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Chapter

Advanced Oxidation Processes: A Powerful Treatment Option for the Removal of Recalcitrant Organic Compounds

Damodhar Ghime and Prabir Ghosh

Abstract

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. AOP can combine with ozone (O_3) , catalyst, or ultraviolet (UV)irradiation to offer a powerful treatment of wastewater. Future research should be focused on enhancing the properties of heterogeneous catalysts in AOPs. This chapter reports general review of different AOPs utilized for the removal of various phenolic compounds and textile dyes in wastewater. The chapter also aimed at an investigation of efficiency for different photochemical AOPs. The authors have carried out the experimental runs at a laboratory scale for the removal of malachite green oxalate (MGO) dye with photochemical AOPs. The influence of ferrous ions and oxidant dosage on percentage decolorization of MGO in wastewater has been reported. The discussion extends to the utilization of different modified photocatalysts for the photocatalysis process. The future challenges, such as the adoption of strategies for the integration of processes and the decrement in operational cost of AOPs, are discussed. The discussion covers the utilization of different heterogeneous catalysts, the reduction of input demands of chemicals and energy for the processes.

Keywords: advanced oxidation processes (AOPs), wastewater treatment, organic contaminants, heterogeneous catalysts, hydroxyl, sulphate radicals

1. Introduction

Advanced oxidation processes (AOPs) are the most attractive and favorable option for the effective removal of organic pollutants in wastewater. In recent years, many harmful chemicals in the released effluent from industries have been identified in the environment. These chemicals are hazardous to both human beings and aquatic biota. They can cause severe damages to human beings and the marine environment. So, there is a strong need to treat toxic pollutants with a proper treatment option. Thus, AOPs find its suitable applicability for the treatment of wastewater containing harmful chemicals. Advanced oxidation technologies are based on the in situ generations of strong oxidants, i.e. hydroxyl radicals and sulphate radicals, for the oxidation of organic pollutants [1]. Some of the AOPs use ozone and UV irradiation for better treatment efficiency. These AOP technologies have already been established and started at full scale for the treatment of drinking water and the facilities of water reuse. Also, several AOPs such as electrochemical treatment, use of electron beam, plasma, microwave and ultrasound related processes are continually being studied by the environmental researchers and scientists.

AOPs are very well known for bridging the gap between the treatability obtained by traditional physicochemical and biological processes, and from day to day more stringent regulations are set by the environmental laws. As extensive research is going on in AOPs from the last two decades, still these technologies are well matured, and there are again some fields worthy of research [2].

1.1 Different AOPs for wastewater remediation

Different AOPs such as photocatalysis, Fenton-like processes and ozonation were investigated for the treatment of an integrated solution of produced water that contains toluene, xylene, naphthalene, phenol, malonic and acetic acids. This integrated solution of produced water was referred as a seawater matrix [3]. The efficiency in terms of total organic carbon (TOC) was studied. Among different applied AOPs, photocatalysis was less efficient for the treatment of a seawater matrix. The removals of TOC lower than 20% were noticed in 4 h of treatment. The better results were obtained in the ozonation process that combined with oxidant hydrogen peroxide (H_2O_2). The combined effect of H_2O_2 and ozone leads to the complete removal of organics, consisting of a high-percentage removal of acetic acid that was not entirely oxidized with the rest of AOPs used. 74% TOC removal efficiency was observed in ozonation for the optimum conditions of 4 g/h ozone, 500 mg/L H_2O_2 and pH of 10 in 2 h of treatment time.

Several AOPs such as O₃, O₃/H₂O₂, UV, UV/O₃, UV/H₂O₂, O₃/UV/H₂O₂, Fe²⁺/H₂O₂ and photocatalysis processes were investigated for the oxidation of phenol in aqueous medium [4]. The comparison of these different AOPs was undertaken considering some of the experimental parameters. Among all, the Fenton process showed the fastest removal rate for phenol in wastewater. The lower costs were observed for ozonation. Single ozonation provides the best results for phenol degradation in ozone combinations.

The viability of the ozonation and ozone combined with UV photolysis processes was investigated for the degradation of direct blue 86 dye in wastewater [5]. The batch experiments were conducted for studying the effect of solution pH, initial dye concentration and reaction time. The obtained results show that the pH and initial dye concentration controls the efficiency of the process. More than 98% decolorization efficiency was obtained at pH 11, 100 mg/L of initial dye concentration in 35 min of ozone treatment.

All AOPs are characterized by their potential of exploiting the higher reactivity of hydroxyl radicals while driving in oxidation processes. The different AOPs with their essential features are discussed and presented for the wastewater remediation [6]. The number of investigations was carried out with the Fenton process for the abatement of organic pollutants in wastewater. Fenton's reagent can destroy the toxic contaminants in the wastewater like phenols and pesticides. The use of Fenton's reagent for the oxidation purpose is an attractive option for the wastewater treatment due to the iron abundances, and hydrogen peroxide is more comfortable to handle and environmentally safe. Sometimes, UV irradiation is also used with the Fenton process to increase the process efficiency. This process can overcome the shortcomings of the Fenton process. Photolysis allows the regeneration of

ferrous ions (Fe²⁺) from ferric ions (Fe³⁺) that produce sludge in the Fenton process. Besides, the titanium dioxide (TiO₂) in anatase form finds its application in photocatalysis at a greater level due to its higher stability, low cost and better performance for the degradation of organics.

1.2 Organic pollutants in industrial wastewater

The contamination of water resources continuously exists due to the release of organic pollutants from industrial discharges. The presence of organic pollutants affects the freshwater quality and causes severe effects on aquatic biota. The recent improvement in very sensitive analytical techniques is able to measure the concentration of organic pollutants, even in μ g/L concentrations. The organic content generally measures in terms of total organic carbon, chemical oxygen demand (COD), and biological oxygen demand (BOD) [7]. In most of the cases, the dyes and phenolic compounds were taken as model pollutants as per the published literature in recent years. Among different organic pollutants, the phenolic compounds have deserved more attention owing to their high toxicity and frequency of industrial wastewaters [8]. The presence of phenolic compounds in wastewater gives unpleasant smell and taste even at deficient concentrations.

Agro-industrial wastewaters are characterized by the presence of a higher amount of organic and inorganic pollutants of environmental concern [9]. Some parameters, such as high pollutant dose and production of more volume, make the treatment of agro-industrial waters an ecological challenge. For the effective treatment of industrial effluents, the wastewater treatment should consist of degradation as well as the mineralization of water pollutants. During oxidation, the conversion of identified pollutants in wastewater to its more stable oxidation state is necessary.

1.3 Aim and objectives of the chapter

This chapter studies the general review of different AOPs employed for the oxidation of organic pollutants in wastewater. The chapter also aimed at an investigation of the efficiency of different photochemical AOPs. Experimental runs using a UV photochemical reactor were performed at a laboratory scale for the decoloration of malachite green oxalate (MGO) dye using photochemical AOPs. The influence of ferrous ions and oxidant dosage on percentage decolorization of MGO in wastewater has been reported. The discussion was also extending to the utilization of different modified photocatalysts for the photocatalysis process. Some of the future challenges, such as the adoption of strategies for the integration of processes and the decrement in operational cost of AOPs, are discussed.

2. Different photochemical AOPs

Photochemical AOPs are frequently used as an efficient barrier for the oxidation of organic pollutants in wastewater. The combined effect of UV irradiation and hydrogen peroxide (H_2O_2) reaction confirms the removal of a broader range of compounds. Common photochemical AOPs used are UV alone, UV/ H_2O_2 , UV/ Fe^{2+} , UV/ H_2O_2/Fe^{2+} , UV/ O_3 , UV/ $S_2O_8^{2-}$, UV/ TiO_2 , UV/chlorine and UV with some other photocatalysts [10]. Therefore, UV AOPs can show better removal rates with both the radicals, i.e. hydroxyl and sulphate radicals.

2.1 Hydroxyl radical-based photochemical AOPs

The removal of targeted micro-pollutants, including naproxen, carbamazepine, diclofenac, gemfibrozil, ibuprofen, caffeine and mecoprop, were reported with photolysis and UV/H₂O₂ processes [11]. The medium-pressure UV lamp was used for the UV irradiation. The obtained results indicate that UV photolysis alone is not useful for the removal of micro-pollutants, excluding diclofenac and mecoprop. So, there was a need to treat the rest of the micro-pollutants with the UV/H₂O₂ process. It was found that the overall rates of oxidation for all selected organic pollutants were enhanced in the presence of H₂O₂, especially in the case of caffeine and carbamazepine at an oxidant dose of 25 mg/L on comparing with direct UV photocatalysis. Accordingly, the addition of H₂O₂ is critical in the photolysis process for better removal efficiency as it can promote the degradation of micro-pollutants in wastewater. The radical species are generated through the transfer of electrons by transition metal oxidation of oxidants or by the photolysis of H₂O₂ or thermolysis of H₂O₂ and in heterogeneous TiO₂ photocatalysis [12].

Recently, UV photolysis of hydrogen peroxides has earned considerable significance as they are comparatively a benign process concerning the environmental and economic considerations. The activation of symmetrical peroxides, mainly H_2O_2 with UV irradiation, generates the hydroxyl radical species (°OH) through the homolytic cleavage of the peroxide bond. These hydroxyl radicals are responsible for the oxidation of water pollutants.

2.2 Sulphate radical-based photochemical AOPs

Sulphate radical-based photochemical AOPs have drawn considerable attention because of the typical advantages of sulphate radicals such as higher redox potential (2.5–3.1 V), stability, selectivity and its use on the broader pH ranges. These radicals could be generated from different persulphates under the application of light, heat and transition metal and sometimes in an alkaline atmosphere or during electrochemical treatment [13]. The combined effect of UV irradiation with persulphate confirms the destruction of a broader range of water pollutants in wastewater.

The UV-activated persulphate advanced oxidation process was investigated for the degradation of sulfamethoxypyridazine (SMP) [14]. The sulphate radical-based photochemical AOP can significantly remediate SMP from aqueous solutions. In a comparative study of kinetics, the second-order reaction rate constant for SMP with sulphate radicals was obtained to 2.73×10^{10} M⁻¹ s⁻¹, whereas the use of hydroxyl radicals was 2.22 × 10^{10} M⁻¹ s⁻¹. Therefore, the faster reaction kinetics was in the case of sulphate radical-based photochemical AOPs.

Moreover, the uses of different heterogeneous catalysts such as cobalt, silver, copper and ruthenium-based catalysts have been studied to efficiently activate peroxymonosulphate (PMS) and peroxydisulphate (PDS) for the removal of water contaminants. Comparing to other activation methods, the activation of persulphates with heterogeneous catalysts has its own merits, such as the requirement of lesser energy, ease of scale-up and higher reactivity. Nevertheless, the use of cobalt- or copper-based catalysts might create some health problems because of the leaching of harmful metal into the aqueous phase, which can stop their practical applications. Again, the use of ruthenium-based catalysts for persulphate activation can be more costly. Thus, Mn-based catalysts are the best option to use for the activation of persulphate. Mn-based materials show some crucial advantages like its higher natural abundance, lower level of toxicity and environmental friendliness.

So, these materials have paid more attention to further improvement in Mn-based catalysts for the activation of persulphates [15].

3. Decolorization of malachite green oxalate dye through photochemical AOPs

The cylindrical type of photochemical reactor was used for the decolorization of malachite green oxalate dye in wastewater. The technical details for the reactor were that it was having the effective volume of 1.47 L and feeding tank volume of 5 L. The reactor was also built with an inside reflecting surface and four tubes of UV source (low-pressure mercury-vapor lamp of intensity 8 W each) surrounded with glass/quartz tube which was placed centrally in reactor [16]. The feeding tank was made up of SS304 material. The wastewater was circulated into the system with the help of a magnetic pump that was composed of polypropylene material. The maximum working temperature for the photochemical reactor was 80°C. The unknown concentration of MGO dye in wastewater after the treatment was measured with a UV–vis spectrophotometer. Absorbance values were detected at 618 nm of maximum wavelength for MGO dye.

3.1 Comparative assessment of different photochemical AOPs for MGO dye

The comparative assessment of various photochemical AOPs, including UV alone, UV/H₂O₂, UV/TiO₂, UV/H₂O₂/TiO₂ and UV/Fe²⁺/H₂O₂, were investigated for the decolorization of MGO dye. The reaction conditions used were initial dye and H₂O₂ concentrations of 100 mg/L and 12 mM, pH 3, ferrous ion concentration of 60 mg/L and TiO₂ dose to 0.6 g/L with irradiation of 32 W for 60 min of treatment time. A higher rate of decolorization was found in the case of UV/H₂O₂/TiO₂ and UV/Fe²⁺/H₂O₂ processes. It was about 97 and 98% for UV/Fe²⁺/H₂O₂ and UV/H₂O₂/ TiO₂ processes, respectively. This was happened due to the higher rate of generation for hydroxyl radicals in both the processes. The use of photocatalyst (i.e. TiO₂) enhances the production of hydroxyl radicals in the process, leading to the higher percentage removal of MGO dye. UV photolysis alone was not able to decolorize the MGO completely (77% for a treatment time of 60 min). UV/H₂O₂ and UV/TiO₂ processes almost achieved 95% decolorization efficiency. In short, the decolorization of MGO dye was reduced in the order of [16]:

$$UV/Fe^{2+}/H_2O_2 > UV/H_2O_2/TiO_2 > UV/TiO_2 > UV/H_2O_2 > UV \text{ alone}$$
(1)

3.2 Influence of ferrous ions on MGO dye decolorization in the photo-Fenton process

The influence of ferrous ions on MGO dye decolorization was studied by taking the concentrations of Fe^{2+} in the range of 20–80 mg/L. **Figure 1** shows the effect of varying ferrous ion concentration on MGO dye decolorization in the photo-Fenton process. It was seen that the rate of decolorization was enhanced as the ferrous ion concentrations increased up to 20–60 mg/L. Further increase in ferrous ions to 80 mg/L decreased the rate of decolorization. This might have happened due to the rise in the generation of hydroxyl radicals for a higher concentration of Fe^{2+} ions. It can show some scavenging effects of hydroxyl radicals. More concentration of ferrous ions in solution brings about the brown turbidity of ferric hydroxides, and this can cause the recombination of hydroxyl radicals in the aqueous medium [16].

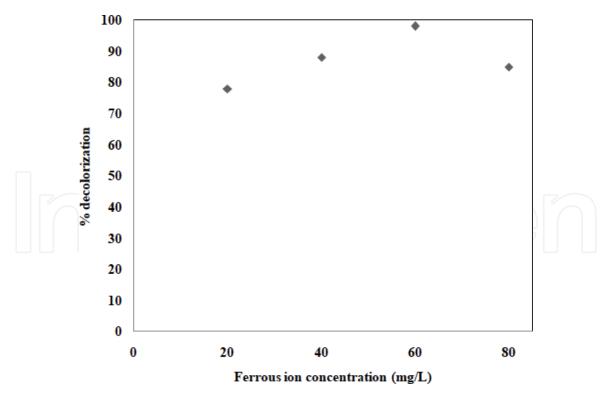


Figure 1.

Influence of ferrous ions (Fe^{2+}) on MGO dye decolorization with the photo-Fenton process by using dye concentration of 100 mg/L, 12 mM of H_2O_2 and pH 3 with irradiation of 32 W for 60 min of treatment time.

3.3 Effect of H₂O₂ concentrations in UV/H₂O₂, photo-Fenton and TiO₂ photocatalysis processes

The concentration of oxidant in the photochemical AOPs (UV/H₂O₂, photo-Fenton and TiO₂ photocatalysis) was varied in the range of 6–36 mM. About 88.61% of decolorization was observed for a dose of 6 mM in the photo-Fenton process [16]. This shows the existence of the generation of hydroxyl radicals into the system. Increment in percentage decolorization was observed with an increase in oxidant concentration up to 12 mM. Beyond 12 mM, it was found to decrease. **Figure 2** shows the influence of H₂O₂ concentration in UV/H₂O₂, photo-Fenton and TiO₂ photocatalysis processes at an oxidant concentration of 12 mM. The decrement in % decolorization beyond 12 mM of oxidant was due to the scavenging action of using extra H₂O₂ or possible recombination of hydroxyl radicals. Sometimes, the excessive amount of H₂O₂ acts as a hole scavenger, which can produce per-hydroxyl radicals. The per-hydroxyl radicals are less reactive than hydroxyl radicals. Thus, it is always advisable to use an optimum dose of oxidant.

3.4 Pseudo-first-order reaction kinetics for hydroxyl and sulphate radical-based AOPs

The pseudo-first-order reaction kinetics was reported for the decolorization study of MGO dye in wastewater. Both processes, hydroxyl and sulphate radical-based AOPs, were employed to calculate the reaction rates and compared to study their efficiencies. The parameters of kinetic models for decolorization efficiency of MGO dye at 12 mM dose of H_2O_2 and 1.68 mM of persulphate concentrations using UV irradiation were studied with the application of linear regression analysis to $[\ln(C_0/C_t)$ versus t] data for pseudo-first-order model [16]. The obtained values of correlation coefficients for sulphate and hydroxyl radical-based AOPs were 0.9904 and 0.9261, respectively. The reaction rate constant values were 0.0276 min⁻¹ for

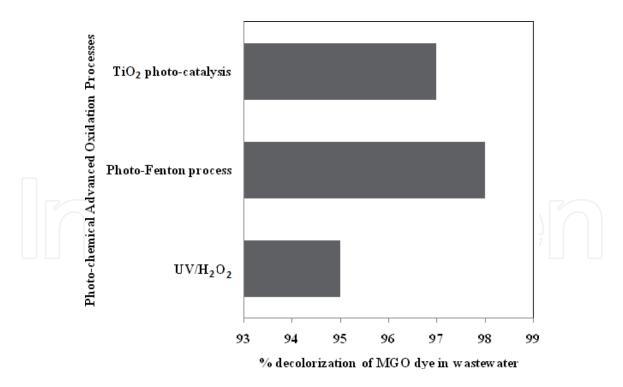


Figure 2.

Influence of H_2O_2 concentration in UV/ H_2O_2 , photo-Fenton and TiO₂ photocatalysis processes (experimental conditions: flow rate of 10 LPH, 12 mM of oxidant (H_2O_2), ferrous ion concentration of 60 mg/L, TiO₂ dosage of 0.6 g/L with irradiation of 32 W for 60 min of treatment time).

sulphate radical-based AOP and 0.0031 min⁻¹ for hydroxyl radical-based AOP. This means that the decolorization was faster in sulphate radical-based AOP.

4. Utilization of different modified photocatalysts in photocatalysis

The use of different photocatalysts in the photocatalysis processes is a useful alternative and attractive option in advanced researches. The photocatalyst should possess higher photochemical stability and can effectually use UV irradiation for wastewater treatment. The word 'photocatalyst' is made up of two parts: photo and catalyst. The word 'photo' means light, and a 'catalyst' means the process in which the oxidation rate is enhanced with catalyst.

Activation energy 'Ea' was found to decrease with an increase in the rate of reaction. In addition, the photocatalysts possess some properties such as higher surface area, better crystallinity, low cost, non-toxicity, ease handling and cost-effective applicability [17]. Recently, the application of semiconductors like titanium dioxide (TiO₂) shows great potential in photocatalysis processes. This is because of its low cost and environmentally friendly nature. TiO₂ is generally used as a standard reference for the comparison of the photochemical activity of other catalysts. The fine powdered form of TiO₂ is normally used in the slurry form. This leads to the higher volumetric production of reactive oxygen species (ROS) that can be proportional to the number of active surface sites [18].

The post-separation of TiO_2 after the treatment is an additional step while using this photocatalyst in slurry form. It is an important step for avoiding any loss of catalyst and can be reused for the photooxidation of new pollutants. The catalyst can be recovered through conventional sedimentation or via crossflow filtration or some types of membrane filtrations. The applicability of immobilized form of photocatalysts can increase the operational difficulty as the photon might not penetrate to every single surface active site for its activation. Silver orthophosphate (Ag₃PO₄) has also gained much attention for its use as photocatalysts as it shows higher photooxidative capabilities for O_2 evolution from the split-up of water and better removal efficiency for textile wastewater [19]. Approximately 90% of quantum efficiency can be achieved with Ag_3PO_4 at a wavelength of 420 nm in the oxidation of wastewater. During this phenomenon, $AgNO_3$ acts as a scavenger. This mentioned quantum efficiency was extremely higher than other photocatalysts, such as 20% in the case of N-doped TiO₂ and BiVO₄ photocatalysts. But the uncontrolled and unwanted photo-corrosion of Ag_3PO_4 is the main hurdle for its application in photocatalytic systems.

The photo-corrosion of Ag_3PO_4 happens due to its slighter solubility (0.02 g/L) in water and its characteristics of energy band structure. Thus, it's a big challenge for the researchers to improve and modify Ag_3PO_4 for their stability and to increase its activity without the help of any sacrificial reagents. Sometimes, the semiconductor, TiO₂, is doped to slow down the recombination of fast charge and to enable the visible light absorption with the generation of defect states in the bandgap. In the earlier case, the conduction band electrons or valence band holes are trapped in the defect sites, retarding their recombination and enhancing the interfacial charge transfer. In the latter case, the electronic transitions from the valence band to defect state or from defect state to conduction band are permitted under UV irradiation [20, 21].

The dopants are categorized into metal and nonmetal ions, and their selection is critical in considering their overall photocatalytic activity. Using noble metal ions (platinum, gold and silver) as dopants has gained less attention than the use of transition metals. Metallic doping serves the purpose of both a conduction band electron trapping and a cocatalyst in photocatalysis. Doping of TiO_2 is also possible with some nonmetallic elements such as carbon, sulfur, nitrogen and boron for studying their visible light photocatalysis.

The photocatalytic decomposition of water pollutants on nitrogen-doped TiO₂ under the illumination of visible light is essentially induced by the surface intermediates formed by oxidation of water or by the reductive activation of oxygen. It does not happen by the direct reactions with positively charged holes trapped at N₂-induced mid-gap level. Moreover, the use of the carbon-doped TiO_2 in photocatalysis is also controversial. Different methods have been proposed for the synthesis of C-doped TiO₂, such as carbon coating and carbon mounting. Two carbon dopants are there: the first one is anions that can substitute oxygen (O_2) in the lattice structure, and the second one is cations that can occupy an interstitial lattice site. The oxidation states that are linked with its bonding phases are +4 (carbonates having C-O bond) and -4 (carbides having Ti-C bond). These oxidation states are strongly dependent on the method of catalyst preparation and operating conditions used. Both states can be even copresent in the prepared photocatalysts. Sometimes, the TiO₂ is also doped with more than two elements for further enhancement in the photocatalytic activity under UV and visible light. This type of activity makes the doped TiO₂ more stable via compensation of charge. The metal/metal (Pt/Cr), nonmetal/nonmetal (N/S) and metal/nonmetal pair (Pt/N) combinations can be used further for doping [20]. Therefore, TiO₂ photocatalysts are doped with different elements, as discussed above, to overcome its demerits before use in oxidation, i.e. less absorbance with visible light and possible recombination of electron-hole pairs [22].

5. Conclusions

Advanced oxidation technologies represent a powerful option for the removal of organic pollutants in industrial wastewater. Different AOPs have been investigated,

and thus, it allows the selection of the most suitable process for the specific wastewater treatments. Different photochemical AOPs also have much potential for the reduction of organics in wastewater. The combination of UV and radicals (hydroxyl or sulphate) can effectively eliminate the organics with higher removal efficiency than direct UV photolysis or persulphate oxidation alone. Decolorization of malachite green oxalate was performed with different homogeneous and heterogeneous AOPs. The influence of ferrous ions and oxidant concentration and comparative assessment of different photochemical AOPs are reported. 12 mM of H_2O_2 and 60 mg/L of ferrous ion concentrations have been observed to be optimum in the photo-Fenton process. In the comparative assessment studies of photochemical AOPs, the percentage dye decolorization efficiency has been decreased in the order: $UV/Fe^{2+}/H_2O_2 > UV/H_2O_2/$ $TiO_2 > UV/TiO_2 > UV/H_2O_2 > UV$ alone. The experimental results show that the MGO dye removal efficiency from wastewater is higher in the case of sulphate radical-based AOP than hydroxyl radical-based AOP. Although the pure form of TiO_2 is a well-liked photocatalyst for many reasons, it suffers from lower efficiency for photochemical oxidation and becomes deficient in the visible light activity that obstructs its practical applications. Thus, the surface-modified TiO₂ photocatalysts have been continuously investigated to overcome the shortcomings of pure TiO₂.

6. Future perspectives

The conceptual proof of advanced oxidation processes has already been offered after years of research for wastewater treatment. However, something is indeed missing, the application of AOPs from laboratory scale to large scale (i.e. totally operational use). The main hindrance is the level of operating cost (money required for per unit mass of organic pollutant eliminated or unit volume of wastewater treated) allied with AOPs in comparison with other traditional wastewater treatment techniques. How can advanced oxidation treatment techniques become more striking cost-wise? Some useful thoughts that can be considered for its broader applicability are as follows:

- The utilization of renewable energy is essential in this case. AOPs driven by sun irradiation have an apparent head start not only for the treatment of industrial wastewater but also for the production of energy simultaneously. This is the more economical solution for the issue of water purification and its reuse.
- Every time, AOPs should not be our first choice for the treatment of wastewater. It must be imposed to become selective, i.e. they must have a definite treatment target. Otherwise, valuable and expensive elements such as oxidants, energy and different catalysts are wasted without cause.
- Advanced oxidation technologies have been intensely benefitted from the advances in the field of material science. Novel materials have been identified continuously with better characteristics. Consequently, the use of AOPs for environmental applications is a topic in engineering, science and other multiple disciplines. All the researchers and scientists working on the same field from various disciplines must join forces to solve the problem successfully. Moreover, the use of waste materials as a catalyst, such as red mud (waste from bauxite processing industries), can reduce the cost of AOP treatment. This phenomenon is waste valorization. It is a new, impressive and somewhat unexploited concept.

So, it is not possible to replace accessible wastewater treatment technologies with AOPs from an economic standpoint. But this problem can be solved by using the combination of advanced oxidation processes with the ancient treatment technologies. It is feasible and cost-effective, also comparing to a fully AOP application.

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