



Advanced Oxidation Processes for Wastewater Treatment: Facts and Future Trends

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Abstract

Pollution provoked by antibiotics and antibiotic-resistant bacteria must be placed as a worldwide health and ecological worry. Advanced oxidation processes (AOPs) are viewed as a greatly reasonable technique concerning water treatments for eliminating organic contaminants categorized as bio-recalcitrant and for the demobilization of pathogens, which are not touchable via traditional methods. This work discusses briefly the main recent facts and future trends in the field of applying AOPs in wastewater treatment. There is an urgent need for strengthening the studies on eliminating pollutants and microorganisms for wastewater reuse utilizing various AOPs. In-depth investigation about their reduction pathways employing AOPs is needed. Coupling AOPs with membrane processes should be kept in mind for better safety and pollutant removal.

Subject Areas

Environmental Sciences

Keywords

Wastewater Treatment, Advanced Oxidation Processes (AOPs), Contaminants of Emerging Concern (CECs), Antibiotic-Resistant Bacteria (ARB), Antibiotic Resistant Gene (ARG), Water Reuse

1. Introduction

Water is the heart of sustainable development; however, it is a restricted resource [1] [2]. Global warming gave rise to a terrifying deterioration of freshwa-

ter resources and their accessibility, therefore causing a large dare throughout the world. As an illustration, the number of people and regions touched by water shortage augmented nearly 20% in the summer of 2017 in Europe [3]. This tendency is awaited to persist, provoking worry across the European Union (EU) and neighboring nations and generating significant ecological and economic sequels.

Immoderate water extraction for agricultural irrigation and for industrial implementations remains one of the main threats to the aquatic ecosystems in the EU; however, the supply of potable water is a crucial condition for the expansion of economic sectors that depend on water [4]. Thus, hydric resources have to be handled more efficiently [5] [6] [7]. A handy option is to implement ecologically potential treatments to wastewater to recuperate it for the following objectives. Water reuse is an approach with a small number of negative ecological effects if contrasted to desalination or water transfers and presents economic and social advantages [8] [9] [10]. These days, even if water reuse could never be solved by itself water lack problems, at least it may assist to enhance the quality and quantity of the planet's water supplies [1] [11] [12] [13].

Selecting the best treatment for recuperating wastewater is a function of its ultimate targets. As a consequence, lower cost, robust, and more efficient techniques to clean and sanitize wastewater are needed without threatening human health or stressing nature by the remediation itself. In this direction, advanced oxidation processes (AOPs) are viewed as a greatly reasonable technique concerning water treatments for eliminating organic contaminants categorized as bio-recalcitrant [14] [15] and for the demobilization of pathogens not touchable via traditional methods [16] [17] [18]. AOPs were primarily proposed for potable water treatments in 1980. In the following years, they were largely tested as oxidizing treatments implemented to various wastewaters. AOPs are described as the oxidation processes linked to the production of reactive oxygen species (ROS) like hydroxyl radicals ($\bullet\text{OH}$) in adequate amount to generate recuperated effluents [16]. Hydroxyl radicals possess elevated redox potential (2.8 eV) and are non-selective [1]. They are able to attack organic matter via four routes: hydrogen abstraction, integration or addition of radicals, and electron transfer [1]. Their reaction with organic pollutants produces carbon radicals ($\bullet\text{R}$ or $\bullet\text{R-HO}$), which may be converted to organic peroxy radicals (ROO) with O_2 . All the radicals moreover react joined by the production of additional reactive species like super oxide ($\text{O}_2^{\bullet-}$) and hydrogen peroxide (H_2O_2), conducting to chemical demolition and, in some situations, the mineralization of water target contaminants [16]. If an AOP is utilized as tertiary treatment, $\bullet\text{OH}$ radicals are formed *in situ* because of their short lifetime via various techniques, comprising a mixture of oxidizing agents (O_3 and H_2O_2), ultrasound (US) or irradiation (UV), and catalysts [1]. The most usual catalyst remains titanium dioxide (TiO_2). If the TiO_2 particles are illuminated by UV light, they are excited and produce valence band holes where $\bullet\text{OH}$ are generated in contact with water [1]. Nevertheless, re-

cycling and recuperating such suspended TiO_2 particles become inconvenient and costly, making the employment of suspended systems not feasible. As an option, fresh setups have been presented employing such an immobilized catalyst [1]. Nevertheless, integrating hydrogen peroxide with Ultraviolet-C light (UVC) radiation leads to the most performant manner for generating $\bullet\text{OH}$ [1]. Further, employing iron [19] species as free catalysts for forming $\bullet\text{OH}$ in Fenton methods has been broadly investigated; however, its implementation in wastewater like tertiary treatments in actual circumstances stays limited because the optimal pH is 2.8 [20]. For such cause, researches have suggested three amended Fenton process: heterogeneous Fenton, photo-Fenton, and electro-Fenton [20]. With a view to produce the photo-Fenton response, the classical Fenton device is open to the UV light with the objective of enhancing the photo-reduction of dissolved ferric iron (Fe^{3+}) to ferrous iron species (Fe^{2+}) [20] [21] [22] [23]. In the electro-Fenton reaction, the two Fenton reagents can be formed with electrochemical techniques [20] [24]. Additional $\bullet\text{OH}$ and hydrogen peroxide production setup is the water sonolysis [1]. This technique remains less tried in wastewater as the initial operational costs are too elevated, and the treated volumes are very small as compared to other processes. Not only are $\bullet\text{OH}$ oxidant agents, but they are also sulfate radicals. Such species are highly reactive, and they possess a short life cycle. Consequently, the $\bullet\text{OH}$ may be produced from them at alkaline conditions.

As aforesaid, $\bullet\text{OH}$ radicals, formed throughout the implementation of AOPs on secondary effluents are apt to eliminate wastewater hazardous products and convert them into less poisonous contaminants, furnishing an option for remedying wastewater [16]. In addition to oxidation via $\bullet\text{OH}$, other concurrent reactions may occur throughout the treatments employing AOP, giving rise to the demolition of wanted compounds in wastewater [16]. The action of such non-radical oxidative pathways in eliminating pollutants may be unimportant or controlling following the reaction circumstances and the utilized AOP kind [1].

Lately, small levels of inorganic, organic, and mineral compounds in the aquatic medium have increased observably, mostly by human activities like immoderate and fast industrialization, urban encroachment, and enhanced agricultural procedures [8]. One practical solution for removing organic contaminants from wastewater is the usage of AOPs or their integrations with additional treatments [24] [25] [26] [27]. Such techniques have been frequently categorized as being greatly able to eliminate recalcitrant contaminants or being employed as a pre-treatment to convert pollutants into shorter-chain chemicals that can be treated after via conventional biological processes [16] [28] [29]. One AOP or integration must be suitably selected for dealing with a particular industrial or domestic wastewater, taking into account parameters like wastewater properties, technical applicability, and economical considerations [1] [30] [31].

It is as well proved that municipal wastewater treatment plants (MWWTPs) constitute a significant stock of ecological water pollution. The global load of

organic contaminants discharged by MWWTPs is a function of the number of inhabitants and the contamination received from local industries connected to the domestic sanitary system. A large set of poisonous remains (chemical or biological products) are produced every day by various sectors, which may be considered as toxic because of the likely negative influences that may induce (neurotoxicity, endocrine disruption, cancer) [1] [9]. Between the pollutants existing in WWTP effluents are personal care products, pharmaceuticals, pesticides, gasoline additives, flame retardants, drugs, plasticizers, and numerous chemicals frequently categorized as “contaminants of emerging concern (CECs)” [30] [31]. Such chemicals are detected at ng/L- μ g/L levels in MWWTP effluents; however, they stay not regulated [8]. WWTPs are seen as the principal route of entry of CECs to nature. Throughout the water treatment techniques, or once in nature, such products may as well be converted by a set of chemical, photochemical, or biological processes that conduct to the generation of transformation products (TPs), which may, at last, be more persistent or poisonous than the initial chemicals [6]. The inefficacious reduction of CECs via MWWTPs is a severe restriction for water reuse concerning the safety/sustainability of reuse usages like irrigation in agriculture or gardens and golf courses [8]. For that reason, the regulations do not authorize that the biologically treated wastewater may be directly reused since its content of health hazard micropollutants [32]. Such hazardous chemicals may be gathered in vegetables and soils with a huge effect on potable water resources and food security [33]. This status necessitates the expansion of substitutional treatment techniques to restrict the discharge of such products in nature [34]. Several publications established the efficacious reduction of micropollutants carried in real domestic wastewater [35]. Membrane bioreactor (MBR) technology merging conventional activated sludge (CAS) treatment with a membrane filtration setup was noted as an alternative to increasing the effluent quality reducing the membrane price [28]. Conversely, MBR is not a convenient technique for removing micropollutants because of membrane-fouling control [31] [36]. In this context, membrane aeration, permeability loss, and membrane replacements are parameters with elevated running costs [37] [38] [39] [40]. In order to substitute MBR technique, it may be preferred to employ solar AOPs as tertiary treatments for CECs elimination because the usage of solar energy diminishes investments costs [8] [41], conducting to wastewater treatments that are easy, robust, and inexpensive. Between the AOPs, the more investigated remains the heterogeneous photocatalysis employing TiO_2 as a catalyst. Nevertheless, it was proved that it is not performant as long treatment periods are required for the complete removal of microcontaminants [8]. One more AOP that generated low microcontaminant decomposition is the solar photo-Fenton running at neutral pH [20]. With a view to averting the iron precipitation in neutral pH circumstances, chelating agents are necessitated for maintaining the catalyst in solution. Usually, the complexing agents are existing in the wastewater; however, they are eliminated throughout secondary biological treatment or drinking water

treatment stages. In fact, there is a broad variety of agents that can be so helpful for maintaining the dissolved iron in the course of the solar photo-Fenton method and to stabilize the free radical formation [1].

Wastewater effluents carry not only toxic inorganic and organic chemicals but as well pathogens. Chlorination, UV-C radiation, and ozone are processes usually utilized for killing microorganisms in WWTPs. Disinfecting wastewater mostly concentrates on particular classes of microbe comprised in the water reuse regulations for various usages, which involve total and fecal coliforms, *Cryptosporidium* sp., or *Legionella* sp. [42] [43] [44] [45] [46]. Injecting chlorine products (chlorine gas, sodium hypochlorite, or calcium hypochlorite) stays the most cost-efficient remediation and has shown to be lethal against a large variety of wastewater pathogens [32] [47] [48] [49] [50]. Demobilizing bacteria is obtained through various cellular oxidation routes and inhibition of enzymatic activity together with the demolition of the cell membrane [51] [52] [53]. As related with the wastewater disinfection under UV-C radiation, it was proved that the photons were absorbed by the microorganisms' genetic material (DNA), therefore averting the cellular replication. Moreover, the aggregated UV-A energy per unit of treated water volume dose, in terms of kJ/L, is a fundamental factor to control the microorganism demobilization upon UV radiation in the function of treatment period when the system is photo-limited. However, the attention on ozone applications is growing because this compound possesses the potential to demobilize microbial cells and to diminish the charge of organic matters [1].

Actually, AOPs are established as substitutional techniques thanks to their capability to demolish a wide set of pollutants and to eliminate microbes from wastewater. Nevertheless, such processes are being studied with a view to reduce related running prices and guarantee the practicality. The leading AOPs examined for treating water remain TiO_2 , photocatalysis, UV/O_3 , $\text{UV}/\text{H}_2\text{O}_2$, Fenton, and photo-Fenton [20]. With a view to assess their chance, model microbes of fecal pollution are chosen because of their considerable immunity to most water disinfection techniques traditionally implemented. Among them, the most frequent are *Escherichia coli* [54], *Cryptosporidium* sp., or *Bacillus* sp. Such microbes are very hazardous to human beings. *E. coli* can hydrolyze conjugated estrogens by sulfatase and glucuronidase enzyme. Concerning protozoan microbes, *Cryptosporidium* is known for its resistance to chlorination methods, provoking intestinal infections in the human population. In addition, *Bacillus* sp. is a facultative anaerobic bacteria that may exist with a low quantity of dissolved oxygen. There are numerous *Bacillus* species present in nature—some of them possess an elevated wastewater purification capability to degrade highly concentrated organic matter in a short period, and they liberate a huge amount of enzymes that can degrade excess sludge. Nevertheless, additional species are dangerous if they appear in the treated water after secondary treatments, and tertiary processes like chlorination are not able to demobilize those [1].

Utilizing UV radiation integrated with H_2O_2 or TiO_2 augments the performance of the demobilization method. The work pathway of pathogen demobilization via UV varies from the pathogen demobilization via UV/ H_2O_2 . When both wastewater disinfection methods are contrasted, the treatment periods are shorter if the integrated technique is used [1]. One more sort of UV radiation is UV-C, which has been proved to be more performant than UVA/ TiO_2 or UV/sono-chemical treatments as it has a huge disinfectant potential, reaching a demobilization decrease of 6-log during 10 min of application period. Nevertheless, the photo-reactivation of microbes happened at 72 h following the end of the used technique [1]. A further performant, solar driven AOP is the photo-Fenton process. If the pH levels are augmented, the ferrous iron solubility diminishes, conducting to its precipitation as Fe^{3+} hydroxides. This reality may be a problem if the photo-Fenton process is chosen for disinfecting wastewater; consequently, the existence of the plurality of the controlled microorganisms diminishes or even dies at $pH < 3$. With a view to handle such blockage and assess the demobilization of microbes via the photo-Fenton process, many investigations were realized at $pH > 4$, where microorganisms are apt to endure [20]. To demobilize microbes by photocatalysis via TiO_2 , a crucial truth is the internal cellular destruction generated by actions of ROS, like $\bullet OH$, just as for photocatalysis by TiO_2 . Such impact on vital compounds of the cell starts with the photon absorbance via the plasmatic membrane of microbes provoking lethal physical demolition pursued by an oxidative attack by hydroxyl radicals on the cellular membranes, inducing oxidative stress and pores and forming loss of their permeability [43]. Such condition is a function of the quantity of $\bullet OH$ and the accessibility of iron during the photocatalytic application [48].

In spite of that, present microbial pathogen identification procedures in wastewater have noted the existence of a large collection of other microorganisms. Such microbes are viewed as “emerging pathogens” with an intrinsic alarm to the population because of their manifestation in recuperated waters and discharged waters to nature. An illustration of emerging pathogens is the antibiotic-resistant bacteria (ARB) [15]. The occurrence of antibiotics in effluents has augmented in the last year [14]. Especially, municipal and hospital wastewaters are between frequent ARB diffusion and anthropogenic sources into the aquatic medium. Discovering a performant and advanced technique to eliminate antibiotic compounds from treated water has been the main research focus for several years. Physical manners like nanofiltration (NF) and ultrafiltration (UF) following a CAS have proved that the reduction of antibiotics augmented by up to 30% [1]; however, such methods do not eliminate microcontaminants and they only transfer them from one point to another. For such cause, the AOPs may be utilized to purify the effluent carrying pharmaceutical products as an environmentally friendly technology through employing reusable catalysts and solar light. TiO_2 photocatalysis, Fenton, and photo-Fenton processes have surged as encouraging wastewater treatment technologies [20].

The occurrence of ARBs in the WWTP effluents released into aqueous ecosystems or reused for irrigation in agriculture shows that disinfection monotone usages do not efficiently dominate the diffusion of such pathogens into nature [15]. ARBs are commonly observed in wastewater effluents from hospitals, MWWTPs, and wastewater from cattle (known as “grey waters”) [14]. Actually, the principal classical disinfection techniques are chlorination, application of ozone, and UV-C. Concerning chlorine chemicals, ARB demobilization rates are not lower than those of total heterotrophic microbes, and surprisingly the part of several ARBs may be elevated following injecting chlorine products [1].

2. Conclusions

From this work, the following conclusions can be drawn:

- 1) That being the case, the external and the internal pathways of how the chlorination method affords to augment the levels of ARB and antibiotic resistant gene (ARG) in wastewater stay not defined; thus, more investigations in this domain are necessary [14] [15]. Regarding UV-C radiation influences, the accessible data show that this technique is not performant in killing ARB and eliminating ARG upon UV-doses about 30 mJ/cm², which are frequently employed. If the mJ/cm² is elevated, the microbe demobilization rate is elevated too, therefore attaining the total log removal. Following some researches, demobilization of 4 - 5 log of cell ARB needs low UV-doses from 10 - 20 mJ/cm² comparatively with those necessitated for removing ARG (UV-doses from 200 to 400 mJ/cm²). This shows that chlorination and UV techniques may not result in a significant effect over levels of ARB and ARG in wastewater, even if the mechanisms of removing such biological products are not evident. In addition, the impact of ozonation on ARB has been assessed in a few studies. They noted that such a technique is not practicable for killing ARB or eliminating ARG. None the less, different technologies are being examined with a view to ameliorate the performance of wastewater disinfection and to beat many disadvantages of the aforesaid traditional techniques, diminishing the related working costs as well. AOPs driven under solar light like Fenton and photo-Fenton processes have been estimated on wastewater for demobilizing ARB and ARG. The findings found in such researches established its lethal potential on natural ARB from MWWTPs secondary effluents. On the contrary, and following the kind of resistant gene, its performance is smaller if the ARG levels are analyzed, reaching total destruction in some controlled AG [1]. The existence and the expansion of resistant microorganisms in the effluents from MWWTPs disposed into reused effluents are some of the greatest dangers to humanity linked with the domestic usage of wastewater. These realities divulge the inefficiency of classical wastewater treatments and disinfection methods for dominating the diffusion of pathogens and microbial resistance into nature. For the time being, even if some investigations are being performed to monitor the expansion of ARB and ARG in nature, the biological processes to efficiently demobilize such microbes stay va-

gue.

2) The present investigations in wastewater treatments using AOPs are mostly concentrated at a pilot-scale in batch mode. Estimating in practical circumstances and in continuous flow mode is required for large-scale application [1].

3) Strengthening the studies on eliminating CECs and bacterial demobilization for wastewater reuse utilizing various AOPs [1].

4) Pollution provoked by antibiotics and ARBs/ARGs must be placed as a worldwide health and ecological worry. In-depth investigation about reduction pathways of antibiotics and ARBs/ARGs employing AOPs is needed [1].

5) Coupling AOPs with membrane processes should be kept in mind for better safety and pollutants removal.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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