

A Review on Types and Applications of Advanced Oxidation Processes

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ABSTRACT

Due to rapid increase of the population, the protection and conservation of natural resources becomes very important and hence also difficult. Among these resources, water related crisis alarming globally. In order to protect such resources and its management, we need to treat our wastewater before direct disposal into river streams. So, there are several methods for treatment of water and wastewater like biological methods, physicochemical methods, conventional methods etc., but some highly stable and persistent compounds are not easily treated by such methods. For that advanced treatment technology either alone or in combinations are required. Among the most widely used techniques, advanced oxidation processes (AOPs) is considered as the very good and efficient methods for the removal of such highly chemical stable compounds from the waste water. Currently, several researchers are focusing on this technology and are working on the improvement by providing the solutions of its limitations. The present study, a brief overview of advanced oxidation processes (AOPs) and its applications with suitable examples discusses and also highlights the merits and demerits of the process.

Keywords: Advanced oxidation processes (AOPs), Recalcitrant compounds, Fenton process, Wastewater treatment, refractory compounds.

INTRODUCTION

Water is an essential natural resource used by all living for various functions to sustain life in ecosystem. Out of total quantity of water, 97% exists in oceans and remaining 3% present on earth's

surface as freshwater. Out of total amount of fresh water, only 0.65% of the water is directly used by human beings for different purposes [1]. This valuable asset is limited and many countries across the globe are facing severe shortage. About 2.8 billion people in approximately 48 countries in the world are expected to face grave problem of availability of freshwater on earth by 2025[2]. India is one of the countries which is also facing the scarcity of potable water. As the population and development of India is increasing, the per capita demand of the water is also increasing and hence fresh water availability is becoming scarce. This availability of the freshwater for an average annual per capita, has been reducing since 1951 from 5177 m³ to 1869 m³, in 2001 and 1588 m³, in 2010. And in 2025 and 2050, It is projected to reduce further to 1341 m³ and 1140 m³ respectively [3]. The resources which provide water are highly under pressure to overcome this scarcity in order to meet the growing demand of potable water of the population and climate change [4][5]. The given graph (Fig.1) shows the demand of the fresh water as per CPCB report, 2010. According to CPHEEO, 70-80% of the supplied water to the domestic use is converted into wastewater. Nearly, 61754 MLD sewage generates for that there are 920 sewage treatment plants in India, having sewage treatment capacity 22963 MLD in which 38% is treated 38791 MLD [6]. Hence, to address this problem, there is an urgent need for efficient water resource management and treatment of the waste water, contaminated by various activities of human beings.

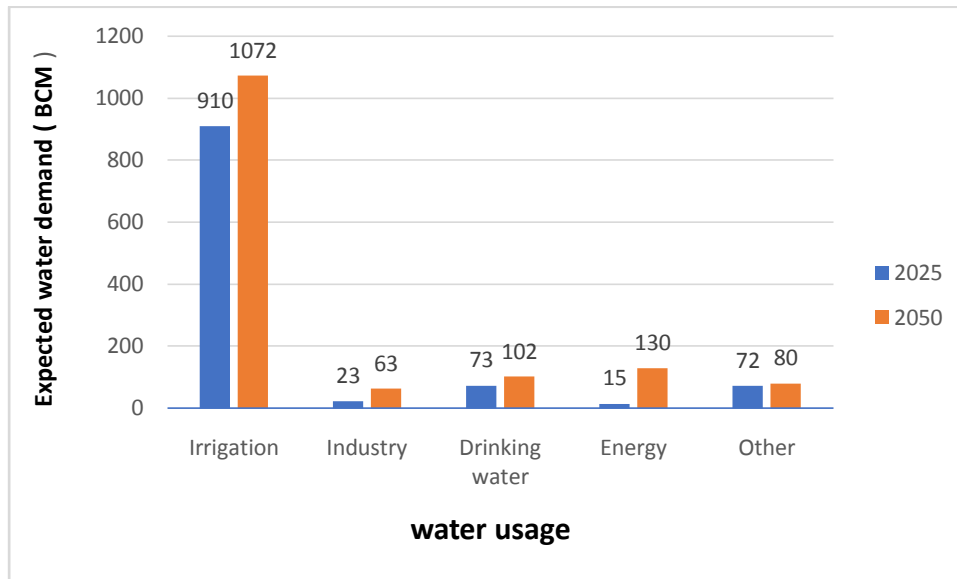


Fig.1 Demand of the fresh water as per CPCB report ,2010

The researchers have been searching and applying the new technologies in the field of treatment of the wastewater in the last many years. There are many methods and treatment processes like member treatment technologies, desalination processes including reverse osmosis, electrodialysis, ion exchange, freeze desalination etc. [7][8], and conventional systems including physical(Screening, Sedimentation,Floatation, Filtration, Absorption,

Adsorption, Centrifugation etc.), chemical (Coagulation, Absorption, Oxidation-Reduction, Ion-exchange, Disinfection) Biological (Aerobic Treatment, Anaerobic Treatment) and combination processes for the removal of solids, organic matter and sometimes nutrients from the wastewater[9].The table 1 shows the suitability of the various water treatment options for the various wastewater [10].

Quality of wastewater	Treatment Options
Low organic effluent	Chemical treatment
Organic effluent	Combine aerobic or anaerobic treatment
Highly organic effluent	Chemical Oxidation by H ₂ O ₂ or O ₃ or NaCl Chemical + biological treatment
Inorganic salts	Solar evaporation forced evaporation Membrane separation
Refractory	Chemical oxidation + biological treatment

Table-1 various water treatment options for the various wastewater

The conventional system for the treatment of the wastewater is always not suitable because of the complex operation and high maintenance cost. In India it has been seen that removal of sludge, its treatment and handling is being neglected in the operation of the sewage treatment plants. Furthermore, many treatment plants remain closed most of the time because of the plant design is not

appropriate, existing facilities do not work properly, maintenance is poor, irregular availability of the electricity etc. One of the major issues with this method of the treatment is that there is no economical return and hence no local authorities do not want to take up such wastewater treatment.

In case of the Pharmaceutical and other compounds like phenols, drugs, pesticides, consumer

products, and industrial chemicals the biological treatment processes are not able to oxidized easily due to low biodegradability problem [11][12]. Hence, the existing plants are being redesign and upgrading by implementing new technologies which should be environmental benefits and human health protection [13][14]. Amongst the various advanced competing technologies, The Advanced oxidation processes is one of the most widely and preferably used for the treatment methods for the such compounds in water. The objective of the present study is to review the art of the Advanced oxidation process and its application in the various waste water treatment. The process requires many improvement and has to work on its limitations.

1. Advance Oxidation Processes (AOPs)

Earlier, Advanced Oxidation Processes used to treat portable water only. Later, AOPs were widely adopted to treat different types of wastewater too. This method is completely based on in-situ generation of strong oxidizing agents namely hydroxyl radicals (OH^*). Advanced oxidation Processes (AOPs) are the oxidation of the various highly chemically stable compounds such as recalcitrant, refractory, POPs etc., present in the

wastewater which cannot be treated easily by any other conventional systems or methods [15][16]. In this process, high oxidizing agents such as hydroxyl radicals (later extended for the sulphate radicals) [17] are formed in sufficient quantity and remove such compounds from wastewater [18]. This method of treatment was first introduced in 1980s only for the treatment of the potable water but due to lot of advantages, it was started later for treatment of the wastewater [19][20] and till today, this method has been using widely and effectively. Because of simplicity in its operation, pathogens inactivation and high proficiency in organic mineralization, this technique is most potentially used in the post-tertiary sewage treatment.

The steps involved in AOPs are explained below:

1. Strong oxidants are formed such as OH^*
2. The reaction of these oxidants produces biodegradable intermediates.
3. These biodegradable intermediates are then reacted with these strong oxidizing agents and convert into minerals.

The general reaction involve in the AOPs can be shown in Fig 2:

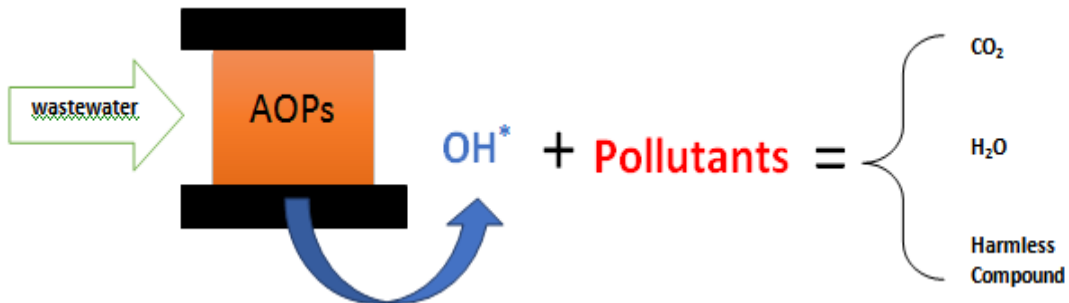


Fig. 2 Reaction involve in AOP

Earlier, the aim of the AOPs is to improve the biological process for the treatment but later s these are adopted for the degradation of the highly stable and low degradable compounds [21] [22].

The strong bonds (like azo) are broken by the pre-biological treatment process which uses the biodegradable compounds and then Advanced oxidation process is applied for the treatment of the toxic aromatic amines and other recalcitrant compounds which produce due to the bioremediation process [23]. The AOPs should not be applied before the pretreatment process because of generation of the

highly stable intermediates which are more toxic to the biological system and less biodegradable than the actual molecules [24]. Furthermore, the high amount of the oxidants used during the oxidation process of AOPS may results condition of the toxicity to the microorganism [25]. Due to the high amount of the chemicals used during process, Pretreatment of AOPs is become more costly, therefore it is proposed that pre-biological treatment for the wastewater is beneficial to use for AOPs treatment [26][27][28].

2. Characteristics of the OH radical

The primary role of an AOP system is to produce a strong oxidizing agent such as OH^{*} agent so that low biodegradable pollutant can be removed effectively which cannot be treated by conventional methods.

The property of the generated hydroxyl radicals during the process is very important for efficiency of the AOPs. The oxidizing potential of the OH^{*} is of the order of 2.8 V-1.95 V (pH=0 to 14). The fig.3 shows the various oxidizing agents with their oxidizing potential [29]. OH^{*} is non-selective in nature and reacts with other species with very fast rate (reaction rate constant $10^8-10^{10} \text{M}^{-1} \text{s}^{-1}$). The radicals react on the organic pollutant in four ways – hydrogen's abstraction, transferring an electron, the addition of radicals and radical combination [30]. When OH reacts with organic compound it leads to the production of Alkyl Free Radical (R) or Alcohol (R-OH). These might react with oxygen transforming into organic peroxy radical (ROO^{*}). Hydrogen peroxide and superoxide, which comes in the group of more reactive species, are generated when further reactions take place which can degrade recalcitrant organics like phenolic and organochloro-compounds. The process transforms complex organic compounds into more biodegradable organic compounds which are easily removable in biological treatment. OH^{*}

having a very low lifetime requires in situ production by assortment of irradiation, catalysts and oxidizing agents [31]. A complicated downpour of oxidative reactions take place with the initiation of free radical by H₂O₂ or ozone. The initiated radicals degrade the target compounds quickly. The kinetic rate of degradation of AOPs relies on concentration of pollutant, temperature, reactants and presence of varied scavengers like bicarbonate ion [32]. It has also been noticed that AOPs can give better COD deduction when merged with biological treatment (as pre or post treatment) as compared to any standalone technology under identical dispositions. Membrane Bioreactors (MBRs) have arisen to be a reliable developed biological treatment procedure. As recorded by Ioannou and Fatta-Kassinos (2013), COD disposal of around 70% was accomplished on treatment of waste water with solar photo-fenton method which was pre-treated by MBR, in which the primary COD removal (58%) was attained in initial 30 min of solar treatment [33]. The primary drawback of membrane bioreactor is the issue of membrane fouling which pivots on microbial population pattern and microbial cell density. It was recorded that nitrifying activated sludge, created at short C/N ratio, considerably reduced the issue of fouling and boosted nitrifying efficiency [34].

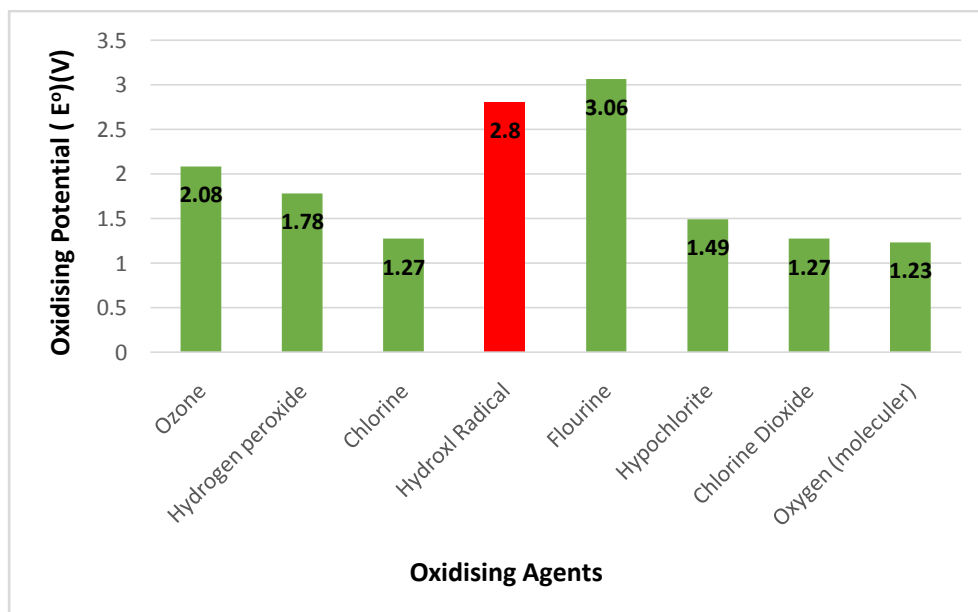


Fig: 3 Oxidizing Agents with their Oxidizing potential

3. Application of the AOPs

The following are some applications of the AOPs discussed below but they are not limited:

3.1 Micro pollutant Treatment:

Micro pollutants are pollutants that are present in water in low concentrations e.g. volatile organic compounds, herbicides, endocrine-disrupting compounds, pharmaceuticals etc. Several micro pollutants potentially effect on health because of not easily treated and detected by conventional methods. The endocrine-disrupting compounds like dioxine are one of the examples which affect the hormonal system of the mammals and can cause the several diseases and disorders[35].

3.2 Removal of refractory compounds from dye wastewater:

The combined processes of biological, physical, and chemical treatment methods have been developed[36][37] to remove refractory compounds from textile wastewater particularly azo dyes ,(i.e., molecules that have one or more azo (N=N) bridges that link substituted aromatic structures) [38]. These methods are efficient but not good from economic point of view. On the other hand, traditional physicochemical processes such as membrane filtration, ion-exchange, and adsorption, have been efficiently applied but these processes are not attractive commercially. In contrast, advanced oxidation processes (AOPs) uses the strong oxidizing agent and oxides such compounds easily.

3.3 Treatment of compounds produce Taste-and-Odor:

Taste and odor compounds may produce in drinking water due to growth of algae in warmer season. These compounds have not profound effect on human health because of their presence in less concentration but due to presence of some algal toxins such as microcystin may have adverse effect on human health [35].

3.4 Treatment of Recycled Water:

There are several techniques to treat or remove the pollutants in the wastewater but still there are many contaminants such as N-nitrosodimethylamine which are not typically found in drinking water applications. Therefore, AOPs is a good method (whether the water is directly or indirectly supplied) to remove such micropollutant from the water [35].

3.5 Destruction of terephthalic acid (TPA): -

Terephthalic acid (TPA) is widely used for the manufacturing of polyester fiber, polyethylene terephthalate bottles, polyester films, etc. It is a toxic compound and can have effect such as irritation on skin, nose, throat and eyes, coughing, and shortness of breath. Kidneys may also be affected by the repeated exposure to TPA. The wastewater containing the TPA is treated by biological processes but advanced oxidation processes such as photocatalytic oxidation, ozonation, Fenton oxidation etc. have been effectively used for its removal from wastewater[39][40].

3.6 Paper and pulp mills: -

The effluent from Pulp and paper mills may contain the wide variety of contaminants such as hemicelluloses, lipophilic extractives, lignin, lignin-related substances, and carboxylic acids etc. [41] which depends on the type of pulping process and the following treatment. [42] Moreover, it has been studied that more than 40 % low biodegradable compounds are produced during such pulping processes as by products such as [43], sulfite processes which produce compounds of low biodegradability[44]. Hence the application of the advance oxidation process becomes effective for the treatment of this type of wastewater as an additional process with conventional chemical and/or biological processes.

3.7 Landfill leachate:

When water passes through a solid waste and the water content is above to field capacity then a liquid formed known as leachate. Landfill leachate may be defined as highly polluted and toxic wastewater containing a variety of organic and inorganic waste. It is considered in the category of high strength wastewater. The large recalcitrant organic compounds present in leachate not easily removed by biological treatment processes. Advanced oxidation methods such as ozone with hydrogen peroxide, ozone with ultraviolet light, Fenton process etc. has been studied in the last many years for the treatment of such wastewater to improve the removal capacity of the large recalcitrant organic molecules or to convert into them into easily biodegradable matters [45].

3.8 Petroleum Industry:

The wastewater generated from the petroleum industry contains a high volume of hydrocarbons, if not treated well may have serious and hazardous environmental impacts. The conventional techniques for the treatment of these water include physical-chemical, mechanical, and biological treatment. These conventional techniques capable to remove free and suspended emulsion oil from the wastewater. When biological treatment is also associated with these methods, can reduce the BOD. But merely the application of the biological process is not adequate for the wastewater containing highly toxic recalcitrant compounds. In this case, more advanced techniques must be used to degrade these compounds [46] and advanced oxidation processes (AOPs) have been proved most effective powerful methods [47][48]

4. Merits and demerits of the AOPs

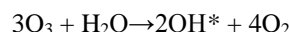
There are many benefits of the AOPs over conventional system of the treatment like a) This method oxidizes the organic pollutants into biologically treated forms and not transfer into other phase b) Very effective in treating most micro pollutants. c) The oxidizing agents produced are non-selective in nature, therefore treat a wide range of contaminants. d) Helps to remove compounds which produces taste-and-odor in drinking water. e) Reactions in AOPs are highly vigorous with a contact times of few seconds in this technique, simultaneous microbial disinfection can be achieved during degradation of contaminants. On the other hands, disadvantages include a) They have high annual operation and maintenance costs. b) The water treated by AOPs should be tested for potential regulated and unregulated by-products. c) Ultraviolet

reactors containing mercury lamps may result in the mercury hazard due to its possible breakage. [35]

5. Methods of treatment: -

Conventionally utilized AOPs are also categorized as homogeneous and heterogeneous procedures relying on the occurrence in single phase or utilizing heterogeneous catalysts like carbon substances, catalysts equipped with metals or semiconductors like oxides of Titanium, Tungsten and Zinc [49]. Some cases of homogeneous procedures are O₃, O₃+H₂O₂, wet peroxide oxidation, photo-Fenton and others whereas heterogeneous procedures comprise of catalytic wet peroxide oxidation (CWPO), catalytic ozonation and others. The classification of the AOPs are shown in the Fig.4

5.1 Ozone: - It is one of the oxidizing agents having strong electrochemical oxidizing potential 2.08 Vs and rate of reaction constant is about equal to the 1.0×10⁰–10³/M/s [50]. The use of the ozone is not limited to disinfection but it is also used for the treatment of the waste water as an AOPs in the presence of the OH⁻ and some other transition metal cations in an alkaline medium. The overall reaction is as follows [51].



However disadvantage of this type of the AOP is that it requires the high energy for the generation of the ozone and the efficacy of the ozone is totally depend on the gas liquid mass-transfer [52]. Furthermore, the oxidizing cost of this process is about 5.35 \$/m³ [53]. This process is most efficiently used for the removal of the various compounds from gasoline such as BTX, MTBE etc. [54]

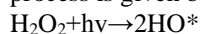
Processes	Reactions	Applications/ salient information	References
O ₃ +UV radiation	H ₂ O+O ₃ +hv →2OH*+ O ₂	Used for the degradation of <ul style="list-style-type: none"> • Dinitrotoluene (DNT) • Trinitrotoluene (TNT) • Dimethyl sulfoxide (DMSO) • Carbofuran (CBF) etc. 	[55][56]

O3 and H ₂ O ₂	$\text{H}_2\text{O}_2 \rightarrow \text{HO}^{\cdot-2} + \text{H}^+$ $\text{HO}^{\cdot-2} + \text{O}_3 \rightarrow \text{OH}^* + \text{O}^{\cdot-2}$	This method is also called peroxone.	[57]
O3 and Ultra sound	$\text{O}_3 + \text{H}_2\text{O} + \text{h}\nu \rightarrow \text{H}_2\text{O}_2 + \text{O}_2$	Used for removal of p-Aminophenol (PAP) compounds (a very harmful compound to the environment, used for the production of the several medicines, leather dyes etc.) [35]	[58]
O3+ UV + H ₂ O ₂	$2\text{O}_3 + \text{H}_2\text{O}_2 + \text{h}\nu \rightarrow 2\text{OH}^* + 3\text{O}_2$	Used widely in textile industries for discoloration of dyes. With combination of H ₂ O ₂ or UV or Ultra sound, the efficiency of the ozone can be enhanced for the oxidation of the low biodegradable compounds	[59]

Table 2 ; shows the Ozone with hydrogen peroxide(H₂O₂) or UV radiation or Ultrasound or in combination with all:-

5.2 Hydrogen Peroxide and Ultraviolet Radiation (H₂O₂/UV):

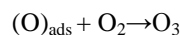
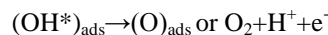
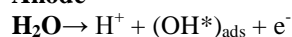
-H₂O₂ is used for the removal of low level contaminants in the waste water and it becomes more effective when it is used with other reagents or energy sources which are much able to produce hydroxile radicals. Usually used for the degradation compounds like naproxen, TNT and its aminodinitro- (ADNT) and diaminonitrotoluene (DANT) [60] etc. The reaction occurs during the process is given below [61].



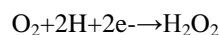
5.3 Electrochemical process: -This is the method that approaches the direct or indirect anodic reaction for the treatment of the waste water [62]. In the direct process , the absorption of the pollutants take place on the anodic surface for the degradation but it does not involve any oxidant. This method requires the further development for the industrial

acceptance. On the other hand, electrochemical fuel cells are used in indirect method where oxygen is sparged in the cathode chamber for the generation of the H₂O₂ by applying electric current directly [63]. The reaction of the whole process is as follows [64].

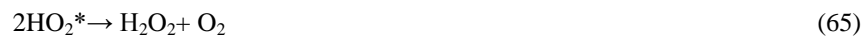
Anode



Cathode

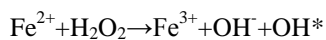


5.4 Fenton Process:- Fenton Process is the oldest and most widely used method in AOPs, discovered by British Chemical Engineer Henry John Horstman Fenton in 1894. In this method, the soluble mixture of the Iron (II) and hydrogen peroxide (H₂O₂), known as Fenton Reagents, is used for treatment of POPs, recalcitrant and other low biodegradable compounds [65]. There had always been controversy regarding Fenton's reaction

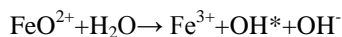
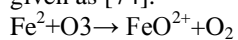


mechanism. Weiss and Haber in 1934 interpreted the classic Fenton reaction in which aqueous solution of hydrogen peroxide (H₂O₂) and ferrous ions (Fe²⁺ and Fe³⁺ in an acidic medium, decomposes the hydrogen peroxide into a hydroxyl ion and a hydroxyl radical. The oxidation of Fe²⁺ to Fe³⁺ and classical Fenton reaction mechanisms is shown in the equations below [66]

5.5 Photo-Fenton: -Photo-Fenton process is one of the most efficient AOPs for degradation of the organic or inorganic contaminants. By using the certain organic acids to Fe³⁺, the efficiency of this method can be improved. There are several advantages of using this reaction that includes the production of the low toxic products, need small quantity of iron salts, using of the solar radiation is not costly and environmentally friendly. The typical reaction is given below.[73]



5.6 Catalytic Ozonation: -Ferrous (III) is used as catalyst in this process and general reaction is given as [74].

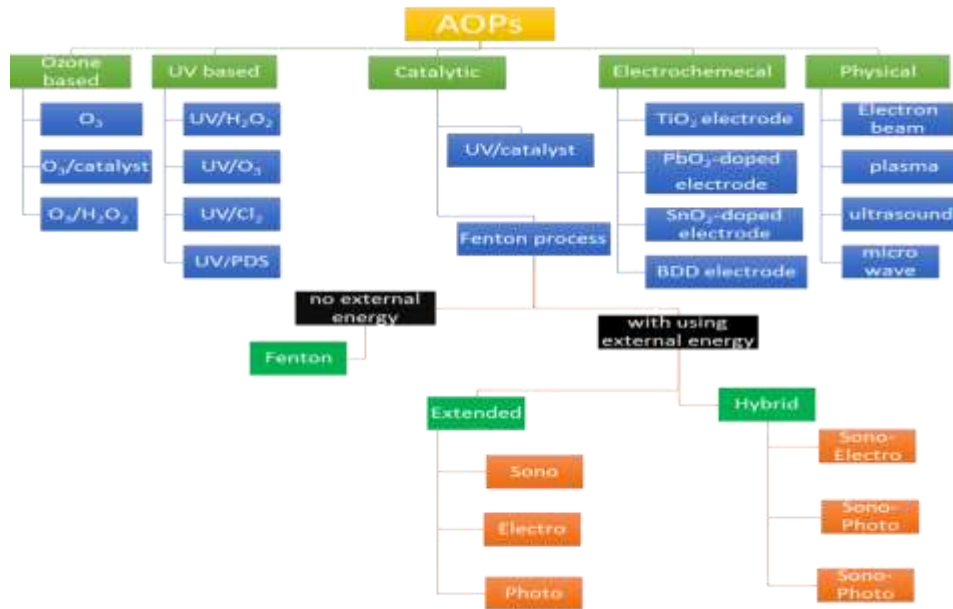


The transfer of the matter is taken place in 3 phases that's why it is one the limitation of using this method.

CONCLUSION

The use of AOPs to treat high-strength wastewater is a viable option. In general, we can conclude that each AOP has some advantages and disadvantages that can be best utilised based on the appropriate treatment scheme selection. Several AOPs in combination would be appropriate for removing more than 85 percent of the chemical oxygen demand (COD) and boosting biodegradability. The treatment option is determined by the wastewater discharge requirements. AOPs can be used as a treatment before or after treatment. Conventional biological treatments such as SBR, UASB, and ASP can easily treat the effluent of AOPs. In summary, a single treatment plan would be insufficient for high-concentration wastewater containing stubborn chemicals. They require a correct

combination of AOPs and other traditional approaches.



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