation of azure-A with peroxydisulfate ion was studied by Soni et al.[76] Although a number of photocatalysts have been used for the treatment of dye-containing wastewater effluents, but, no attention has been paid to the use of colored oxides like  $\text{NiO}$  and  $\text{CuO}$  as photocatalyst to extend the wave length into the visible region. The use of colored semiconductor oxide  $\text{NiO}$  and  $\text{CuO}$  for photocatalytic bleaching of dye rose Bengal was investigated by Malkani et al.[77]

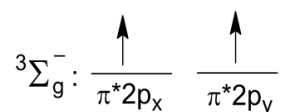
The homogeneous AOPs employing peroxydisulfate[78] have been found to be very effective for degrading dyes and pollutants. Lin et al.[79] investigated the removal of trace level amount of twelve sulphonamides from drinking water by UV activated peroxymonosulfate. Photochemical oxidation of methylene blue was investigated in the presence of  $\text{Fe}^{2+}$  and peroxydisulphate ions at normal temperature and pressure were reported by Soni et al.[80]

## 2.5 Singlet Molecular Oxygen

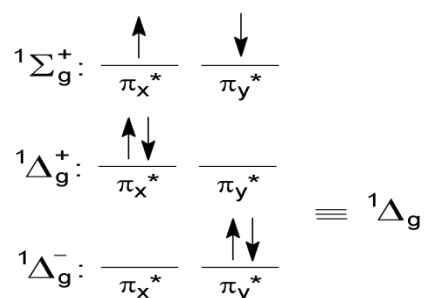
Singlet molecular oxygen has been used as an effective photooxidant for carrying out a number of chemical reactions. It has been found to be an active oxidizing species for a number of biological compounds.[81, 82] Singlet molecular oxygen, the excited state of molecular oxygen, has been used as an oxidizing agent to oxidize various organic and inorganic substrates like pyrrole[82], fullerenes[83] etc.

Photodegradation of tyrosine derivatives by singlet molecular oxygen in the presence of cationic and neutral micellar systems have been reported by Criado et al.[84] Bhatnagar et al.[85, 86] reported the photooxidation of some pharmaceutical drugs (thiopentone, phenytoin, benzylpenicillin, and streptomycin) by singlet molecular oxygen.

The molecular orbital configuration for oxygen is  $\text{KK } \pi_{2s}^2 \pi_{2s}^{*2} \pi_{2pz}^2 (\pi_{2px})^2 (\pi_{2py})^2 (\pi_{2px})^1 (\pi_{2py})^1$ . The last two valence electrons enter one each into the degenerated  $\pi_{2px}$  and  $\pi_{2py}$  pair of orbitals with parallel spin giving triple ground state  $^3\Sigma_g^-$ , which is represented as:



The two electronically excited states are  $^1\Delta_g^+$  and  $^1\Delta_g^-$ . The pairing of two electrons in one molecular orbital result in the  $^1\Delta_g$  state. This is a doubly degenerated state ( $^1\Delta_g^+$  and  $^1\Delta_g^-$ ). The spin pairing in different orbitals results in the  $^1\Delta_g^+$  state. These states can be represented as:



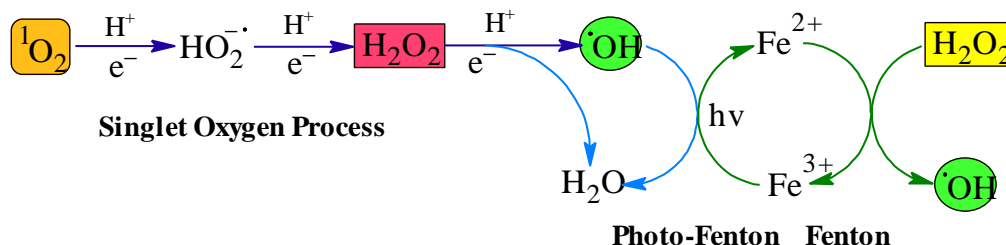
Klotz et al.[87] highlighted the toxic and signalling effects of photochemically or chemically generated singlet molecular oxygen in biological systems. So, it is quite necessary that one must understand the mechanism of oxygen involvement in biological systems and its role in the atmosphere.

## 2.6 Singlet Oxygen-Photo-Fenton Process

The reaction of singlet oxygen is carried out at low temperature and the photo-Fenton reaction takes place at the room temperature. If the photo-Fenton reaction is carried out at a low temperature, then the rate of reaction become very low. And if the singlet oxygen reaction is carried out at normal temperature, then rate of



reaction also become very low. But when both reactions are done together, it is called singlet-oxygen-photo-Fenton process. In this reaction, the rate of reaction is greater than the rate of reaction of both. Because singlet oxygen and photo-Fenton form the same intermediate ( $\cdot\text{OH}$  radical). That's way the rate of reaction of singlet-oxygen-photo-Fenton process is more. Both the reactions support each other. Singlet molecular oxygen with photo-Fenton reaction accelerates the rate of photochemical oxidation of acriflavine was studied by Kumar et al.[88]. This can be explained by the following diagram.



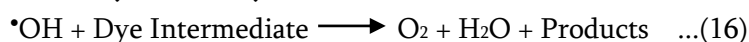
**Fig. 4: Mechanism of Singlet-oxygen-photo-Fenton process.**

## 2.7 UV-Based Processes

The UV oxidizer system involves direct excitation of the substrate due to the radiation with the subsequent oxidation reaction. There may be synergism also between the oxidizer and the ultraviolet radiation, which causes the global effect to be different from the additive effect.

### 2.7.1 Fenton/UV

Vujevic et al.[89] investigated decolourization of reactive blue-2 dyes in aqueous solution by a combination of UV radiation and Fenton process. In this process, the  $\cdot\text{OH}$  radicals are produced directly and it react with dye molecules (eq. 14 - 16).



### 2.7.2 UV/O<sub>3</sub> Process

The UV/O<sub>3</sub> system is an effective method for the oxidation and destruction of organic compounds in water. Normally, aqueous systems saturated with O<sub>3</sub> are irradiated with UV light.

The AOP with UV radiation and ozone is initiated by the photolysis of ozone. Peyton and Glaze[90] investigated that the photodecomposition of ozone leading to two hydroxyl radicals, which do not act as oxidant become they recombine to produce hydrogen peroxide.



This system contains three components to produce  $\cdot\text{OH}$  radicals and to oxidize the pollutant for subsequent reactions: UV radiation, ozone and hydrogen peroxide. Guittonneau et al.[91] reported that the UV/O<sub>3</sub> process was found to be more efficient than the UV/H<sub>2</sub>O<sub>2</sub> system for the degradation of p-

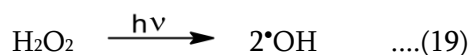
chloronitrobenzene. The viability of the ozonation and ozone combined with UV photolysis processes was investigated for the degradation of Direct blue-86 dye in wastewater.[92]

### 2.7.3 UV/O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> Process

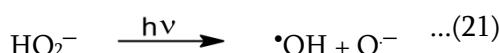
The addition of H<sub>2</sub>O<sub>2</sub> to the UV/O<sub>3</sub> process accelerates the decomposition of ozone resulting in the increased rate of •OH radical generation. This is a very powerful method that allows a considerable reduction of the TOC. Trapido et al.[93] reported that the combination of ozone with UV radiation and hydrogen peroxide was found to be more effective for the degradation of nitrophenols. Contreras et al.[94] demonstrated that the addition of H<sub>2</sub>O<sub>2</sub> to UV/O<sub>3</sub> system slightly improves the rate of TOC removal in solutions of nitrobenzene.

### 2.7.4 UV/H<sub>2</sub>O<sub>2</sub>

The UV/H<sub>2</sub>O<sub>2</sub> processes use ultraviolet radiation to cleave the O–O bond in hydrogen peroxide and generate the hydroxyl radical (•OH).



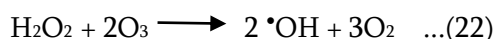
Also HO<sub>2</sub><sup>−</sup>, which is in an acid-base equilibrium with H<sub>2</sub>O<sub>2</sub> absorbs the UV radiation and produce •OH radicals.



The oxidation of reactive blue-19 dye with UV/H<sub>2</sub>O<sub>2</sub> was also carried out by Rezaee et al.[95] The effectiveness of the UV/H<sub>2</sub>O<sub>2</sub> system in the treatment of aromatic compounds such as phenol has been widely studied.[95] Alaton and Balcioglu[96] showed the effectiveness of UV/H<sub>2</sub>O<sub>2</sub> system to pre-treatment or in combination with other advanced oxidation processes in the treatment of textile wastewater. UV/H<sub>2</sub>O<sub>2</sub> process has been successfully used for the destruction of chlorophenols and other chlorinated compounds. The removal of targeted micro-pollutants, including naproxen, carbamazepine, diclofenac, gemfibrozil, ibuprofen, caffeine and mecoprop were reported with photolysis and UV/H<sub>2</sub>O<sub>2</sub> processes.[97]

### 2.7.5 H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> Process

Addition of hydrogen peroxide to ozone offers another way to accelerate the decomposition of ozone, leading to the formation of •OH radicals. Hydrogen peroxide in aqueous solution is partially dissociated in the hydroperoxide radical (•O<sub>2</sub>H), which reacts with ozone, decomposing this and giving rise to a series of chain reactions with the participation of hydroxyl radicals. Glaze and Kang[98] reported that in the global reaction, two ozone molecules produce two hydroxyl radicals.



This system does not depend on the UV radiation transmission to activate the ozone or hydrogen peroxide molecules, its greatest advantage of being able to work with turbid water without problems. The improved H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> oxidation process was used for wastewater reclamation by Yang et al.[98] while Balcioglu

and Arslan[99] studied the efficiency of ozonation and  $\text{H}_2\text{O}_2/\text{O}_3$  of reactive dyes and textile dye-bath wastewater.

### 2.7.6 UV/ $\text{Fe}^{2+}/\text{O}_3$ Process

A novel combination of wastewater treatment UV/ $\text{Fe}^{2+}/\text{O}_3$  was investigated. Addition of  $\text{Fe}^{2+}$  to the UV/ $\text{O}_3$  process increased the mineralization rate of 4-chlorophenol as reported by Ruppert et al.[100]

## III.DEGRADATION OF SOME ORGANIC COMPOUNDS

### 3.1 Hydrocarbon

Somrani et al.[101] reported the photocatalytic degradation of some hydroxylated bicyclic aromatic compounds (2,2'-dihydroxy biphenyl, 4,4'-dihydroxy biphenyl and 4-hydroxy biphenyl) by  $\text{TiO}_2$  catalyst. Novio et al.[102] observed the partial or complete heterogeneous photocatalytic oxidation of neat toluene and 4-picoline in liquid organic oxygenated dispersion containing pure and iron-doped titania photocatalysts.

Lee et al.[103] suggested the Fenton oxidation process for polycyclic aromatic hydrocarbons by Frontier electron density. Cho et al.[104] treated benzene, toluene, ethylbenzene and xylene with advanced oxidation processes, such as  $\text{TiO}_2$  photocatalysis and  $\text{Fe}^{2+}/\text{H}_2\text{O}_2$  exposed to solar light. Barreto et al.[105] investigated the decolourization of dye solutions of different DHBs (dihydroxybenzene) and gallic acid (trihydroxybenzene) by Fenton systems ( $\text{Fe}^{2+}/\text{H}_2\text{O}_2$ ,  $\text{Fe}^{3+}/\text{H}_2\text{O}_2$ ).

### 3.2 Carboxylic Acids

A recent hydroxylation example involving the oxidation of barbituric acid to alloxane by Fenton process was studied by Bromme et al.[106] Photo-oxidation of p-aminobenzoic acid by Fenton reagent was reported by Kumar.[107] Zantal and Martinez-Huitleii.[108] degraded 2-hydroxybenzoic acid by Fenton and UV/ $\text{H}_2\text{O}_2$  processes. Benitez et al.[109] reported the degradation of gallic acid (3, 4, 5-trihydroxybenzoic acid) which is a major pollutant present in the wastewater in aqueous solution by UV/ $\text{H}_2\text{O}_2$ , Fenton's reagent and photo-Fenton system. Ustun et al.[110] studied degradation and mineralization of 3-indole butyric acid using Fenton and Fenton-like processes.

The photo-Fenton process using potassium ferrioxalate as a mediator was employed for the photodegradation of dichloroacetic acid and 2,4-dichlorophenol by Nogueira et al.[111] Yue et al.[112] worked at the photo-assisted Fenton degradation of salicylic acid using strongly acidic ion exchange resin (SAIER) exchanged with Fe ions as catalyst in the presence of UV light (254 nm) and  $\text{H}_2\text{O}_2$ . Oxalic acid destruction by combined heterogeneous photocatalysis and photo-Fenton reaction using UV/ $\text{Fe}/\text{H}_2\text{O}_2$  and the UV/ $\text{TiO}_2/\text{Fe}/\text{H}_2\text{O}_2$  was studied by Litter et al.[113]

Photo-Fenton-assisted ozonation of p-coumaric acid present in olive oil mill wastewater was investigated by Monteagudo et al.[114] The photo-Fenton degradation of salicylic acid and trichloroacetic acid catalyzed by alizarin violet-3B dye complete mineralization was studied by Lofrano et al.[115]

### 3.3 N-Containing Compounds

Fenton process has proved to be efficient in the removal of colour and chemical oxygen demand from the aqueous environment. This includes the treatment with ferrous oxide through coagulation followed by integrated Fenton process. The aim of this process is removal of azo dye in an aqueous environment.[116] 2, 4-Dinitrotoluene has[117] been completely oxidized by the photo-Fenton process. Fenton, UV/Fenton, and

UV/H<sub>2</sub>O<sub>2</sub> reagents were used to mineralize dinitrotoluenes and trinitrotoluene in toluene nitration process.[118, 119]

The degradation of nitrophenols namely 2-nitrophenol, 4-nitrophenol and 2,4-dinitrophenol with Fenton process, solar and UV-assisted Fenton processes was investigated by Lei et al.[120] Oxidation of the nitroaromatic explosives namely 2,4,6-trinitrophenol, ammoniumpicrate, 2,4-dinitrotoluene, 2,4,6-trinitrotoluene (TNT) and hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX) and octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine by Fenton's reagent has also been investigated.[121-124] The photocatalytic oxidation of nitrobenzene and 2,4,6-trinitrotoluene in aqueous suspensions of titanium dioxide was investigated by Chung et al.[125]

3-Aminopyridine is used as an intermediate in the manufacturing of pharmaceuticals compounds. It is one of the emerging pollutants because of its toxic nature, carcinogenic potential and hazardous effect on the natural environment. Its degradation by Fenton and photo-Fenton has been reported by Karale et al.[126] Nitrogen containing heteroaromatic pharmaceutical compounds like pyridine have received a large attention recently because their presence in the environment causes toxic, carcinogenic and hazardous effect on natural environment and discharge of pyridine-containing waste does great damage to human health and the environment.[127] The wastewater generated from pyridine and cyanopyridine manufacturing plants is highly complex in nature. So Padoley et al.[128] investigated pretreatment of wastewater containing pyridine and 3-cyanopyridine by Fenton oxidation.

### 3.4 Alcohols

The oxidative degradation of polyvinyl alcohol by the photochemically enhanced Fenton reaction was observed by Braun et al.[129] and the photo-Fenton process was studied by Guardani et al.[130] Winery wastewater being seasonal and experiencing substantial flow variations are difficult to treat by conventional biological process. The photo-Fenton reaction in homogeneous and heterogeneous phase was studied by Ovellerio et al.[131] and Mosteo et al.[132] respectively.

Hirashima et al.[133] worked at the photocatalytic oxidation of alcohols. They found that alcohols were oxidized to the corresponding carboxylic acid in the presence of a catalytic inorganic bromo source, for example, lithium bromide, bromine and hydrobromic acid, under photo-irradiation. Photooxidation of alcohols to the corresponding aldehydes with a porphyrin/quinone/TEMPO (TEMPO = 2,2,6,6-tetramethyl-1-piperidinyloxy free radical) system has been described by Nagata et al.[134] This photoreaction is a combination of a photo-induced electron transfer from the porphyrin to the quinone and a TEMPO-catalysed liquid-phase selective photooxidation of 2-butanol (crotyl alcohol) to 2-

butenal crotonaldehyde and gas-phase selective photooxidation of 2-propanol to acetone. Fenton reagent is used in organic synthesis in coupling reactions. As an example, tert-butanol is dimerized with Fenton's reagent and sulfuric acid to 2,5-dimethyl-2,5-hexanediol.[135] Walling and Kato[136] investigated oxidation of methyl alcohol, ethyl alcohol, isopropyl alcohol and t-butyl alcohol by Fenton reagent.

### 3.5 Phenols

Phenols are widely used and have become common pollutants in water bodies.[137] These compounds are highly toxic and biorefractory products. They often appear in the effluents from many heavy chemical plants, petrochemical plants and oil refining industries, which can cause adverse effects on the aquatic ecosystems and human health. Because of its toxicity and less biodegradability, phenolic wastewater must be

specially treated before disposing off. Various effective and economic treatment methods have been applied to remove or eliminate phenolic chemicals from wastewater.

The role of  $\text{Fe}^{2+}$  ion in Fenton and photo-Fenton processes for the photochemical oxidation of phenol was examined by Kavitha and Palanivelu.[138] Photochemical oxidation of phenol by solar photo-Fenton using compound parabolic collectors reactor has been carried out by Alalm et al.[139]

Phenol degradation by photo-Fenton reactions in highly concentrated wastewater and most intermediate species have been identified by Fourier Transform IR-spectroscopy with ATR device and reported by Pena et al.[140] The degradation of p-chlorophenol using the combination of UV/ $\text{H}_2\text{O}_2$  and the photo-Fenton reaction were investigated on lab-scale.[141] The photo-Fenton process was also used for the degradation of phenol red by Punjabi et al.[142]. Photo-Fenton treatment of water containing natural phenolic pollutants like vanillin, protocatechuric acid, syringic acid, p-coumaric acid, gallic acid and L-tyrosine was studied by Malato et al.[143]

The degradation kinetics and mechanism of phenol in photo-Fenton type processes;  $\text{Fe}^{2+}/\text{H}_2\text{O}_2$ ,  $\text{Fe}^{3+}/\text{H}_2\text{O}_2$ , UV/ $\text{Fe}^{2+}/\text{H}_2\text{O}_2$ , UV/  $\text{Fe}^{3+}/\text{H}_2\text{O}_2$  and UV/ $\text{Fe}^0/\text{H}_2\text{O}_2$ , and  $\text{Fe}^0/\text{H}_2\text{O}_2$  was carried out by Le-Cheng and Feng.[144] Heterogeneous photo-Fenton degradation of phenolic aqueous solution over iron-containing SBA-15 catalyst was carried out by Melero et al.[145] Degradation of cresols in photo-Fenton system was reported by Kavitha and Palanivelu.[146] Role of intermediate in the degradation of 4-chlorophenol, 4-nitrophenol and phenol by Fenton process was investigated by Selanec et al.[147]

Molina et al.[148] suggested the iron species incorporated over different silica support for the heterogeneous photo-Fenton oxidation of phenol. Photo-Fenton and photodegradation of bisphenol-A with iron oxides in aqueous solution was investigated by Li et al.[149] and Katsumata et al.[150] Catalytic effect of inorganic additives like sodium thiosulfate and potassium bromate on the photo-Fenton degradation of phenol red was observed by Jain et al.[151]

Degradation of p-chlorophenol by electro-Fenton and photo-Fenton process under acidic condition using batch recirculation reactor has been studied by Manivasagan et al.[152] Degradation of 2,4-dichlorophenol by  $\text{H}_2\text{O}_2/\text{UV-C}$ , Fenton, photo-Fenton and toxicity has been evaluated by Karci et al.[153] Wang et al.[154] investigated the effect of amount of  $\text{H}_2\text{O}_2$  and pH on the photo-Fenton oxidation of catechol using nano- $\text{Fe}_3\text{O}_4$  as photocatalyst. Photochemical treatment of a phenolic compound tyrosol (present in olive mill wastewater) by  $\cdot\text{OH}$  and sulphate radical based AOPs was carried out by Kilic et al.[155] The use of immobilized rose Bengal in the oxidation of phenol was studied by Nowakowska and Kepczynski.[156] The use of Fenton reaction in the decomposition of phenol and formaldehyde from their aqueous solutions and from industrial wastewater containing them was studied by Rashed et al.[157]

### 3.6 S-Containing Compounds

The oxidation of sulfide salts to sulfate in aqueous solution is also important in wastewater treatment. In a study by Iliev et al.[158],  $\text{Zn(II)-2,9,16,23-phthalocyanine tetracarboxylic acid}$  was found to be effective in the photosensitized oxidation of both; sulfide and thiosulfate to sulfate. Sulfamethazine antibiotic was degraded using photo-Fenton process by Graells et al.[57] Oxidation of methionine residues by  $^1\text{O}_2$  can produce methionine sulfoxide (Met-SO) that can be further oxidized to methionine sulfone.[159]

### 3.7 Pharmaceuticals

Hospitals and pharmaceutical industries are the main sources of wastewater effluents so the photodegradation of pharmaceuticals is of great interest. They have detrimental effects on aquatic organisms, water ecosystem and contaminated the soil including toxicity, endocrine disruption, and genetic damage.[160]

Li et al.[161] have reported degradation of pharmaceutical compounds and effluent industrial organic matters by Fenton treatment. Under dark light and solar irradiation, photodegradation of the pharmaceuticals bezafibrate, paracetamol and amoxicillin in aqueous solution via the photo-Fenton process was studied by Gustavo et al.[162]. Simultaneous degradation of the antibiotics ciprofloxacin, sulfathiazole, sulfamethazine and removal of hospital effluent after biological treatment via photo-Fenton/UV process was studied by Perini et al.[163].

Ampicillin has been detected in wastewater effluents. The solar photo-Fenton oxidation process was used for the removal of ampicillin, resistant *E. coli*, total cultivable and eco-toxicity from secondary treated wastewater effluents under various experimental conditions by Ttofa et al.[164] Souza et al.[165] investigated the removal of 3-amino-5-methylisoxazole, which is a recalcitrant by-product resulting from the biological breakdown of some pharmaceuticals, by applying a solar photo-Fenton process assisted by ferrioxalate complexes (SPFF) and classical solar photo-Fenton process (SPF). The degradation of atenolol and metoprolol pharmaceutical compounds in aqueous solutions by means of the photo-Fenton reaction was investigated by Veloutsou et al.[166]

### 3.8 Pesticides and Herbicides

The photodegradation of pesticides is of great interest because of the scale of agriculture and the intensive use of chemicals. Pesticides are however synthesized not to, photodegrade readily in sunlight in order to allow them to exert their biocidal activity. Thus, additional modalities are implemented to enhance their photodegradation, including the use of photosensitizers, photocatalysts (e.g., titanium dioxide) and the addition of reagents such as hydrogen peroxide that would generate hydroxyl radicals that would attack the pesticides.[167] Malato et al.[168] reported the degradation of water-soluble pesticides like diuron, imidacloprid, formetanate and methomyl by photo-Fenton and TiO<sub>2</sub> using solar energy. Organophosphorus pesticide, fenthion was photodegraded using singlet molecular oxygen by Hirahara et al.[169]

## IV. MISCELLANEOUS

The discharge of industrial wastes directly into water bodies has raised serious problems during the last few decades. Due to the increased disposal of organics, there is a pressing demand for newer technologies, so Ameta et al.[50] suggested Fenton and photo-Fenton reagents for the complete mineralization of wastewater. Water pollution caused by organic wastewater has become a serious concern worldwide. Fenton optimization processes is a novel and highly efficient organic wastewater treatment method. The feasibility and application of the coupled Fenton optimization processes (photoelectro-Fenton, heterogeneous electro-Fenton, heterogeneous photo-electro-Fenton, three-dimensional electro-Fenton) for organic wastewater treatment were discussed by Zhang et al.[170]

Esplugas et al.[171] compared different AOPs for phenol degradation. Sarria et al.[172] treated the bio-recalcitrant wastewater generated in textile activities using photo-Fenton and the biodegradability of the



photo-treated solution. The degradation of the organic contaminants in paper pulp effluents by Fenton and photo-Fenton was studied by Perez et al.[173]

Pulgarin et al.[174] studied new integrated  $\text{Fe}^{3+}$  ion photo-assisted biological treatment and mineralize the 5-amino-6-methyl-2-benzimidazolone. Bozzi et al.[175] compared the Fenton homogeneous reactions with superior biodegradability mediated by immobilized Fe-fabrics of wastewater. Gurses and Arslan-Alaton.[176] degraded the procaine penicillin-G formulation effluent using  $\text{Fe}^{3+}/\text{H}_2\text{O}_2$  and  $\text{Fe}^{3+}/\text{H}_2\text{O}_2/\text{UV}$  processes while Nogueira et al.[177] used  $\text{Fe}(\text{NO}_3)_3$  and complex ferrioxalate ( $\text{FeOx}$ ) as different iron sources for the photochemical oxidation of 4-chlorophenol, dichloroacetic acid, malachite green, formaldehyde and herbicides (diuron and tebuthiuron). The degradation of p-nitroaniline[178] in water, and formaldehyde[179] in the presence of methanol by solar photo-Fenton was also investigated. Matilainen and Sillanpaa[180] removed the natural organic matter from drinking water using Fenton and photo-Fenton reagent. Cruz et al.[181] highlighted the possibility of the degradation of micropollutants such as pharmaceuticals, pesticides, etc. in the presence of dissolved organic matter at near  $\text{pH} = 7.0$ . Ebrahiem et al.[182] investigated removal of residuals organic pollutants present in cosmetic and industrial wastewater by applying photo-Fenton oxidation technology.

Pollutants can be removed by number of methods like adsorption, burning, filtration etc. but advance oxidation processes have got an edge over these methods because of low cost, easiness and eco-friendly nature. Although various advance oxidation processes are there, but Fenton reaction is preferred over other reactions. It was planned to carry out photochemical degradation of some organic pollutants by photo-Fenton, singlet molecular oxygen and their combinations. Gentian Violet, Acriflavine and Evan's Blue were selected as model contaminants for the present study.

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