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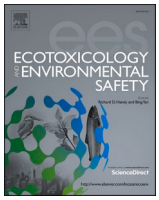

Removal of organic pollutants through hydroxyl radical-based advanced oxidation processes

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ABSTRACT

The use of Advance Oxidation Process (AOPs) has been extensively examined in order to eradicate organic pollutants. This review assesses the efficacy of photolysis, O_3 based (O_3/UV, O_3/H_2O_2, O_3/H_2O_2/UV, H_2O_2/UV, Fenton, Fenton-like, hetero-system) and sonochemical and electro-oxidative AOPs in this regard. The main purpose of this review and some suggestions for the advancement of AOPs is to facilitate the elimination of toxic organic pollutants. Initially proposed for the purification of drinking water in 1980, AOPs have since been employed for various wastewater treatments. AOPs technologies are essentially a process intensification through the use of hybrid methods for wastewater treatment, which generate large amounts of hydroxyl (•OH) and sulfate (SO4·−) radicals, the ultimate oxidants for the remediation of organic pollutants. This review covers the use of AOPs and ozone or UV treatment in combination to create a powerful method of wastewater treatment. This novel approach has been demonstrated to be highly effective, with the acceleration of the oxidation process through Fenton reaction and photocatalytic oxidation technologies. It is clear that Advance Oxidation Process are a helpful for the degradation of organic toxic compounds. Additionally, other processes such as •OH and SO4·− radical-based oxidation may also arise during AOPs treatment and contribute to the reduction of target organic pollutants. This review summarizes the current development of AOPs treatment of wastewater organic pollutants.

1. Introduction

In recent years, the use of advanced oxidation processes (AOPs) such as cavitation, photolytic oxidation, Fenton's, and Ozonation has become increasingly significant in wastewater treatment (Zhao and Yin, 2023; Yang et al., 2022). This method has been successfully employed in the elimination and degradation of organic pollutants (Elmolla and Chaudhuri, 2009; Giwa et al., 2021). The most advanced and modern developments in H_2O treatment involve the oxidation of intractable organic constituents, resulting in the creation of highly reactive moieties

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that can degrade the most difficult molecules into small, biologically degradable species or inorganic compounds such as CO_2 and water (Oturan, 2014; Yuan et al., 2023). Emerging contaminants such as pharmaceuticals, pesticides, hormones, UV filters, surfactants, and dyes have presented a major challenge for environmental researchers (Taheran et al., 2018; Gogoi et al., 2018). AOPs, or hybrid processes such as ultrasound-assisted Fenton, sono-photocatalytic, O_3/H_2O_2, have been developed to improve the efficiency and minimize the limitations and drawbacks of separate AOPs in treating pollutants (Adewuyi, 2001; Bae et al., 2023). Antibiotics, which are used to treat microbial infections and are utilized to treat human and animal diseases (Ghisi, 2014; Sun and Pittman, 2000), can cause hazardous pollution when misused or mishandled (Iqbal et al., 2021a; Focazio et al., 2008; Li et al., 2022).

It has been found that conventional biotic treatments are not only ineffective but also problematic in degrading persistent antibiotics (Imran et al., 2021; Wan and Wang, 2016; Liu et al., 2023). Recent studies have shown that it is not possible to degrade the majority of new compounds through artificial, mechanical, chemical, biotic, or physical treatments (Cincinelli et al., 2015; Iqbal et al., 2021b). Additionally, the occurrence of antibiotics and their metabolites in manure run-offs is a major cause of their entry into marine ecosystems (Manyi-Loh et al., 2018; Guo et al., 2022). Low concentrations of antibiotics in surface and drinking water pose potential threats to the environment and serious health risks for humans (Hussain et al., 2021; Kim et al., 2019). These drugs and their hazardous metabolites can lead to various problems, such as chronic toxicity, endocrine disruption, and soil toxicity (Hussain et al., 2021). Antibiotics have been detected in various expected environments, such as rivers (Khan et al., 2021a; Ye et al., 2019; Khan et al., 2021b), surface water (Khan et al., 2021b), drinking water (Shah et al., 2021; Xiao et al., 2023), topsoil (Shi et al., 2022). Long-term exposure to antibiotics can lead to the generation of antibiotic-resistant genes (ARGs) and bacteria (ARBs), which poses a threat not only to the environment but also to human health and the ecosystem as a whole (Ben et al., 2019; Peng et al., 2022). Various techniques, such as coagulation, membrane separation, sorption and biodegradation have been discussed to treat a range of drugs found in drinking and discarded water (Cerqueira et al., 2019; Zhuang and Wang, 2019a). However, these approaches are often not applicable due to their high operational cost and low removal efficacy. Nanotechnology, on the other hand, is being explored as a promising alternative for this purpose due to its applicability in many areas of life (Gaur et al., 2022).

Nanomaterials have recently gained a lot of attention and interest from the scientific community due to their extraordinary properties, such as size and shape, and their diverse applications (Zhuang and Wang, 2019b). Furthermore, nanomaterials can be easily delivered and bound to the intended target site due to their small size (Zhuang, 2019). Some novel and modified nanotechnologies that can be used to remove contaminants from wastewater include carbon nanomaterials (CNTs), graphene (GRA) and fullerene (C_{60}) (Ul-Islam et al., 2017; Dong et al., 2022), electrochemical and ionizing radiation (Ul-Islam et al., 2014), reverse osmosis (Peng et al., 2020) and adsorption on activated carbon. Advanced oxidation processes such as Fenton or Photo-Fenton system, UV light peroxidation, and photocatalysis through TiO_2 have been used for remediation of groundwater, drinking water, and wastewater (Oturan, 2014).

Recently, nanotechnology has been employed to improve, represent, create, and display nanomaterials at the nanometer scale for the same purpose (Zhuan, 2020). This application of nanomaterials has led to the development of efficient treatment systems, which are simpler and less expensive than conventional technologies (Zhuan, 2020; Knepper et al., 1999). Pesticides are often converted into persistent metabolites that remain in the environment for years. Their ability to enter the food chain makes them especially hazardous, as their bioaccumulation can be dangerous (Jia et al., 2022; Lazartigues et al., 2013). Assessing the effects of organic pollutants on the ecosystem is difficult, as their effects depend on both their mode of action, persistence time, and degradation byproducts (Lazartigues et al., 2013). It is difficult to accurately estimate the effects organic pollutants have on the ecosystem due to factors such as their mode of action, persistence time, and the byproducts created by their breakdown. In some cases, these by products may be even more toxic than the original pollutant and require a more time for degradation (Dong et al., 2022). For the purpose of eliminating detrimental organisms in the environment, pesticides are used to protect agricultural products. Only 10–15% of functional pesticides have a meaningful effect on the targeted pollutants, reducing their impact on the environment (Lazartigues et al., 2013). Research has demonstrated that most of these pollutants are not only hazardous but also have carcinogenic properties. This has led to a global water pollution crisis due to the presence of toxic and persistent organic contaminants. To combat this, researchers worldwide have been utilizing nanotechnology for environmental remediation (Sayan et al., 2013), as nanomaterials are highly effective for the removal of organic pollutants (Cahill et al., 2011; Jiang et al., 2023). Nanomaterials have proven their effectiveness in improving the detection of organic and inorganic pollutants at trace levels in both aquatic and terrestrial systems. Adsorption, photocatalysis, bioremediation, and nanoparticle reduction are all techniques used to control contamination and its effects on the environment. Nanomaterials are also utilized for the detection of pesticides, herbicides, and drugs, as well as for the degradation and transformation of pollutants into less toxic forms (Dolar et al., 2012; Herrera et al., 2023).

The selective capture and elimination of pollutants via oxidation or reduction reactions are made possible by the functional and novel characteristics of nanomaterials (Zhuang and Wang, 2020; Zhuang, 2019). A variety of AOPs are used to speed up the photo-degradation process, including hydrogen peroxide and/or ozone, metallic salts, and semi-conductors such as titanium dioxide (TiO_2) (Brame and Alvarez, 2011; Tang et al., 2023). Nanostructures of TiO_2 and zinc oxide (ZnO) are commonly used as photo-catalysts. The most effective AOPs for pesticide degradation are photocatalytic oxidation (e.g., TiO_2/UV), Photo-Fenton, and Fenton-like coordination (H_2O_2/UV/Fe^{3+}). In this review, different nanomaterials and techniques such as Fenton or Fenton-like reaction, catalytic O_3 process, POR (photocatalytic oxidation reaction), and EOIP (electrochemical oxidation, ionizing process techniques) will be discussed in regards to their ability to degrade various pharmaceuticals and organic pollutants such as herbicides and pesticides (Tang et al., 2023; Zhao, 2015).

1.1. Advanced oxidation process

Advance Oxidation Process (AOP) is commonly used to oxidize complex organic pollutants that originate from industrial wastewater. These contaminants are difficult to breakdown into smaller molecules through biological processes (Gangupomu and Ramirez, 2014; Manisankar, 2015). In chemical advance oxidation (CAOPs), ozone and hydrogen peroxide are used to breakdown organic toxins or at least break them down into harmless products. In certain degradation processes, the byproducts of the solution can demonstrate equivalent or increased toxicity when compared to the original compounds (Manisankar, 2015). In such cases, contaminants can be removed through a special type of oxidation known as Advanced Oxidation Processes (AOPs) (Zhao, 2015). The biodegradability and rate of removal of antibiotics and their inhibitory effect on microbes can be improved by the use of AOPs, which can effectively breakdown antibiotics into small molecules. These AOPs employ strong oxidation proxies such as Hydroxyl radical ($^{\bullet}OH$), ozone (O_3) and superoxide radical ($O^{2-\cdot}$) (G. Covinich et al., 2014; Kurt et al., 2017). Further, Fenton oxidation, photo-catalytic oxidation reaction, and electrochemical oxidation reaction are very useful and versatile methods of AOPs for generating oxidizing agents (Macwan and Chaturvedi, 2011; Srivastava, 2020).

1.2. Theory of advanced oxidation

The generation of Hydroxyl Radical (·OH) serves as a potent oxidizing agent for the breakdown of organic contaminants which cannot be oxidized with commonly used oxidants such as O_3, O_2 and Cl_2 (Munter, 2001). The hydroxyl radical is essential in the destruction of toxic molecules. Once created as a result of a chemical reaction, the hydroxyl radical starts a chain reaction leading to the complete degradation of organic pollutants (Munter, 2001). The process involves a series of steps, as outlined in Eqs. 1–3 and summarized in Table S1, which involve the production of OH radicals, a non-selective chemical oxidant (Munter, 2001).

$$R + HO^{\cdot} \rightarrow OH \tag{1}$$

$$R + OH^{\cdot} \rightarrow R + H_2O \tag{2}$$

$$R_n \quad HO^{\cdot} \rightarrow R_n^{-1} + OH^{-} \tag{3}$$

Most advanced oxidation processes do not allow for complete oxidation of the given compounds or portions of them. This process leads to the total conversion of organic carbon pollutants to CO_2 (Carey, 1992). As a result of the advanced oxidation process, changes in structure occur, which can lead to an increase in toxicity (Carey, 1992; Al-Asadi, 2018).

1.3. Advance oxidation classification

The AOP has been segmented into various categories. The most successful of these are those that involve an oxidizer, such as hydrogen peroxide, combined with a catalyst and ultraviolet (UV) light (Al-Asadi, 2018; Lei et al., 2023). Studies have revealed that the most effective AOPs are those that combine titanium dioxide (TiO_2) with UV light, Fenton's reaction and hydrogen peroxide with UV light, which produce the hydroxyl radical (•OH) as a byproduct (Oller and Sánchez-Pérez, 2011; Zhang et al., 2022). The AOPs are divided into two main categories: homogeneous and heterogeneous. The homogeneous AOPs (HOAOPs) and the heterogeneous AOPs (HTAOPs) are further subdivisions of these categories. HTAOPs use UV radiation to degrade organic contaminants, which absorb light within the given range of the spectrum. On the other hand, heterogeneous AOPs rely on catalysts to breakdown compounds (Oller and Sánchez-Pérez, 2011). The production of photoelectrons and holes lead to the oxidation and reduction process, respectively.

The hydroxyl radical is formed in H_2O solution when water molecules are oxidized by a catalyst, resulting in radioactivity yield that is dependent on the linear energy transfer (LET) and pH in the range of 6–8.5, as depicted in the following reaction (Wasiewicz and Getoff, 2006; Wang et al., 2023).

$$H_2O \rightarrow HO^{\bullet}(2.8), H^{\bullet}(0.6), e_{aq}^{-}(2.7), H_2(0.45), H_2O_2(0.72), H_{aq} + (3.2), HO_{aq}^{(0.5)^{-}}$$

The advance oxidation process had a direct impact on the types of chemicals and catalysts used in order to achieve the goal of oxidizing the target compounds. Fig. S1 demonstrates the various chemicals and techniques used in the degradation of organic pollutants (Gogate, 2021).

Without the metal ions, the radiation shows lower potential in the degradation of the organic compounds. The have disproportion to hydrogen peroxide and oxygen at pH < is of importance as shown in Eqs. 4–9 (Gogate, 2021; Kubesch et al., 2005).

$$H^{\cdot} + O_2 \rightarrow HO_2^{\bullet}, k = 2.0x \quad 10^{10} M^{-1}s^{-1} \tag{4}$$

$$e_{aq+}O_2 \rightarrow O_2^{\cdot -}, k = 1.9x \quad 10^{10} \quad M^{-1} \quad s^{-1} \tag{5}$$

$$H_2^{\cdot} \leftrightarrow H^{+} + O^{-} \quad pK = 4.8 \tag{6}$$

$$HO_2^{\bullet} + O_2^{\cdot -} \rightarrow H_2O_2 + O_2 (pH < 7), k = 9.7x \quad 10^7 M \tag{7}$$

$$HO_2^{\bullet} + HO_2 \rightarrow H_2O_2 + O_2, k = 8.3x10 M^{-1}s^{-1} \tag{8}$$

$$O_2^{\cdot -} + O_2^{\cdot -} \rightarrow H_2O_2 + O_2, k < 10 M^{-1}s^{-1} \tag{9}$$

During the advance chemical reaction the recombination of free radical, caused radical reaction (Wang et al., 2023; Basfar et al., 2007; Jay-Gerin, 1992). During this process the hydroxyl radical react with solvated electron e_{aq}^{-} hydrogen radical, and hydroxyl radical to create negative hydroxyl, water and hydrogen peroxide as shown in Eqs. 10–12, which caused to decrease in the effective radical concentration. The reactivity of reductive classes also occurs during this oxidation process, as shown in the Eqs. 10–15 (Jay-Gerin, 1992; Ashraf, 2009).

$$HO^{\cdot} + e_{aq}^{-} \rightarrow HO^{-}, k = 3.0x10 M^{-1}s^{-1} \tag{10}$$

$$HO^{\cdot} + H^{\cdot} \rightarrow H_2O, k = 7.0x10^9 10 M^{-1}s^{-1} \tag{11}$$

$$O^{\cdot} + HO^{\cdot} \rightarrow H_2O_2, k5.5x \quad 10^9 M^{-1}s^{-1} \tag{12}$$

$$e_{aq}^{-} + H^{\cdot} \rightarrow H_2 + HO^{-}, k = 2.5x10 M^{-1}s^{-1} \tag{13}$$

$$e_{aq}^{-} + e_{aq}^{-} \rightarrow HO^{-} + H_2, k = 5.4x10^9 M^{-1}s^{-1} \tag{14}$$

$$H^{\cdot} + H^{\cdot} \rightarrow H_2, k = 7.8x \quad 10^9 M^{-1}s^{-1} \tag{15}$$

Hydrogen peroxide reacted with solvated electron (e_{aq}) and •H, which caused the production of •OH radical (Ashraf, 2009; Raman and Daud, 2015). A solvated electron refers to an extra electron confined within a cavity created by surrounding solvent molecules (Lakhno, 2007). This species, which appears temporarily, can be discovered in a wide range of biological and chemical systems where the transfer of electrical charge occurs within a medium. Its significance is evident in various applications, including cancer radiation therapy and the elimination of hazardous waste materials (Sun and Pittman, 2000). The hydroxyl radical react with hydrogen peroxide created in the solution and produced $HO_2^{\bullet}$ class as represented in Eqs. 16–19. In the presence of strong basic environmental/condition, the hydroxyl radical react as weak acid as shown in Eqs. 16–19.

$$e_{aq}^{-} + H_2O_2 \rightarrow HO^{\cdot} + OH^{-}, k = 1.2x \quad 10^{10} \quad M^{-1}s^{-1} \tag{16}$$

$$H^{\cdot} + H_2O_2 \rightarrow OH^{\cdot} + H_2O, k = 9.0x \quad 10^7 \quad k = 1.2x \quad 10^{10} \quad M^{-1}s^{-1} \tag{17}$$

$$HO^{\cdot} + H_2O_2 \rightarrow H_2O + HO_2^{\cdot}, k = 2.7x \quad 10^7 \quad M^{-1}s^{-1} \tag{18}$$

$$HO^{\cdot} + H_2O_2 \rightarrow O^{-} + H_2O, k = 1.2x \quad 10^{10} \quad M^{-1}s^{-1} \tag{19}$$

1.4. H_2O_2 and UV-radiation

During advance oxidation process, •OH radical are generated through photolysis of hydrogen peroxide and caused chain reaction which is corresponding to the process (Raman and Daud, 2015; Keskin and Özdemir, 2023). The hydroxyl radical is generated, when H_2O_2 absorbs applied UV light, as shown in Eq. 20.

$$H_2O_2 + hV \rightarrow 2HO^{\cdot} \tag{20}$$

H_2O_2 is the primary source of OH radical generation, and these OH radicals play a significant role in the degradation mechanism. Deficiency of hydroxyl radicals •OH occurs due to insufficient H_2O_2 dosage resulting in declining the degradation efficacy. In contrast, for pollutants degradation excessive concentration of H_2O_2 is also not suitable (Keskin and Özdemir, 2023; Garoma and Gurol, 2004). According to the subsequent given Eq. 21, the mandatory theoretical H_2O_2 dose value could be designed (Garoma and Gurol, 2004).

$$C_aH_bN_cO_d + (2a + \frac{1}{2b} + \frac{5}{2c-d})H_2O_2 \rightarrow aCO_2 + (2a + b + 2c - d)H_2O + cHNO_2 \quad (21)$$

Theoretically, one mole of $C_aH_bN_cO_d$ obliged ($\mathbf{2a + 1/2\ b + 5/2\ c - d}$) moles of H_2O_2. As compared to the designed value, actual added amount of H_2O_2 must be higher according to the above chemical equation, and it can be scrutinized by way of the earliest experimentations. The literature showed that rate of photolysis of H_2O is dependent and increases with the increase in alkaline environment (Raman and Daud, 2014)·H_2O_2 /UV combined system have the ability to degrade completely the organic pollutants to water and CO_2. The procedure, is not usually necessary as the oxidative products is not a problem, because their degradation is easy (Raman and Daud, 2014). Addition of H_2O_2 to the reaction, either as multiple steps or single dose. To calculate the optimal dose of H_2O_2 need pilot-scale and bench testing (Raman and Daud, 2014). The main advantage of this process are that hydrogen peroxide is soluble and may added at high amount and the fact that hydrogen peroxide /UV are able to produced higher number of $^{\bullet}$OH radicals, compared to O_3/UV under same environmental condition (Raman and Daud, 2014). This process is costly, expensive because of the additional cost of necessary devices and requirement of the energy consumed and the detail due to the remaining of H_2O_2 in the treated sewage leads biological re-growth in the dissemination (Leverenz and Tchobanoglous, 2007). The photolysis process directly encompasses the interaction of light with molecules, and caused the dissociation of the molecules into fragments why fragmentation pathway as shown Eqs. 22–23.

$$R + hV \rightarrow Intermediate \quad (22)$$

$$Intermediates + hV \rightarrow CO_2 + H_2O + R^- \quad (23)$$

This method is less operative when compared with other protocol in which UV radiation is combined with O_3 or H_2O_2 (Raman and Daud, 2015).

1.5. Catalytic ozonation (CO)

Catalytic Ozonation is an ecofriendly process in wastewater treatment (Raman and Daud, 2015; Xu et al., 2023). O_3 and organic molecules reacted either through electrophilic reaction or through radical chain reactions. The literature showed that catalytic behavior of O_3 is much slower as compared of hydroxyl radical, which showed, that the treatment of hydroxyl radical with organic pollutants is the main mechanistic pathway in UV/O_3 environment (Raman and Daud, 2015; Garoma and Gurol, 2004; Wang et al., 2019b). During photolysis ($\lambda < 300$ nm) decomposition of ozone occur into oxygen molecules and atom as shown in equation (Wang et al., 2019b; Javed et al., 2023). Nascent oxygen [O] atom is very energetic, reactive and practically reacts with all possible available substrates, inclusion H_2O. It is confirmed that, the nascent oxygen reacts predominately through insertion into the C-H and O-H bond (Wang et al., 2019b; Kusic and Bozic, 2006). The additional energy of hydrogen peroxide, so caused the fragments of O-O as represented in Eqs. 24–30 (Garoma and Gurol, 2004; Kusic and Bozic, 2006).

$$O_3 + hv \rightarrow O_2 + O(1_D), \varnothing_O \approx 0.9 \quad (24)$$

$$O(1_D) + H_2O \rightarrow H_2O_2 \quad (hot), k = 1.8x \quad 10^{10} M^{-1}s^{-1} \quad (25)$$

$$H_2O_2 \quad (hot) \rightarrow 2HO^{\cdot} \quad (26)$$

The O_3 is degraded in short chain reaction, which initiated through •OH radicals (Garoma and Gurol, 2004).

$$O_3 + HO^{\cdot} \rightarrow HO_2^{\cdot} + O_2, k = 1.1x \quad 10^8 \quad M^{-1}s^{-1} \quad (27)$$

$$HO_2^{\cdot} \leftrightarrow H^+ + O_2^{\cdot -}, pK = 4.8 \quad (28)$$

$$O_3 + O_2^{\cdot -} \rightarrow O_3 + O_2, k = 1.6x \quad 10^9 \quad M^{-1}s^{-1} \quad (29)$$

$$O_3 + HO_2^{\cdot} \rightarrow HO^{\cdot} + 2O_2, k = 1.2x \quad 10^6 M^{-1}s^{-1} \quad (30)$$

An assortment of refractory biological chemicals oxidize by ozone by means of 2.07 V oxidation potential and organic pollutants ruined directly by ozone particles (Garoma and Gurol, 2004). Moreover, ozone forms hydroxyl radicals $^{\bullet}$OH with help of catalyst when it undergoes reaction with water and hydroxyl radical has stronger oxidation capability as depicted in Eqs. 31–35.

$$O_3 + H_2O \rightarrow 2OH^{\cdot} + O_2 \quad (31)$$

$$O_3 + HO^- \rightarrow O_2^{\cdot -} + HO_2 \quad (32)$$

$$O_3 + HO_2 \rightarrow 2O_2 + HO^{\cdot} \quad (33)$$

$$O_3 + HO^{\cdot} \rightarrow O_2 + HO_2^{\cdot} \quad (34)$$

$$2HO_2^{\cdot} \rightarrow O_2 + H_2O_2 \quad (35)$$

The rate of removal /deprivation of organic contaminants increases through the of H_2O_2 with UV/O_3 combination because the generation of hydroxyl radical production increases (Kusic and Bozic, 2006). In both homogeneous catalytic (HOC) and heterogeneous catalytic (HTC) Ozonation methods, degradation efficiency of organic toxic waste can be enhanced. Ozone molecule excites by using these catalysts and generate hydroxyl radicals $^{\bullet}$OH which improvise proficiency of degradation (Yang et al., 2023). Solid catalysts like metal oxides, activated carbon, permeable materials and their complex materials are supplementary into reaction elucidation in heterogeneous catalytic Ozonation methods (Kusic and Bozic, 2006). The mechanistic routs for the creation of hydroxyl radical for O_3/Mn^{II} are proposed in reaction (Kusic and Bozic, 2006; Bautista et al., 2008). In the reaction O_3/Fe^{2+} & O_3/Fe^{3+}, the decomposition of O_3 occur and generation of radicals ions are observed as shown in reaction **36–41** (Bautista et al., 2008; Ji et al., 2023).

$$Mn^{2+} + O_3 + H^+ \rightarrow Mn^{3+} + HO^{\cdot} + O_2 \quad (36)$$

$$Fe^{2+} + O_3 \rightarrow Fe^{3+} + O_3^{\cdot -} \quad (37)$$

$$O_3^{\cdot -} + H^+ \rightarrow O_2 + HO^{\cdot} \quad (38)$$

$$Fe^{2+} + O_3 \rightarrow FeO^{2+} + O_2, k = 8.2x \quad 10^5 \quad M^{-1}s^{-1} \quad (39)$$

$$FeO^{2+} + H_2O \rightarrow HO^{\cdot} + OH, k = 1.3x \quad 10^{-2} M^{-1}s^{-1} \quad (40)$$

$$Fe^{3+} \quad O_3 + H_2O \rightarrow FeO^{2+} + H^+ + HO^{\cdot} + O_2 \quad (41)$$

1.6. Fenton & Fenton-like reaction

Reaction of ferrous salt with hydrogen peroxide is named as Fenton reagent (Fenton, 1894) used mostly in the various processes of wastewater treatment plants and when it is combined with wastewater (Fenton reagent (Fe^{2+} and H_2O_2) it undergoes a fast reaction and generate $^{\bullet}$OH which can oxidize and degrade the water contaminants as described in Eqs. 42–45**.** Fenton reaction defined the power of H_2O_2 to the degradation of several of various organic compounds in which hydroxyl radical are produced by the reaction of H_2O_2 and Fe^{II} as the catalyst (Bautista et al., 2008; Brillas and Oturan, 2009; Meyerstein, 2021).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO^{\cdot} + HO^- \quad (42)$$

$$HO^{\cdot} + H_2O_2 \rightarrow HO_2^{\cdot} + H_2O \quad (43)$$

$$2OH^{\cdot} \rightarrow H_2O_2 \quad (44)$$

$$H_2O \rightarrow HO^{\cdot} + H^{\cdot} \quad (45)$$

The homogeneous catalytic Fenton reaction, originated through the production of •OH radical in acidic solution. During homogeneous process, number of cyclic reaction are generated that utilized the Ferric ion as catalyst and course decomposition of H_2O_2 (Brillas and Oturan, 2009). The Ferric ion (Fe^{2+}) is generated in the first type of Fenton type reaction called propagation, which caused reduction Fe^{3+} with hydrogen peroxide (H_2O_2) from, $HO_2^{\bullet}$, alkyl radical $R^{\bullet}$, or $O_2^{\bullet}$ from reactions. The reaction related with two step revolution, in which Fe (III)-complex generated in equilibrium (Brillas and Oturan, 2009). The organic contaminants degradation through hydroxyls radical, by abstracting H from CH or NH or •OH bond and the addition of -C═C- or aromatic rings mainly depend upon the ionization potential of the organic contaminants (De Laat et al., 1999). The organic radical/-intermediate generated during the process may reacted with Fe^{3+} and hydrogen peroxide, generated R^{+} and ROH shown in equation, which can be oxidized further (De Laat et al., 1999). Further in the presence of alkyl radical ($R^{\bullet}$) maybe reacted with O_2 to produced $HO_2^{\bullet}$, peroxyl radical ($ROO^{\bullet}$) or Oxyl radical, which eventual converted into CO_2 and H_2O and moiety of organic acid (Brillas and Oturan, 2009). The degradation process is shown in Eqs. 46–62.

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO^{\cdot} + HO^{-}, k = 63M^{-1}s^{-1} \quad (46)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{3+} + HO_2^{\cdot} + H^{+} \quad k = 0.002 - 001M^{-1}s^{-1} \quad (47)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe(OOH)^{2+} + H^{+} \quad k = 3.1x \quad 10^{-3}M^{-1}s^{-1} \quad (48)$$

$$Fe(OOH)^{2+} \rightarrow Fe^{2+} + HO_2^{\cdot} + H^{+}, k = 2.7x \quad 10^{-3}M^{-1}s^{-1} \quad (49)$$

$$Fe^{3+} + HO_2^{\cdot} \rightarrow Fe^{2+} + O_2 + H^{+}, k = 2.0x \quad 10^{3}M^{-1}s^{-1} \quad (50)$$

$$Fe^{3+} + R^{\cdot} \rightarrow Fe^{2+} + R^{+} \quad (51)$$

$$Fe^{3+} + O_2^{-} \rightarrow Fe^{2+} + O_2 + H^{+} \quad k = 2.0x \quad 10^{3}M^{-1}s^{-1} \quad (50)$$

$$HO^{\cdot} + RH \rightarrow H_2O + R, k = 10^{7} - 10^{9}M^{-1}s^{-1} \quad (52)$$

$$R^{\cdot} + Fe^{3+} \rightarrow R^{+} + Fe^{2+} \quad (53)$$

$$R^{+} + HO^{-} \rightarrow RO \quad (54)$$

$$R^{\cdot} + H_2O_2 \rightarrow ROH + HO^{\cdot} \quad (55)$$

$$R^{\cdot} + O_2 \rightarrow R(-H^{+}) + H_2^{\cdot}O \quad (56)$$

$$R^{\cdot} + O_2 \rightarrow ROO^{\cdot} \rightarrow \rightarrow RO^{\cdot} \quad (57)$$

$$HO^{\cdot} + Fe^{2+} \rightarrow HO^{-} + Fe^{3+}, k3.2x \quad 10^{8} \quad M^{-1}s^{-1} \quad (58)$$

$$HO^{\cdot} + H_2H_2 \rightarrow H_2O + H_{2O}^{-}, k = 2.7x \quad 10^{7}M^{-1}s^{-1} \quad (59)$$

$$HO^{\cdot} + H_2H_2 \rightarrow H_2O + H_2O^{-}, k = 2.7x \quad M^{-1}s^{-1} \quad (60)$$

$$HO^{\cdot} + HO^{\cdot} \rightarrow H_2O_2, k = 4.2x \quad 10^{9}M^{-1}s^{-1} \quad (61)$$

Fenton oxidation technique proves more beneficial due to its easy operation as well as sophisticated degradation efficacies. More influence on the usage efficacy take place owing to the pH value, Fe^{2+} and hydrogen peroxide concentration as well as temperature (Brillas and Oturan, 2009; Wang, 2018c; Bai, 2017).

1.7. Photo-Fenton type reaction (PFR)

The Fenton reaction aided through ultraviolet visible light irradiation, called photo-Fenton process, which leads to the increase of degradation (Brillas and Oturan, 2009; Bai, 2017). The photolysis of H_2O_2 occur using UV light because hydrogen peroxide has maximum absorbance under range of 210–230 nm Eq. 62.

$$H_2O_2 + hv \rightarrow 2HO^{\cdot}, \varnothing OH = 0.5 \quad (62)$$

By visible UV light radioactivity, dilapidation of different dyes has been perceived in to PFR because of transfer of electron from the visible light, Fe^{3+} clause the catalytic cycling of Fe^{3+}/Fe^{2+} as shown in Eqs. 63–64 (Brillas and Oturan, 2009; Ma et al., 2005).

$$Dye + hv \rightarrow Dye^{*} \quad (63)$$

$$Dye* + Fe^{3+} \rightarrow Fe^{2+} + Dye^{+} \quad (64)$$

In Fenton process, an important factor is catalytic dosage, which put crucial impact on the organic pollutants degradation. Due to overdose of hydroxyl radical (catalyst), organic pollutant degradation may be inhibited and excessive catalyst dosage may raise the cost and practical applications might be affected (Wang, 2018c; Ma et al., 2005). Ciprofloxacin degradation by aqueous solution studied by diao et al. and FeS_2/SiO_2 microspheres as a heterogeneous Fenton catalyst used by the diao et al. to activate H_2O for degradation purpose. By FeS_2/SiO_2, microspheres activated H_2O_2 degradation reach almost 100% within 60 min only. Due to the attachment of SiO_2 microspheres on the surface of FeS_2 resulting in the smooth reaction between Fe^{2+} and H_2O_2 by controlling the aqueous Fe^{2+} release from FeS_2 (Wang, 2018c; Ma et al., 2005). By increasing the catalytic dosage from 0.005 to 0.1 g/L, degradation efficiency also enhanced from 40.96% to 84.29% just because of the huge active sites, which cause the production of •OH by the breakup of H_2O_2. Table 1 show antibiotic removal by Fenton type's

Table 1
Representation the antibiotics removal by Fenton like Oxidation Process.

Antibiotics	Catalyst (dosage); pH range	Removal efficiency (%)	References
	[Fe_3O_4 (1.0–2.5 g/L)]; [pH = 3 – 11]	89 (%)	(El-Temsah et al., 2016)
	[CNTs/FeS (5–35 mg)]; [pH = 1–12]	91.03 (%)	(Mitra and Varshney, 2013; Shoiful et al., 2016)
	[H_2O_2/Fe^{2+} = 1.75 mM]; [pH = 3]	95 (%)	(Shoiful et al., 2016)
	[(SBC)] = [pH= 4]	> 80 (%)	(Lopes et al., 2008)
	FeS_2/SiO_2 microspheres activated H_2O_2	100 (%)	(Affam et al., 2016b)
Ciprofloxacin	Fe_3O_4/MWCNTs (30–200 mg/l), H_2O_2 (5–25 μmol/l); pH= 4–10	95%	(Ghisi, 2014)
	Fe_2O_3/RGO-ATP, H_2O_2 (2.9724 mmol/l) pH= 5	88.27%	(Iqbal et al., 2021a)
	FeCu/ABC (1 g/L), pH= 5.8	92%	(Sun and Pittman, 2000)
	0.03TiO_2/γ-Fe_2O_3/GO [Fe(II)] (0.8–3 mM)	99%	(Focazio et al., 2008) (Wasiewicz and Getoff, 2006)
Norfloxacin	Alg/Fe (0.2–1.4 g/L); pH = 3	100	(Macwan and Chaturvedi, 2011)
	[Alg/CDTA/Fe (0.01–0.09 g)]; [pH = 3]	100	(Macwan and Chaturvedi, 2011)
Ofloxacin	[CQDs/Cu-MIO (0.1–0.25 g/L)]; [pH = 3.6 – 10]	100	(Oller and Sánchez-Pérez, 2011)
	[Fe-Cu@MPSi (0.5–1 g/L)]; [pH = 3 – 9]	100	(Oller and Sánchez-Pérez, 2011)

CNTs (Cabon Nano Tubes), (SBC), Sesbania bispinosa biochar (MWCNTs), Multiwall Carbon Nano Tubes), RGO-ATP (Reduced Graphene Oxide-Attapulgite), ABC (Activated Biochar), Alg/CDTA, (Alginate cyclohexane dinitrilo tetraacetic acid), CQDs, (carbon quantum dots),

oxidation.

1.8. Photo-Fenton types of reaction through chloride ion

Because of the presence of Cl^- ion, the amount of mineralization in (PFR), compare to that detected in the equivalent thermal Fenton reaction which proceed in dark environment both in the presence and absence of Cl^{-1} ion as shown in Fig. S3. However, this had attributed (Maciel and Dezotti, 2004; Devi et al., 2013), that the inhibition to the special generation of less reactive Cl_2^- radical ion instead $^{\bullet}OH$. The nanosecond laser lash was used in order to examine the effect of the added Cl^- ion at the photocatalytic step which caused back conversion of iron (III) to iron (II). However, finding of $^{\bullet}OH$ directly through spectroscopic has showed intangible, the chloride radical anion, that absorb at the range 340 nm, can be detected at the excitation of aqueous solution of Fe(III), at acidic media along with NaCl at 355 nm and third harmonic of a Nd-YAG laser. Because of differential absorption spectra and kinetic traces as shown in Fig. S3, exhibited the formation of chloride radical anion, within life time of the laser pulse. The absorption decrease for long time comparative of the laser pulse, returns the adaptation of Fe (III) to Fe (II) which does not absorbed in the spectral rang. The consequent reaction why Photo-Fenton type of reaction in summarized in Table S3.

1.9. Catalytic behavior of photo-Fenton (CPF) type reaction with complexation of Iron (III)

The proficiency of the CPF can be improved by the combination of CH_3CO_2H with Fe (III) (Pignatello and MacKay, 2006). The oxalic acid is the best example for the self-explanation of CPF. Consequently, thermal Fenton Reaction (TFR), oxalic acid is intractable intermediate, act as catalyst in the PFR. The $[Fe\ (C_2O_4)_3]^{3-}$ absorbed light up to 750 nm. Furthermore, due to radioactivity the decay proficiency to iron (II) and carbon dioxide. The results show that the CPFR rate effected and increase because in the presence of $C_2O_4^{2-}$. The literature reported (Kim and Vogelpohl, 1998), that with UV radiation the PFR was at least 30% is much better due to the presence of oxalate than in the absence of oxalate. The Ferrioxale-catalyzed PFR is more sensitive to UV and UV/visible light, particularly sun light is more attractive sources (Silva et al., 2010; Nogueira, 2005). Sulfate ion frequently present in PFR due to which addition of Fe in the form of eagerly accessible sulfate salts (Nogueira, 2005). Sulfate ion strongly complexes with iron (III) at different pH range as shown in Fig. 1. and the quantum yield creation of iron (II^{2+}) and SO_4^{2-} radical from photolysis of $Fe(SO_4)^+$. Iron phosphate is even more photo inert than the iron Sulphate complex and more influential inhibiter of the PFR (Lee et al., 2003).

1.10. Electrochemical advanced oxidation processes

EAOPs used widely as an effectual and effective wastewater treatment techniques. During this techniques different types of reactive species are generated that responsible the oxidative removal of organic contaminants (Chaplin, 2014). The main benefit of Electro oxidation (EO) of the environmental compatibility, flexibility, energy effectiveness seeming as a procedure for eradicating contaminants from wastewater (Liu et al., 2009). The improvement and solicitation of the ECT (electrochemical technologies) in wastewater conduct have been studied and the oxidation mechanism of pollutants at anode have been reviewed (Song et al., 2010). At anodic oxidation, H_2O molecules decomposes into radical, whereas, H_2 gas is produced and not participate in the oxidation of organic toxins. The EC process used for the treatment of waste water mainly focus on the anodic oxidation. During EC reaction the electrode materials play significant rule in electrochemical oxidation through the prompting the efficiency of the oxidation, dilapidation and mechanistic pathway (Zhu et al., 2008). On the basis of mechanistic pathway of the organic pollutants oxidation, the materials are classified into two main categories, which is active anode like Pt, IrO_2 & RuO_2, while inactive anode including PbO_2 and SnO_2 (Brillas and Oturan, 2009; Zhu et al., 2008). The initiated reaction in both kind of anodes (which is generally recognized as M), corresponds to the oxidation of H_2O and created active O_2, absorbed OH (M) radical Eq. 65 (Brillas and Oturan, 2009).

$$M + HO^{\cdot} \rightarrow MO + H^+ + e^- \tag{65}$$

The hydroxyl radical is strongly interacted through the surface of the active anode which caused transformation into higher oxide (MO) Eq. 66 (Ferro, 2006). When superoxide sate are available for metallic anode, the standard potential for oxygen evaluation ($E^{\circ} = 1.23$ vs SHE), the adsorbed OH may form chemisorbed active oxygen (Ferro, 2006).

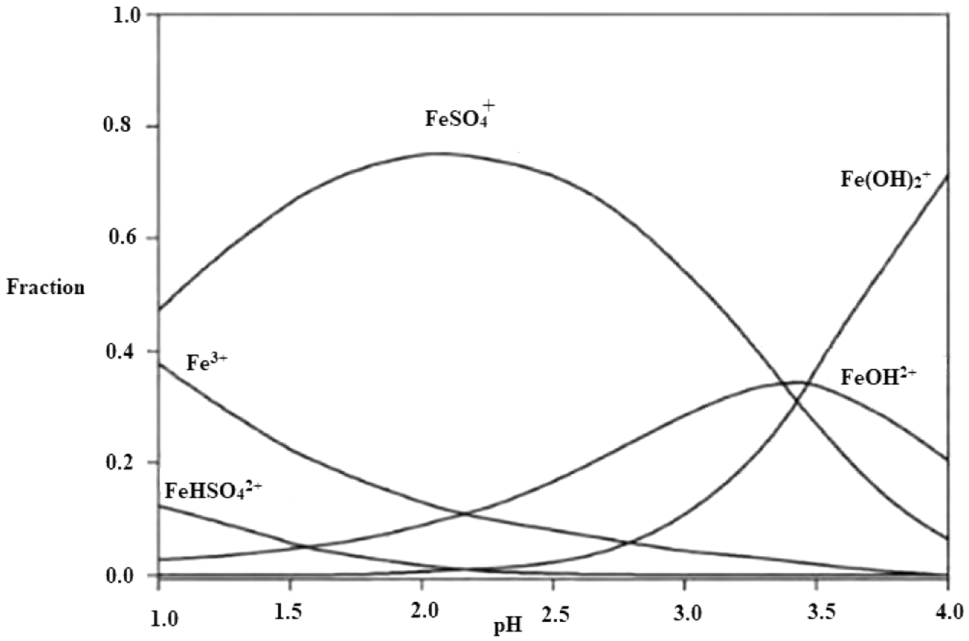

Fig. 1. 1.0 mM Iron III b/w pH one & four at room temperature at 0.2 ionic strength with 1.8 mM Sulphate.

$$M + HO^{\bullet} \rightarrow MO + H^{+} + e^{-} \quad (66)$$

The MO show weaker oxidizing capability as compared to OH radical, which involve in the oxidation of organic pollutants (R), and MOM, (redox couple) acts as a mediator as shown in reaction **67**, whereas, competition, reaction take by the evaluation of O_2 through chemical decomposition of MO shown in **68** (Ferro, 2006).

$$MO + R \rightarrow M + RO \quad (67)$$

$$MO \rightarrow M + \left(\frac{1}{2}\right) O_2 \quad (68)$$

Weaker oxidant O_3 can be produced from H_2O discharge at anodic surface ($E = -1.51$ V vs SHE) as shown in reaction **69**. During the process, small amount of hydrogen peroxide also generated as shown in reaction **70**. The other reactive oxygen species like hydrogen peroxide are produced in the electrochemical oxidation.

$$3H_2O \rightarrow O_3 + 6H^{+} + 6e^{-} \quad (69)$$

$$3H_2O \rightarrow H_2O_2 + 2H^{+} + 2e^{-} \quad (70)$$

The mechanistic oxidation of the organic contaminants at the surface of nonnative anode are elucidated in Fig. S5. Table S5 showed the role of different AOPs in the removing the CEC from wastewater.

It has been reported that electrochemical oxidation of Tetracycline at anode surface (Ti/Ti_4O_7) an intermediate is created in the tetracycline deprivation pathway having different toxicities and the toxicity delayed the degradation process. The Table S6 show the summaries of the most of the antibiotic treatment with heterogeneous /electro-Fenton reactions.

The main advantage of the electrochemical system is the lower effective cost, ecofriendly and lower labor are required and most safety risk that is useful in the degradation of drugs and other organic pollutants. The literature reported that dual anode and Pd/CeO_2 Nano catalyst for the degradation and removal of sulfadiazine antibiotic while using electro-Fenton system. Table 1 show the summaries more antibiotic treatment with EFR (Zhang et al., 2020).

1.11. Characterization of $^{\bullet}OH$ at anodic surface

The distinctive target of concert for electrochemical advance oxidation process, anode materials is comparative production of hydroxyl radical. Two different method are involved in the production of •OH radicals and have distinct disadvantage which can be lead to the misinterpretation of Hydroxyl ion production. The first one is related to electron spin resonance [ESR] with spin trap compound which reacted with hydroxyl radical to form an adduct species or involve the generation of •OH radical is conditional by the disappearance of the probe constituents. It is clear that both the method has deficiency, when used to characterize EC produced •OH radical. Both type of molecules react with on the surface of electrode either by transfer of *en* followed by Nucleophilic substation reaction through H_2O to form adduct which give false positive •OH radical (Leinisch et al., 2011; Mason, 2011). The process is called inverted trapping as shown in Eq. 71.

$$R \quad \rightarrow^{e^-} R^{\bullet +} \rightarrow^{OH^-} R - OH \quad (71)$$

Another reaction mechanism result in false positive detection, which is known as the Forrester-Hepburn mechanism and the reverse o inverted spin trapping. During this proceed the Nu attack of the spin trap by water followed by the direct *en* transfer oxidation reaction as shown Eq. 72 (Chaplin, 2018).

$$R \rightarrow^{OH^-} ROH^{-e} \rightarrow^{e^-} ROH \quad (72)$$

The results of the both the method show the same results, the generating of radical adduct (R-OH) that is same to that prepare by the hydroxyl radical attack on the original spin trap.

2. Mineralization of pollutants

Usually, antibiotics are completely mineralized by Ozonation process and during mineralization process carbonate (CO_3^{2-}) and bicarbonate (HCO_3^-) are formed which act as hydroxyl radical scavengers and result in inhibition of antibiotics removal. On the other hand, decreased solution pH during Ozonation reaction is proceeding oppose the generation of hydroxyl radicals ($^{\bullet}OH$). Complete norfloxacin degradation observed by kuang (De Laat et al., 1999) after Ozonation process without any mineralization. Total Organic Compounds removal efficiency of Ofloxacin was 33.5% after 180 min Ozonation of Ofloxacin solution as described by Goncalves (Nidheesh, 2015).

Ciprofloxacin, norfloxacin and Ofloxacin evaluated by Fenton oxidation method (Liu et al., 2012). Impact of pH on ciprofloxacin degradation studied by Wan and Wang (2016) employed Ce°/Fe°-RGO composites as Fenton catalyst (Liu et al., 2012). By increasing pH value from 6.0 to 8.3, ciprofloxacin removal declined because as pH changes, adsorption efficacy of ciprofloxacin on catalyst surface also effected. On ciprofloxacin surface negative charged catalyst would repel its anionic form because 7.42 pH value is pKa_2 of ciprofloxacin which not only declining the adsorption efficiency but also effect the oxidation method (Wang et al., 2015; Xu et al., 2013). Norfloxacin degradation investigated (Wang et al., 2015) and as a Fenton catalyst zero-valent iron and Fe°/CeO_2 employed. The results showed that as pH increased from 3.0 to 5.8 the degradation efficiency of Norfloxacin decreased from 93% to about 50% and *n*ZVC alone employed as a catalyst. Combine use of Fe°/CeO_2 at pH 3.0–5.8 value resulting maximum degradation of Norfloxacin and exhibit extreme reactivity at various pH values (Xu et al., 2013).

2.1. Biodegradability improvement of pollutants

To differentiate biodegradability of a waste product or wastewater typically BOD/COD ratio is employed. Generation of biodegradable and low molecular weight intermediates by Ozonation method is used to improvise the biodegradability of antibiotics containing wastewater. After Ozonation, biodegradability of discarded water is increased as confirmed by Balcioglu and Otker (Kakavandi et al., 2019). The oxidation of one electron of aromatic compounds created a reactive species cation, which is followed by •OH-cyclohexadienyl radical through hydrolysis (Rayaroth et al., 2022). At least hydroxylated product formation take place by the addition of O_2 which caused eliminating of water molecule (Rayaroth et al., 2022). Meanwhile the nitro-products is generated by the de-nitration and re-nitration process. In the initial reaction nitrite NO^{2-} release in de-nitration reaction. The newly generated NO^{2-} again reacted with another reactive species SO4•- and hydroxyl radical to produce a strong nitrating agent NO•2. The rate of nitration of reaction and rate constant is shown in Eqs. 73–74 (Neta et al., 1988). One *electron* oxidation of mono nitro phenol created a nitro hydroxyl cyclohexadienyl radical and successive nitration yield di-nitro phenol as shown in Fig. S5. The main di-nitro phenol seen during reactions is 2, 4-DNP, that generated from both 2-NP and 4-NP, as 2, 6-DNP produced only from 2-NP. Other conversion reaction on di-nitro products from tri-nitro products while other products shown in Fig. S5 (Rayaroth et al., 2022).

$$NO_2^- + SO_4^{\bullet -} \rightarrow NO_2^{\bullet}, k = 8.8x \quad 10^8 M^{-1} s^{-1} \quad (73)$$

$$NO_2^- + HO^{\bullet} \rightarrow NO_2^{\bullet}, k8.8x \quad 10^8 \quad M^{-1} s^{-1} \quad (74)$$

During nitration reaction in the presence of nitrate and nitrite is a common process in the degradation of organic pollutants in H_2O, which contains both the nitrite and nitrate ions (Neta et al., 1988; Goldstein, 2007). Both the species generated during the nitration process have capacity to absorbed maximum wavelength of λ 300–350 nm (Attri et al., 2015; Marussi and Vione, 2021). The reactive nitrates and oxygen

species are generated depending upon the wavelength of radioactivity as shown in Fig. S7.

2.2. Degradation of pharmaceuticals by zero valent metals

Continuously pharmaceutical products introduced into the environment from different sources like hospitals (Yamaguchi et al., 2018; Chen et al., 2019), personal care products (Yamaguchi et al., 2018; Chen et al., 2019), pharmaceutical industry waste (Kakavandi et al., 2019; Yamaguchi et al., 2018; Chen et al., 2019), and inappropriate discarding of drugs (Alheety et al., 2019). Small amount of toxic pollutants is considered possibly injurious to ecosystems. Enormous public health problems arise due to the traces of xenobiotic elements as well as narcotics, which are present in drinkable water and polluted food (Alheety et al., 2019; Yang et al., 2020). Adsorption, reduction and oxidation mechanism followed by the Fe° and oxidants are considered reasonable in degradation of different hazardous pharmaceuticals and these drugs removal mediated by the Fe° surface. Active points blocked by the FeO or $Fe(OH)_2$ and electron transfer also inhibit by the surface that are responsible for antibiotics reductive degradation at same time, when the electrostatic collaboration among iron oxide/Fe° improvements its absorption as well as intermediates also and vital role played by antibiotics in elimination method (Pan et al., 2019). Greater or lesser interaction developed due to the drugs, which depends on charge and iron surface. Best transfer of electron promoted in acidic pH and ROS formation occurred, which involves in oxidative degradation (Pan et al., 2019; Lumbaque et al., 2019). The 17 α-ethinylestradiol as an ideal steroidal estrogen reported by karim and via viably sourced *n*-Fe° its degradation studied under diverse oxygen conditions and among many pH values 3–5 and 7 $^{\bullet}OH$ was main radical at pH 3 as well as O_2^{-} at pH 5 (Pan et al., 2019). From unlike usual waters Fe° could be working to powerfully remove chloramphenicol within few minutes (Liu et al., 2018a, 2018b), representing that to lessen antibiotic selection compression, *n*-Fe° is an favorable solid that can be exploited in the atmosphere. Deduction of amoxicillin described and its degradation based on the incorporation of adsorption and reductive process. In AOPs, fabrication of extremely reactive species ($HO^{\bullet}$, $O^{\bullet}$, $H^{\bullet}$, and H_2O_2) allowed by the usage of ultrasonic radiation and by thermal fragmentation of H_2O particles as well as biological solutes and it involve development and successive fragmentation of bubbles through acoustic cavitation (Ghosh et al., 2012; Hameed, 2010). Fig. S8 represents the diagram of the antibiotics degradation using Fe°/H_2O_2 system and degradation pathways of antibiotic ibuprofen using Fe°/H_2O_2 is represented in Scheme S1.

Scheme S1 represents the degradation pathways of antibiotic ibuprofen using Fe°/H_2O_2 system as given below.

Table S7 show the analysis of the usage of Fe° as a catalyst in dissimilar systems and in latest studies regarding the removal of pharmaceuticals by Fe°, that transports indications about matrix performance and about the identification of TPs (Shah et al., 2020a). The primary gatherings of pharmaceuticals in distilled or deionized H_2O were antibiotics and anti-inflammatory mixtures working in different studied including methods with Fe°. Exclusion percentages of parent mixtures in the majority of situations exhausted 70% but the toxicities and TPs identification are still focus of exploration. According to the utilized substance amount, the reaction time can vary not only the nature of the matrix but physicochemical structures of the antibiotics as well. In acidic pH, prospered degradation efficiency values was higher, but some researches also confirmed the more than 60% removal at pH at 7 (Iqbal et al., 2021a; Sayed et al., 2019; Khan et al., 2017).

Zero valent manganese (*n*ZVMn) used first time for Ciprofloxacin degradation due to its nano dimension, crystalline as well as highly stable nature by addition with $S_2O_8^{2-}$. When Mn° applied alone than CIP degradation was not much higher but CIP degradation reach up to 95% with $S_2O_8^{2-}$. When Mn° applied alone, and reaction conditions set according to the following manner i.e., $[CIP]_0 = 10$ mg/L and $[Mn^{\circ}]_0 = 1.0$ g/L than obtained mortification of CIP was 63% within 80 min of reaction. But as $S_2O_8^{2-}$ combined with Mn° than reaction conditions set in such manner as given here i.e., $[CIP]^{\circ} = 10$ mg/L, $[Mn^{\circ}]_0 = 1.0$ g/L, and $[S_2O_8^{2-}]_0 = 50$ mg/L than degradation proficiency reached up to 95% within 80 min (Iqbal et al., 2020; Shah et al., 2020b). Due to the $^{\bullet}OH$ and $SO_4^{\bullet-}$ radicals, faster kinetics exhibited by the ciprofloxacin but CIP removal repressed to some amount in presence of $^{\bullet}OH$ and $SO_4^{\bullet-}$ scavengers. CIP removal directly linked with the $^{\bullet}OH$ and $SO_4^{\bullet-}$ formation. As $[S_2O_8^{2-}]_0$ and $[Mn^{\circ}]^{\circ}$ amount enhanced in reaction conditions, degradation level also increased and CIP degradation pathway by means of $^{\bullet}OH$ and $SO_4^{\bullet-}$described in Scheme S2. Proposed CIP degradation mechanism was expectable when $^{\bullet}OH$ and SO4.− was undergo H-abstraction, electron abstraction as well as double bod addition to convert CIP into its DPs that also undergo investigation resulting same pathway. It was found that $^{\bullet}OH$ and $SO_4^{\bullet-}$ were also comprised DPs degradation as well and it shows that TPs with CIP undergo opposition for $^{\bullet}OH$ and SO4.− (Shah et al., 2019). Shah et al. applied same pattern for CIP degradation by means of Cu° mediated H_2O_2 based Fenton-like reaction. The use of Cu° led to 63% removal of CIP, which was just because of extraordinary surface area and therefore cause great accumulating of CIP particles (Iqbal et al., 2020; Shah et al., 2019). The addition of H_2O_2 to Cu promoted CIP removal and caused 85% removal at 105 min using $[H_2O_2]_0 = 40$ ppm, $[Cu^{\circ}]_0 = 0.5$ g/L, and $[CIP]_0 = 10$ ppm. CIP removal by Cu°/H_2O_2 was found due to $^{\bullet}OH$ and $^{\bullet}OH$ showed extraordinary reactivity with CIP. CIP removal was promoted at high $[H_2O_2]_0$ and $[Cu^{\circ}]_0$ while declined at high $[CIP]^{\circ}$ (Shah et al., 2020b, 2018; Murtaza et al., 2019). Proposed ciprofloxacin degradation pathways via $^{\bullet}OH$-mediated processes given in Scheme S2.

2.3. Removal of pesticide through advance oxidation process

Due to high reactivity of the $^{\bullet}OH$ radical show high unitability and must generated during Fenton type reaction in AOP at 25 °C (Murtaza et al., 2019; Pham et al., 2020). Due to high reactivity, the $^{\bullet}OH$ reacted with unsaturated more frequently as compared to saturated hydrocarbon like organic acid, which are oxidant products. Mostly, the $^{\bullet}OH$ attack at the *Ortho* and *pera* position of the aromatic hydrocarbon. The inorganic compounds showed lower kinetic constant value as compared to organic compounds. The inductive donor can have stabilized the $^{\bullet}OH$ generated during the reaction. It is observed that the compounds deficient hydrogen atoms and unsaturation are protected to reaction by the •OH radicals.

3. Pesticides degradation by nano-zero valent materials

Many classes of pesticides are classified on basis of their target use as well as their chemical composition (Iglesias et al., 2015). Pesticides can contaminate the natural water sources as a highly soluble material after using the field. In natural sources, a significant increase for agrochemicals observed due to this case. Some essential ecological adverse effects continued due to these compounds on aquatic biomes and collected in the human body and wildlife. In addition, associated to agrochemical contagion of marine atmosphere bad financial and communal impacts are observed. Remediation of carbon-based pollutants becomes a growing concern owing to their hydrophobic nature. Recently, focus of investigators was to evaluate the harms caused by pesticides and herbicides to the earth's bio-network and human health (Njoku and Hameed, 2015; Zhang et al., 2014). In latest papers pesticide removal via zero-valent iron (ZVI) has been explored and reported and in this direction conducted researches assumed on DDT, triallate, nitro aromatic pesticides as well as triazines herbicides and organophosphorus insecticides, benzoic acid and benzimidazole pesticides and carbamate pesticide also (Yu et al., 2007). These pesticides are organo-chlorinated compounds frequently, which means in their arrangement one or more chlorine atoms are present in addition to nitrous and/or carboxylic groups (Marussi and Vione, 2021; Yang et al., 2019b). In the start of the 1990 s, early research carried out and for an massive number of

ecological scientists attractive material continuous to be a ZVI due to its non-poisonous, inexpensive, and influential reducing features (O'Hannesin, 1994; Matheson, 1994). *n*ZVI used as an effective substitute for the specific remediation of chlorinated derivatives that put difficulties towards the atmosphere by their venomousness and harmful effects in soil and water. Chemical oxidation method, photocatalytic oxidation techniques, filtration, adsorption onto activated carbon are different techniques that working to treat classical excesses and underground water but these above technical ways required too much cost (Iqbal et al., 2020). More freshly, inoculation of nano-scale ZVI openly in the aquifers technology employed for eco-friendly remediation. Cost effective solutions represented by this technique by avoiding vast excavation recognition, lessening cost installation as well as reaching deeper polluted areas.

3.1. Carbamate pesticides

Specifically Dichlorvos are absolutely mineralized in the appearance of ZnO and TiO_2 and used in variety of field crops. Apart from nanoparticles, to degrade insecticides there are various information on the use of nanotubes and nanostructures as well as thin films. TiO_2 nanotubes and TiO_2 (Matheson, 1994; Midik Ertosun et al., 2019) thin films were utilized for *atrazine* degradation and of *organochlorine pesticides* degradation respectively (Marussi and Vione, 2021; Yang et al., 2019b). Titania changed graphite anode by electro-oxidation management was efficaciously employed for the degradation of *2,4*-dichlorophenol (Midik Ertosun et al., 2019; Rojas et al., 2015). Carbon built nanomaterials at the nano-scale bonds the sp^2 hybridized carbon bonds properties with the characteristics of physics and chemistry. Over the arrangement and conformation of a material, molecular handling implies mechanism for carbonaceous nanomaterials counting size, dimension, chirality and the number of layers (Rojas et al., 2015). Organochlorine pesticides mortification by photocatalytic degradation of nano-TiO_2 film studied (Rojas et al., 2015). They specified that on TiO_2 surface, attraction of peroxide or hydroxyl radicle and transmission of electron qualifies the photolytic degradation of insecticides (Rojas et al., 2015; Vukčević et al., 2015). .

Table 3 represents the degradation of various types of pesticides by using different kind of nanomaterials.

3.2. Flutriafol

Extremely persistent water/soil triazole pest killer is termed as Flutriafol and it contain high fungicidal actions and after its successful synthesis, it is broadly used since 1981 by the British Imperial Chemical Industries. In the international fungicide market, after its introduction an imperative position accomplished by this complex where Flutriafol products used to control massive diseases that upsets an extensive range of crops (Carmona et al., 2020). Flutriafol degradation via an oxidation method happened with micro scale nZVI, so this can connect the results gotten with nanoscale nZVI in a higher or a lower amount of oxygen, which noticeably expands the applicability of ZVI as a theoretically useful reagent for the water and wastewaters pollutants mortification (Krebsz, 2020).

3.3. Nitro aromatic pesticides

This types of pesticide were quickly condensed with ZVFe to the subsequent amines as major reduction byproducts. The nitro intermediate were initiate with very little amount in some reactions (Affam et al., 2016a; Esteve-Núñez and Ramos, 2001). Quinone, a Dichlone, neutralized the inhibitory effect C_9H_9NO (humic Acid, HA) on the catalyst proficiency of ZVFe for reduction process of $C_{13}H_{19}N_3O_4$, (pendimethalin PD) which recommends that some neutral quinone, be relevant to pollutant contamination via ZVFe (Keum, 2004). Though the use of ZVFe for organic pollutants degradation is inexpensive and reasonable. it is very to develop pathway for the enhancement, catalytic stability and recovery the oxidized iron (Krebsz, 2020). The H_2 gas elimination rapidly improved the catalyst stability and ability of air-exposed iron and cause PD reduction, which suggests H_2 elimination as possible mean to continue a reactive ZVFe barrier for the related remediation technology (Ghauch, 2008; Galdames et al., 2020). It is reported that the ZVFe can be used in combination with microbial approaches which may provide H_2 and quinine for the removal of oxidized organic contaminations. Fenton oxidation with Fe ^{o} removed 99% of MTBE at pH 4–7 along with hydrogen peroxide MTBE molar ratio 220:1. The C_3H_6O (acetone) concentration generated by this process was approximately 400 mg/L (Thies, 2004). When hydrogen peroxide and MTBE was used in molar ratio 440:1, an effective reduction of MTBE take place with and final C_3H_6O reduced to approximately 60 mg/L (Thies, 2004). The 2nd order MTBE removal were found to be 1.9×10^8 M^{-1} s^{-1} and 4.4×10^8 M^{-1} s^{-1} at pH 7.0 and pH 4.0 respectively while second order rate constant for hydrogen peroxide degradation to be 0.44 M^{-1} s^{-1} and 0.23 M^{-1} s^{-1} at pH 7.0 and pH 4.0 respectively.

3.4. Endosulfan (ES) pesticides

Endosulfan (ES) is most broadly utilized organochloride pesticides and is acutely neurotoxic to fishes and other marine creatures (Jayaraj et al., 2016; Esfahani et al., 2023). Endosulfan became contentious, when its acute toxicity and role as an endocrine disruptor were brought to light. According to existing confirmations all recognized natural attenuation of ES residues i.e., ES-metabolites are very much cancerous and they retain the original chlorinated ES skeleton (Ford et al., 2007). The mass spectra of the mortified products were reliable with the loss of single, double and triple chlorine atoms from the parental compounds through dechlorinating by *n*ZVFe (Yang et al., 2016). Further, by aggregating the intensity of the reducing environments the extent of dechlorinating could be enhanced. To conclude that the impact of the current investigation lies in the statement that partially or fully dechlorinated degraded products of ES and ES-metabolites may be least noxious and more responsive to consequent biodegradation and definitive mineralization in the natural surroundings (Nowak et al., 2021). For stepwise reductive dechlorination of ES-isomers and their metabolites is degradation mechanism proposed in a pathway as shown in Fig. S9 (Bose, 2017).

3.5. 2, 4-Dichlorophenoxyacetic acid

The 2, 4-dichlorophenoxyacetic acid (2, 4-D) acts as the herbicide, its broadly utilized in gardening agricultural practices due to its low cost (Yang et al., 2019b; Islam et al., 2018). It more efficiently attacks broad-leaf weeds. 2, 4-D shows extraordinary soluble nature and unfortunately non-biodegradable as well as its free acid exhibit has little soil adsorption coefficient (Yang et al., 2019b). The 2, 4-D can easily transfer by soil to ground water by leakage (Meftaul et al., 2020). According to WHO, 100 ppb is its acceptable maximum concentration and it considered as a moderately lethal (Tolgyessy, 1993; Bao et al., 2019). By *n* ZVI, reductive dechlorination in 2,4-D destruction was main technique and on basis of outcomes main factors for removal are dosage of *n* ZVI and initial pH of resolution as shown in Figures S10 and S11.

Table 2
Constant value for OH radical reaction with inorganic and organic compounds.

Compounds	$CO_2^{\cdot-}$	HSO_4^{2-}	H_2O_2	Fe^{2+}	Cu^{2+}	Ag^{+}	ClO^{-}	Cl	Benzene	Chloro-benzene	Trichlor-ethylene
k(105 xmol/L/s)	8	3.5–17	270	3200	3500	120000	8800	43000	78000	40000	40000

Table 3
Degradation of pesticides by different nanomaterials.

Pesticides	Nanomaterial	Brief summery	Mechanism	References
Thiamethoxam, imidacloprid	Zero-valent metals (Fe, Sn, Zn)	Degradation [90% after a reaction time of 30 min via initial NO_2-[NH_2 reduction	Ultra-sonication	(Abdeen and Mohammad, 2014)
Thiamethoxam, imidacloprid	Fe^0/Fe_3O_4 with/or H_2O_2	Highly efficient in acidic conditions	Redox degradation	(Carneiro et al., 2015)
Terbufos	TiO_2	99% degradation within 90 min	Photocatalysis	(Manimegalai et al., 2014)
Monocrotophos, dichlorvos	TiO_2–zeolite	100% degraded	Photocatalysis	(Rani and Shanker, 2018; Pius, 2015)
Malathion	Fe°	Quantitative oxidation in soil within 8 min	Oxidation	(Konrad et al., 1969)
DDT	Fe°	Effective degradation in water (92%) than soil (22.4%)	Reductive degradation	(El-Temsah et al., 2016)
Endosulfan	Cu°	Completely annihilates in solution	Degradation	(Mitra and Varshney, 2013)
Lindane, DDT and aldrin	Fe° and Fe_3O_4	100%, 81% and 79%, respectively, after 12 h reaction	Redox degradation	(Shoiful et al., 2016)
Thiamethoxam, imidacloprid	Fe°/Fe_3O_4 with/or H_2O_2	Highly efficient in acidic conditions	Redox degradation	(Lopes et al., 2008)
Chlorpyrifos, cypermethrin, chlorothalonil	Coating of Fe–granular activated carbon	With H_2O_2, complete degradation	Oxidation	(Affam et al., 2016b)

Momentous influence played by these variables on the degradation rate (Zhao et al., 2020). As ZVI nanoparticles dosage enhanced, exclusion productivity of 2,4-D also enhanced but its removal efficacy slightly raised after certain levels (Chen and Shih, 2008). *n*ZVFe shows very effective in water as well as polluted soil remediation with 2,4-D. *n* ZVFe is not only a source of Fe(II) but at same time it provides active sites and extreme surface area. Under higher conditions, *n*ZVI is an appropriate applicant for 2,4-D remediation because mortification by nZVI is more operative than Fenton and Photo-Fenton processes (FPRP). In removing chlorinated insecticides from discarded water, nZVI is suggested, as a innovative proficiency and capable handling possibilities in wastewater management skills (Castaño et al., 2021). A mechanistic proposal deprivation pathway of lindane with *N*-doped TiO_2 is shown in Scheme S3 (Philip, 2010; Sayed et al., 2016).

4. Trends observed in removal of pesticides

In the literature, agricultural waste carbonaceous materials, activated carbon clay minerals such as zeolite are wide range of adsorbents for the removal of various reported pesticides (Saleh and Al-Ghouti, 2020; Bish, 2013). In addition, chitosan and alginate like biopolymers (Sabbagh and Sharif, 2021), were also employed in pesticides adsorption. For the heterogeneous degradation of insecticides, diverse nanomaterials containing silver (Benelli, 2018), titanium dioxide (Abdennouri et al., 2016), ZnO and even nanocomposites were employed as a photocatalysis. *N* ZVI as such or in bimetallic form owing to its unique properties was used for the redox degradation of these toxic compounds (Bruckmann et al., 2022). Reactive sorbents like monocrystalline metal oxide were practical for the demolition of hazardous industrial mixtures plus extremely toxic OPs. Recently, the trend of removal of pesticide is following the use of nanocomposite and nano bio-composites via photo catalysis and adsorption mechanism (Bruckmann et al., 2022). For pesticide, removal reports are revealing on usage of nano bio-composites (Özmal, 2023). The PFR (Photo-Fenton reaction) is effective process for the oxidation of organic pollutants. Due the generation of •OH radical from hydrogen peroxide and UV light along with Fe^{2+} and Fe^{3+} ions which increase increases the rate of decomposition of organic pollutants (Özmal, 2023; Sheikhi and Aslani, 2021). The degradation through Fenton reaction is better in acidic media. The literature reported that Photo-Fenton reaction is more significant as compared to Fenton process (Ameta and Ameta, 2018). The photo-degradation of chlorpyrifos (CPF) under photo-Fenton process are summarized in Table 4, the results showed that PFR reaction is more significant and about 50% CP removed through this process (Gandhi et al., 2016).

The degradation of organophosphorus pesticides through AOPs take place at the surface of catalyst during the chemical reaction produced between oxidant species and pesticide in wastewater (Murillo et al., 2010). During this process reactive oxidative species are generated and initiated the removal process of pesticide (Flint, 1991). The degradation process is may effected by different factors like pH, catalyst, light, subtract and oxidant concentration (Murillo et al., 2010). The photo advance oxidation process is summarized in Table S11.

4.1. Effect of pH at photo-Fenton types of reaction

An imperative parameter is pH in Fenton-like practices for effective handling (Hussain et al., 2021; Wang et al., 2016; Vermilyea and Voelker, 2009). Suitable pH value in customary homogeneous Fenton methods is about to 3.0, while the optimal pH in the Fenton-like procedures be determined by on the reaction system exclusively and reaction tools mostly rely on the performance of catalysts (He et al., 2016). Most of the report show that the optimal pH is 3. The main reason of this pH for Photo-Fenton types reaction become perfect at the investigation of the Fe^{III} as function of pH as shown in Fig. 2a, absorption spectra of the relevant Fe $^{(III)}$ species as shown in Fig. 2**b**, pH < 2, main kind is Fe $(H_2O)_6^{3+}$ showed weak absorption in the UV range above 300 nm. Supersaturated Fe $^{(III)}$ solution is prepared at pH > 3 with reverence to preparation of colloidal $Fe(OH)_3$, precipitated of hydrated iron oxides stand for long time. However at pH 3, the most abundant Fe $^{(III)}$ present

Table 4
Removal/Degradation of CPF through Photo-Fenton process in aqueous solution.

Initial concentration	AOP features	Kinetic data	Detection Methods	Ref
CPF = 1 mg/L	UV Photo-Fenton Fe2 + ; H_2O_2 ratio = 0.05	Removal efficiency (30 min UV expose) CPF = 50.30% K= 3.3 × 10–4 min^{-1}	GC-MS	[244]
CPF= 100 mg/L COD= 1130 mg/L	UV Photo-Fenton H_2O_2: COD= (2:1) H_2O_2:Fe^{2+}= (25:1) pH= 3	complete degradation of the pesticides in 1 min Removal efficiency (60 min) COD= 78.56% TOC= 63.76%	HPLC	(Ameta and Ameta, 2018)
CPF= 30 mg/L	solar Photo-Fenton H_2O_2 = 0.01 M $Fe^{3+=}$10 mg/L pH= 3.5	complete degradation of CPF in 15 min	GC-MS	[245]

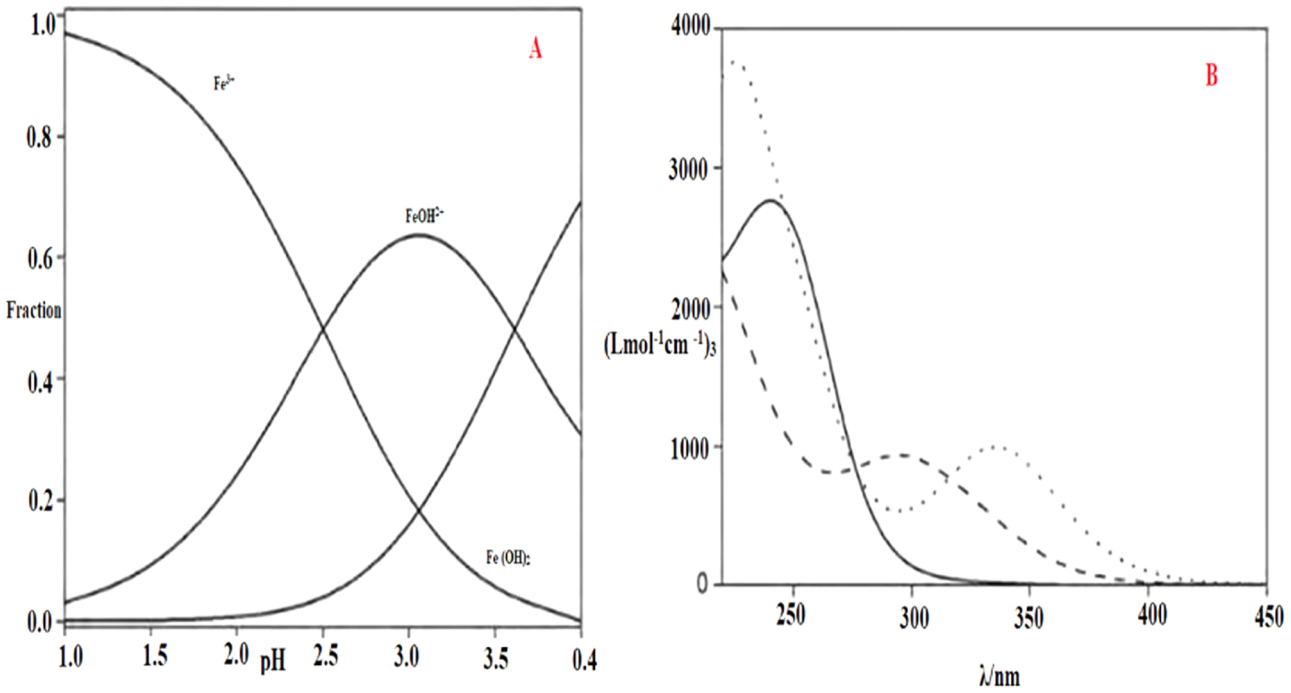

Fig. 2. Sample of 0.5 mM Fe (III) in between pH 1–4 at room temperature; (a). Solid spectra at pH 1.5 with $HClO_4$; 90% Fe (H2O) $_6$ $^{3+}$ or Fe^{3+} as shown in Fig. 2; (b). Dash spectra at pH 2.5 with $HClO_4$ 50: 50 FeOH; Fe^{3+} as shown in Fig. 2; (c). Dotted graph: pH 1.5 plus added 500 mmol L^{-1} NaCl, 30% $FeCl_2^+$ and 65% $FeCl^{2+}$ (2b). Absorption spectra of 0.43 mM Fe (III) studied at three specific environments.

is Fe $(H_2O)_5(OH)^{2+}$ which have the capability to absorbed more of the UV spectral region (Martyanov and Parmon, 1997; Xiao et al., 2020).

4.2. Reusability of catalyst

At industrial level the reusability of the catalyst plays an important rule index beside activity (Kapil et al., 2021). The deactivation of the catalyst is seen with the increase of reaction time because of sintering and change of the valance states (Kapil et al., 2021). The deactivation of the catalyst is estimated but in depth investigation on the causes of the catalyst deactivation can successfully recover the deactivation or sensitive the catalysts (Otor et al., 2020; Yuan et al., 2021). An essential application of catalyst processes involves the capability to isolate the catalyst from the reaction mixture, subsequently recovering and using it again for succeeding reactions within a heterogeneous system is performed with filtration, centrifugation and work up procedure (Kamalzare, 2022; Huang et al., 2023). The industry requirements the usage of catalysts for many reactions and the capability to improve and reuse the catalysts, making the catalyst process carefully reasonable for ascending up in industrial applications. Magnetic catalysts, which can be easily and rapidly separated from the reaction mixture using magnets, have recently garnered significant attention in both scientific and industrial research (Bayramoğlu and Ergan, 2021).

5. Conclusion

The advance oxidation process has developed as promising technologies for water decontamination remaining to their wide applicable, complete removal of organic toxic substances. The photolysis, electrophotocatalytic, cavitation, and electrochemical process organize some of the common AOP active for water decontamination. Advanced oxidation processes are highly effective techniques for breaking down organic contaminants in wastewater. These processes are capable of eliminating pesticides, drugs, dyes, plasticizers, and toxic heavy metals. These processes involve the production of free radicals, which react with the target contaminants, resulting in decomposition. Photo catalysis is the most significant of these processes due to its reliance on sustainable solar energy for the purification of wastewater. The Ultra violet/ sulfite process is recognized as the major advance discount procedure working in water decontamination. The AOP could be stated to a development based on the oxidation of the target toxic contaminants. The AOP is the most highly efficient technology and process in the degradation and removal of micro-pollutants from wastewater. All types of organic toxic pollutants including, organic dyes, plasticizers, toxic metal, drugs are successfully eliminated from water through advance oxidation process. The advance oxidation process, which uses O_3 for disinfection, in water worth, the removal of color and degradation of the organic materials. The AOP is alterative to the conventional treatment method for dyes removal. During AOP the OH radical initiated the reduction and removal of organic pollutants. Ionizing radiation and cavitation are also important processes due to their free radical nature. Ionizing radiation is deemed one of the most versatile advanced oxidation/reduction technologies because of its ability to simultaneously produce oxidizing and reducing species. These processes have shown great potential as a future alternative due to their high efficiency and versatility. Practical applications of these processes take into account various factors, such as initial pH concentrations, organic pollutant concentrations, concentrations of catalysts, the wavelength and intensity of light.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2023.115564.

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