Environmental Chemistry Letters (2018) 16:947–967
https://doi.org/10.1007/s10311-018-0738-3

REVIEW

Treatment of organic pollutants by homogeneous and heterogeneous Fenton reaction processes

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Received: 5 March 2018 / Accepted: 6 April 2018 / Published online: 20 April 2018

Abstract

Nowadays, the water ecosystem is being polluted due to the rapid industrialization and massive use of antibiotics, fertilizers, cosmetics, paints, and other chemicals. Chemical oxidation is one of the most applied processes to degrade contaminants in water. However, chemicals are often unable to completely mineralize the pollutants. Enhanced pollutant degradation can be achieved by Fenton reaction and related processes. As a consequence, Fenton reactions have received great attention in the treatment of domestic and industrial wastewater effluents. Currently, homogeneous and heterogeneous Fenton processes are being investigated intensively and optimized for applications, either alone or in a combination of other processes. This review presents fundamental chemistry involved in various kinds of homogeneous Fenton reactions, which include classical Fenton, electro-Fenton, photo-Fenton, electro-Fenton, sono-electro-Fenton, and solar photoelectron-Fenton. In the homogeneous Fenton reaction process, the molar ratio of iron(II) and hydrogen peroxide, and the pH usually determine the effectiveness of removing target pollutants and subsequently their mineralization, monitored by a decrease in levels of total organic carbon or chemical oxygen demand. We present catalysts used in heterogeneous Fenton or Fenton-like reactions, such as H_2O_2–Fe^{3+}(solid)/nano-zero-valent iron/immobilized iron and electro-Fenton-pyrite. Surface properties of heterogeneous catalysts generally control the efficiency to degrade pollutants. Examples of Fenton reactions are demonstrated to degrade and mineralize a wide range of water pollutants in real industrial wastewaters, such as dyes and phenols. Removal of various antibiotics by homogeneous and heterogeneous Fenton reactions is exemplified.

Keywords Homogeneous Fenton reaction · Heterogeneous · Advanced oxidation processes · Mechanism · Dyes · Antibiotics

Introduction

Water is abundant on earth and is critical to life. Of the total water mass, 97.2% is present in oceans and seas and 2.1% exists in glacier. 0.65% is the fraction of total water mass being utilizable for production of drinking water. This amount contains several lakes' waters which are highly polluted (Bakker 2012; Vörösmarty et al. 2010). Therefore, one of the important issues of this century is to provide clean water to humans (Liu and Yang 2012a, b; Shannon et al. 2008). Based on a recent report of United Nations and World Health Organization (WHO), more than 2 billion humans face some kind of risk to have safe drinking water at home (World Health Organization 2017). Water-related diseases have caused more than 360,000 children die every year under 5 years of age. Ecosystems are also subjected to polluted water-related risks (Schwarzenbach et al. 2006, 2010).

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Water pollutants usually contain diseases-containing agents, e.g., parasitic worms, bacteria, viruses, and protozoa, oxygen-consuming wastes, water soluble inorganics, e.g., toxic metals, nutrients, e.g., phosphates and nitrate, water soluble radioactive compounds and organic pollutants, e.g., plastics, oil, detergents, dyes, aryl chlorides, and pesticides (Kralchevska et al. 2016; Schmeller et al. 2018; Sharma and Sohn 2009; Sousa et al. 2018). In recent years, emerging organic contaminants in water have become major concerns. These include endocrine disruptor chemicals (EDCs) and pharmaceuticals and personal care products (PPCPs) (Cizmas et al. 2015; Hajj-Mohamad et al. 2017; Klatte et al. 2017; Sharma et al. 2016). Pollutants come from industrial, agricultural, and consumer products, which contaminate groundwater and surface water, commonly used as drinking water resources (Blum et al. 2018; Sousa et al. 2018). Water treatments have been greatly investigated for depollution of water for freshwater usage and drinking (Kim et al. 2014; Sharma et al. 2015; Werber et al. 2016).

Various treatment approaches have been applied which include adsorption, biodegradation, coagulation, ion-exchange, and oxidation processes (Brillas and Martínez-Huitle 2015; Feng et al. 2018; Ghattas et al. 2017; Sharma and Feng 2017). Among these methods, advanced oxidation processes (AOPs) have been researched tremendously for the last two decades (Anumol et al. 2016; Boczkaj and Fernandes 2017; Gassie and Englehardt 2017; Oturan and Aaron 2014). AOPs is based on generation of a powerful oxidizing agent such as hydroxyl radicals (˙OH) at a significant amount to effectively decontaminate water. Many different kinds of AOPs have been developed to produce in situ ˙OH radicals (Duan et al. 2018; Liu et al. 2018; Sillanpää et al. 2018). Chemical, sonochemical, photochemical, electrochemical processes have been utilized to form ˙OH radicals (Brillas and Martínez-Huitle 2015; Cheng et al. 2016; Ganzenko et al. 2017; Garcia-Segura and Brillas 2017; Gligorovski et al. 2015; Sharma 2013; Sirés et al. 2014; Steter et al. 2018; Trellu et al. 2016). The present review pertains to Fenton's reagent, a chemical strategy to efficiently generate in situ ˙OH radicals. In the literature, many approaches of Fenton reaction have been performed, which include homogeneous and heterogeneous Fenton reactions (Fig. 1). Examples are classical, modified Fenton reactions (e.g., sono-Fenton, photo-Fenton, electro-Fenton, photo-electro-Fenton, and ligand assisted Fenton) and solid-solution-based Fenton (H_2O_2–solid $Fe^{0,}$ H_2O_2–solid Fe^{III} (Barbosa et al. 2016; Clarizia et al. 2017; Gligorovski et al. 2015; Mirzaei et al. 2017; Moreira et al. 2016). In the next sections, fundamental chemistry of the Fenton reaction occurring in different systems to generate ˙OH radicals is presented.

Fenton's reagent

A mixture of ferrous ion (Fe^{2+}) and hydrogen peroxide (H_2O_2) is called Fenton's reagent. The chemistry of this reagent started in 1894 when Fenton applied it to degrade tartaric acid (Fenton 1894, 1896). Fenton's reagent involved complex mechanism of reactions, presented in Table 1. Basically, a Fenton process is initiated by the formation of hydroxyl radical (˙OH) (reaction F1) (Oturan and Aaron 2014). The reaction F1 takes place in acidic

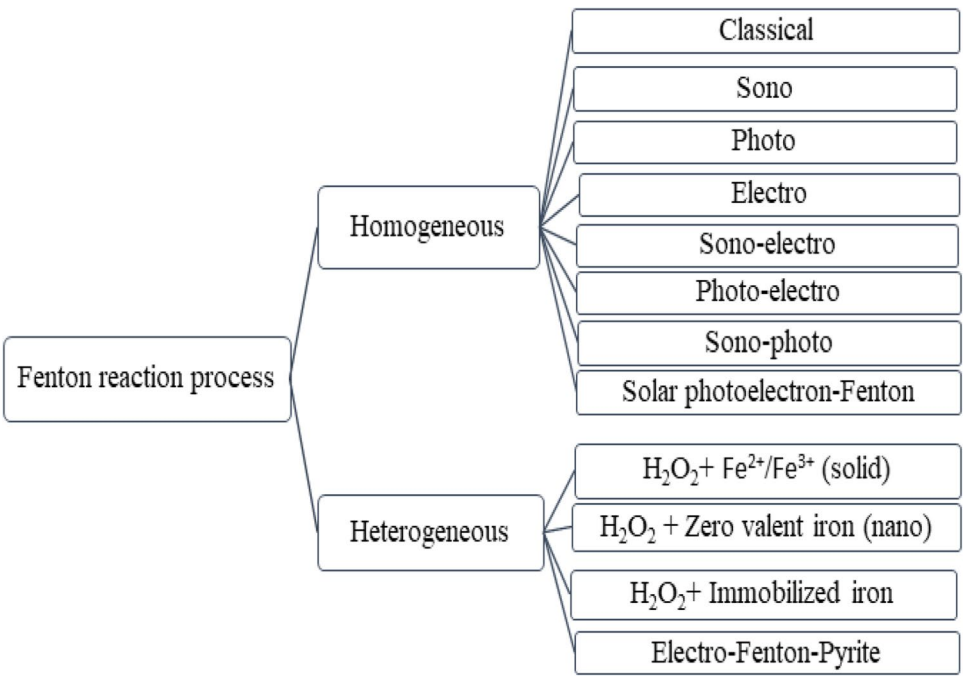

Fig. 1 Types of Fenton reaction processes used in treating organic pollutants in water

Table 1 Reactions involved in Fenton reaction processes

Fenton's reagent		
$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^{\cdot}OH + OH^-$		(F1)
$Fe^{2+} + H_2O + H^+ \rightarrow Fe^{3+} + H_2O + {}^{\cdot}OH$		(F2)
$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^{\cdot} + H^+$		(F3)
$Fe^{3+} + HO_2^{\cdot} \rightarrow Fe^{2+} + O_2 + H^+$	$k_4 = 2 \times 10^3\ M^{-1}s^{-1}$	(F4)
$Fe^{3+} + O_2^{\cdot-} \rightarrow Fe^{2+} + O_2$	$k_5 = 5 \times 10^7\ M^{-1}s^{-1}$	(F5)
$Fe^{2+} + H_2O_2 \rightleftharpoons FeOOH^+ + H^+$		(F6)
$FeOOH^+ + H^+ \rightleftharpoons Fe(H_2O_2)^{2+}$		(F7)
$Fe^{2+} + H_2O_2 \rightleftharpoons Fe(H_2O_2)^{2+}$		(F8)
$Fe(H_2O_2)^{2+} \rightarrow Fe^{3+} + {}^{\cdot}OH + OH^-$		(F9)
$Fe(H_2O_2)^{2+} \rightarrow FeO^{2+} + H_2O$		(F10)
Electro-Fenton processes		
$Fe^{3+} + e^- \rightarrow Fe^{2+}$		(EF1)
$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2$		(EF2)
$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$		(EF3)
$Fe \rightarrow Fe^{2+} + 2e^-$		(EF4)
Photo-Fenton processes		
$Fe^{3+} + h\nu + H_2O \rightarrow Fe^{2+} + {}^{\cdot}OH + H^+$		(PF1)
$FeOH^{2+} + h\nu \rightarrow Fe^{2+} + {}^{\cdot}OH$		(PF2)
$Fe^{3+}\text{-}L + h\nu \rightarrow Fe^{2+} + L^{\cdot+}$		(PF3)
Ultrasound Fenton processes		
$H^{\cdot} + Fe^{3+} \rightarrow Fe^{2+} + H^+$		(UF1)
$H^{\cdot} + O_2 \rightarrow HO_2^{\cdot}$		(UF2)
$HO_2^{\cdot} \rightleftharpoons O_2^{\cdot-} + H^+$		(UF3)
$Fe^{3+} + O_2^{\cdot-} \rightarrow Fe^{2+} + O_2$		(UF4)
$HO_2^{\cdot} + O_2^{\cdot-} + H^+ \rightarrow H_2O_2 + O_2$		(UF5)
Zero-valent-induced Fenton processes		
$2Fe^0 + O_2 + 4H^+ \rightarrow 2Fe^{2+} + 2H_2O$		(ZF1)
$Fe^{2+} + O_2 \rightarrow Fe^{3+} + O_2^{\cdot-}$		(ZF2)
$2O_2^{\cdot-} + 2H^+ \rightarrow H_2O_2 + O_2$		(ZF3)

medium, therefore presented by reaction F2. The optimum pH range for the Fenton process is approximately 2.8–3.0. The catalytic behavior of the redox couple Fe^{3+}/Fe^{2+} propagates the Fenton reaction process. For example, only a small amount of Fe^{2+} is needed because it can be regenerated by the reaction F3, called as Fenton-like reaction. The reaction F3 produces superoxide radical ($HO_2^{\cdot}$), which has lower oxidation power ($E^0 = 1.65$ V vs. NHE) than hydroxyl radical ($E^0 = 2.80$ V vs. NHE). The generation of Fe^{2+} from the reaction of Fe^{3+} with superoxide radical (reaction F4) is slower than the reaction F3. With increase in pH, the protonated superoxide species ($HO_2^{\cdot}$) converts to the deprotonated species ($O_2^{\cdot-}$) ($HO_2^{\cdot} = H^+ + O_2^{\cdot-}$; $pK_a = 4.8$) (Czapski and Bielski 1993; Von Sonntag and Schuchmann 1997). The reaction between Fe^{3+} with $O_2^{\cdot-}$ (reaction F5) is much faster than reaction F4 (see Table 1) (Bielski and Richter 1977; Gallard and De Laat 2000; Rush and Bielski 1985).

The mechanism of Fenton reaction is still not fully understood. The reaction F1 occurs through inner sphere electron transfer step (Fischbacher et al. 2017; Goldstein and Meyersteion 1999). In the first step, the formation of complex, $FeOOH^{2+}$, is formed (Gallard et al. 1999) (reaction F6). This complex goes through equilibrium (reaction F7) (Rachmilovich-Calis et al. 2009). Overall reaction of the complex formation is written as reaction F8. A general assumption is that the complex $Fe(H_2O_2)^{2+}$ could react through either one-electron or two-electron transfer forming ${}^{\cdot}OH$ or Fe^{IV}, respectively (reactions F9 and F10) (Bataineh et al. 2012; Hug and Leupin 2003; Katsoyiannis et al. 2008; Von Sonntag 2008).

Advantages of Fenton's reagent are that it is simple and easy to apply without any requirement of energy input (Bautista et al. 2008). However, Fenton's reagent has some drawbacks which include risk of storage of hydrogen peroxide, adjustment of pH to acidic range, and buildup of iron sludge (Oturan and Aaron 2014). Optimizing the dosages of reactants may minimize disadvantages of Fenton's reagent. Use of iron oxides, iron-modified clays, ion-exchange resins, iron-exchange Nafion membranes, and zeolites, and alumina may reduce the generation of sludge (Bautista et al. 2008; Lucas et al. 2007; Pignatello et al. 2006; Zhang et al. 2011a, b). Fenton's reagent has been applied to degrade and subsequent destruction and mineralization of numerous organic pollutants (Aljuboury et al. 2017; Annabi et al. 2016; Descorme 2017; Ganzenko et al. 2017; Li et al. 2016; Tayo et al. 2018; Usman et al. 2016). Examples include treatment of dyes, phenols, chlorophenols, chlorobenzenes, and antibiotics. Details are described in later sections of the review.

Electro-Fenton process

In the electro-Fenton process, Fe^{2+} and H_2O_2 are produced simultaneously by cathodic reduction of Fe^{3+} and O_2, respectively (reactions EF1 and EF2, Table 1) (Barhoumi et al. 2015, 2016; He and Zhou 2017; Lin et al. 2017a; Mousset et al. 2018; Mousset et al. 2016, 2018; Nidheesh et al. 2018; Sirés et al. 2014; Wang et al. 2016). A small amount of salt of Fe^{2+} (e.g., ferrous sulfate) is initially added, which can react with electrochemically produced H_2O_2 to produce Fe^{3+}. Reaction EF1 is critical to carry out recycling of Fe^{3+} to Fe^{2+}. Glassy carbon and graphite are rarely used as cathodes in electro-Fenton process. The most often used electro-Fenton's cathodes are carbon felt (Oturan et al. 2008) and GDE (gas diffusion electrode) cathodes (Brillas et al. 2009; Oturan and Aaron 2014). These are 3D cathodes. A platinum electrode is applied to perform the anodic reaction in the laboratory setup (reaction EF3, Table 1). In the peroxycoagulation, fered Fenton, anodic Fenton treatment or electrochemical peroxidation, Fe^{2+} can also be produced from the oxidation of sacrificial anode of iron (reaction EF4, Table 1) (Brillas et al. 2009). A few reviews on the

elimination of pollutants including micro-pollutants in water using electro-Fenton process have been published (Annabi et al. 2016; Brillas et al. 2009; Isarain-Chávez et al. 2011; Lin et al. 2017b; Steter et al. 2018; Wang et al. 2015; Özcan et al. 2016). A progress is being made to enhance the efficiency of electro-Fenton process. A research on sono-electro-Fenton and photo-Fenton processes has also been performed in order to improve efficiency and practicality of electro-Fenton methods (Bocos et al. 2016; Espinoza et al. 2016; Gozzi et al. 2017; Kalishwaralal et al. 2016; Oturan et al. 2008; Pliego et al. 2015; Uribe et al. 2015; Vidal et al. 2018). Electro-Fenton process is also being combined with biological methods to mineralize organic pollutants in water (Annabi et al. 2016; Ganzenko et al. 2017). The coupling of electro-Fenton with biological degradation is a new and interesting tool. The coupled process is called bioelectro-Fenton process (Olvera-Vargas et al. 2016a, b).

Photo-Fenton process

In the Photo-Fenton process, the ultraviolet (UV) light can assist the reduction of Fe^{3+} to Fe^{2+} (reaction PF1, Table 1) to react with H_2O_2 to generate $^{\cdot}OH$ through the reaction F1 (Table 1). Efficiency of reaction is greatest at pH 3.0 because Fe^{3+} ions generally exist as $Fe(OH)^{2+}$ under this condition. The photo-Fenton process has possibility of using many several UV regions as light energy source, namely UVA ($\lambda = 315$–400 nm), UVB ($\lambda = 285$–315 nm), and UVC ($\lambda < 285$ nm); therefore, the yield of $^{\cdot}OH$ varies with intensity of light. The $Fe(OH)^{2+}$ has absorbance maximum only in the UVB region. Production of $^{\cdot}OH$ through the UV photolysis of $Fe(OH)^{2+}$ is low (quantum yield of reaction PF2 is 0.2) (Pignatello et al. 2006; Zepp et al. 1992). Furthermore, solar light has fraction of light in the UVB region; only limited solar light irradiation can be absorbed. At the neutral pH, Fe^{3+} precipitated out and efficiency of photo-Fenton process is quite low. This drawback of the photo-Fenton process can be minimized by adding ligands (L) such as polycarboxylates and polyaminocarboxylates (e.g., oxalate, citrate, ethylenediaminetetraacetatic acid, and ethylenediamine *N*,*N*′-disuccinic acid) (Faust and Hoigné 1990; Faust and Zepp 1993; Li et al. 2012; Weller et al. 2013a, b). These ligands form stable complexes with Fe^{3+}, which upon UV and visible light irradiation reduce Fe^{3+} to Fe^{2+} via ligand-to-metal-charge transfer (LMCT) step (reaction PF3, Table 1). Quantum yields of these Fe^{3+}-L complexes are higher than quantum yield of $Fe(OH)^{2+}$. The use of organic ligand complexes of Fe^{3+} is advantageous. However, the ligands are attacked by $^{\cdot}OH$ produced in the process reducing the efficiency. Photo-Fenton process has shown their effectiveness in removing a wide range of contaminants, which include polychlorinated biphenyls, pesticides, and pharmaceuticals (Clarizia et al. 2017; Gligorovski et al. 2015; Matafonova and Batoev 2018; Serpone et al. 2017).

Ultrasound Fenton process

In the ultrasound Fenton process, a high-frequency is applied to split water into $^{\cdot}OH$ and $^{\cdot}H$ radicals (Eren 2012; Ma 2012; Salimi et al. 2017; Özdemir et al. 2011). Sonolysis of solution containing Fe^{3+} ions results in a series of reactions (reactions UF1–UF5, Table 1) to generate both Fe^{2+} and H_2O_2 for Fenton reaction (Gligorovski et al. 2015). Basically, sonochemistry and Fenton reaction generate $^{\cdot}OH$ to carry out transformation of organic pollutants in water (Chakma and Moholkar 2014, 2015). Several investigations have been explored applications of ultrasound Fenton processes to degrade a number of contaminants in water (Durán et al. 2013; Feng et al. 2013).

Heterogeneous Fenton reaction

A heterogeneous Fenton-like process has been investigated by many researchers because of advantages over homogeneous Fenton reactions (Cai et al. 2016; Diao et al. 2017; García-Rodríguez et al. 2017; Li et al. 2018; Lyu and Hu 2017; Velichkova et al. 2017). One of the main advantages of heterogeneous Fenton reactions is its feasibility over a wide pH rage to degrade pollutants in water. If the source of iron is magnetic like magnetite (Fe_3O_4), a magnetic separation can be applied (Morales-Pérez et al. 2016b). A focus of the research in the heterogeneous Fenton process is to increase the catalytic activity of solid iron sources without leaching of iron to aqueous environment. Efforts have been made to characterize catalysts for their pore size, density, and porosity in order to achieve better catalytic efficiency of heterogeneous Fenton-like reactions. A number of catalysts have been applied to carry out heterogeneous Fenton reactions (Costa et al. 2008; Diao et al. 2017, 2018; García-Rodríguez et al. 2017; Mirzaei et al. 2017; Morales-Pérez et al. 2016a; Nidheesh et al. 2017; Oturan et al. 2018; Ouiriemmi et al. 2017; Pi et al. 2018).

The $^{\cdot}OH$ radicals may also be produced in a reductive environment (Cao et al. 2013; Le et al. 2011; Vilardi et al. 2018). It has been shown that an addition of zero-valent iron (ZVI, Fe^0) to an aerated water solution yields the precursors reactants (Fe^{2+} and H_2O_2) through a sequence of reactions (ZF1–ZF3, Table 1) (Kang et al. 2017). In the presence of organic compounds and ZVI, many additional reactions also occur. This may be the reason of limited use of ZVI in Fenton reaction processes (Shimizu et al. 2012).

Iron oxides minerals like pyrite (FeS_2), hematite (Fe_2O_3), goethite (α-FeOOH), and lepidocrocite (γ-FeOOH) have

been studied in the heterogeneous Fenton-like reactions. Numerous support materials in heterogeneous Fenton processes have also been used. These include activated carbon, zeolites, clays, silicas, layered materials, and graphene oxide (Espinosa et al. 2016). More recently, a focus is on metal nanoparticles (e.g., Ag, Cu, and Au) to modify surfaces to obtain more effectiveness of the heterogeneous Fenton reaction processes (Dhakshinamoorthy et al. 2012; Espinosa et al. 2018). Details of a range of catalysts in Fenton reactions are given in reviews on the subject of degradation of several kinds of pollutants in water.

In following section of the review, examples are given to demonstrate applications of homogeneous and heterogeneous Fenton reaction processes.

Treatment of industrial wastewater: homogeneous Fenton reaction process

Effluents released from various industries contain pollutants at high levels, which could be a threat to human life. Removal of these pollutants by biological and eco-friendly methods is not successful as industrial waste contains mainly organic loads comprising of high COD and BOD. In developing countries, 85–90% of the wastewater is discharged directly into surface water bodies without proper treatment, and thus, the pollutants present in the discharged directly to the environment (Shannon et al. 2008). In India, nearly 6.2 million m^3 of industrial wastewater is generated every day, and only 60% of it is being treated (Kaur et al. 2012).

Chemical method is considered as a convenient strategy for removing these pollutants. Among various chemical processes, advanced oxidation process has found to be appropriate approach to minimize contamination from industrial effluents. Table 2 presents examples of Fenton treatment of industrial waste generated by various industries of the world. A wide range of chemicals have been found in industrial wastewater, which include pesticides and pharmaceuticals. Generally, industrial wastewater contains high values of chemical oxygen demand (COD), biological demand (BOD), dissolved organic carbon (DOC), and total organic carbon. Fenton's reagent was applied at various molar ratios of Fe(II) and H_2O_2 (Table 2). Pollutants of industrial wastewater could be degraded almost completely. Moreover, results showed a significant decrease in values of COD, BOD, and DOC after the treatment with Fenton's reagent. For example, at a molar ratio 10 for 0.2 mM Fe^{3+}, TOC removal was 90% in real industrial water (Bouafia-Chergui et al. 2010). This suggests that mineralization of organic pollutants (e.g., maleic acid anhydride, pesticides, 2-ethylhexyl alcohol, urea formaldehyde resin adhesive, α,β,γ-HCH, DDT, DMDT, fenitrothion, chlorfenvinphos) could be achieved using the Fenton's reagent. Values of DOC decreased significantly by the treatment with the Fenton's reagent. Components of petroleum waste like ethylene glycol, 1,4-dioxane, lower $[Fe(II)]/[H_2O_2]$ value could also be degraded at molar ratio of Fe(II) to H_2O_2 as 0.02.

Degradation of pollutants: homogeneous Fenton reaction processes

Degradation of representative phenols and pesticides by homogeneous Fenton reaction is given in Table 3. Phenol and dichlorvos were found to be successfully removed by homogeneous Fenton reaction in the acidic pH range (Table 3). Only 80% removal of 2,4-dichlorophenol was seen. Transformation of bisphenol A formed various intermediates, suggested not significant mineralization of parent molecule under studied conditions. Homogeneous Fenton reaction in combination with ultrasound was highly effectives in degrading different kind of pesticides. Removal of the pesticides was almost complete (Table 3).

Degradation of dyes: heterogeneous Fenton reaction processes

Nowadays, dyes are frequently used for several purposes such as dyeing clothes, leather, furniture, even in our regular life in food, cosmetics, and medicine, etc. Dyes are not easily degraded because some of them are non-biodegradable and have long-term adverse effect. Dyes have shown hazard effects on environment after entering into the ecosystem (Huang et al. 2009). Dyes can be degraded by various methods, including biomass degradation (Hsu et al. 2012; Prachi and Anushree 2009), photocatalytic degradation (Gu et al. 2014), combined treatment (Jafari et al. 2012). Among various advanced oxidation methods, Fenton reaction systems are reliable methods to transform dyes into many smaller fragments, i.e., water, carbon dioxide. Examples of degrading dyes by Heterogeneous Fenton reactions systems are summarized in Table 4. Catalysts used in the systems were iron- and carbon-based materials. Combinations of iron and carbon materials were also utilized to degrade dyes. Significantly, most of the studies shown in Table 4 under different conditions and catalysts could achieve the complete degradation of dyes. However, time of the complete degradation varied with experimental conditions. Nanocomposite materials in heterogeneous Fenton reaction systems seem to take less time to obtain the complete degradation compared to other catalysts.

Table 2 Treatment of real industrial wastewater by homogeneous Fenton reaction process. COD—chemical oxygen demand, BOD—biological oxygen demand, DOC—dissolved organic carbon, TOC—total oxygen carbon

No.	Region	Pollutants	Reaction conditions and comments	References
1.	Tambla Tributary (River Damodar), India	Industrial waste	[Fe(II)] = 6 g/L and $[H_2O_2]$ = 220 g/L COD removal = ~ 95%	Mandal et al. (2010)
2.	Chemical factories, Southern Poland	Maleic acid anhydride, pesticides, 2-ethylhexyl alcohol, urea formaldehyde resin adhesive	$[Fe(II)]/[H_2O_2]$ = 0.33, (maleic acid anhydride), pH = 3.0, COD removal = ~ 88% $[Fe(II)]/[H_2O_2]$ = 0.50, (2-ethylhexyl alcohol), pH = 3.5, COD removal = 86.3% $[Fe(II)]/[H_2O_2]$ = 0.33, (urea formaldehyde resin adhesive), pH = 3.5, COD removal = 88.8%	Barbusinski (2005)
3.	Pesticide-containing wastewater, Southern Poland	α,β,γ-HCH, DDT, DMDT, fenitrothion, chlorfenvinphos	$[Fe(II)]/[H_2O_2]$ = 0.33, (pesticide containing water), pH = 3.2, COD removal = 71.7% All pesticides degraded completely	Barbusinski and Filipek (2001)
4.	Pharmaceutical waste water, Turkey	Variety of pharmaceutical chemicals	$[H_2O_2]$ = 5 g/L and $[Fe(II)]/[H_2O_2]$ = 0.33–0.50 Fenton oxidation followed by sequencing batch reactor, COD removal = ~ 99%	Tekin et al. (2006)
5.	Laboratory mixed waste chemical of 17-month period, Brazil	Different laboratory chemicals waste	$[Fe(II)]/[H_2O_2]$ = 0.22, COD removal = 90%	Benatti and Tavares (2012)
6.	Fish canning waste water, Portugal	Organic matter, salts, oil and grease	Before treatment: DOC = 50 mg/L, COD = 220 mg/L, BOD_5 = 0.8 mg/L (Biological pretreatment of fish canning waste water, followed by treatment with Fenton's reagent) After treatment: DOC = 20 mg/L, COD = 90 mg/L	Cristovao et al. (2014)
7.	Chemical plant that produce acrylic sheets, Mexico	Methyl methacrylate	Maximum removal efficiencies (Fenton adsorption treatment): 96% color, 58% TOC, and 60% COD	Sanchez et al. (2014)
8.	Real industrial biorecalcitrant wastewater, Spain	5-Amino-6-methyl-2-benzimidazolone	$[Fe(II)]/[H_2O_2]$ = 0.0032 Removal: 67% color, 42% COD, and 41% TOC	Sarria et al. (2001)
9.	El-Nasr pharmaceutical and Chemical Company, South-East of Cairo	Pharmaceutical company discharges both industrial and municipal wastewater	Before treatment: COD (4100–13,023 mg/L), TSS (20–330 mg/L), and oil grease (17.4–600 mg/L) Treatment: $[Fe(II)]/[H_2O_2]$ = 0.02 and $COD/[H_2O_2]$ = 1:2.2 Removal was almost complete	Badawy et al. (2009)
10.	Tannery wastewater, Brazil	Containing both organic and inorganic pollutants	H_2O_2/UV at pH 3 and Fenton at pH 3.5; efficiently remove TOC to low level	Schrank et al. (2005)
11.	Petrochemical effluent, India	Ethylene glycol, 1,4-dioxane	$[Fe(II)]/[H_2O_2]$ = 0.02; pH 3.0 COD removal = 97.5%	Ghosh et al. (2011)
12.	Pesticides wastewater from Nubaria, Egypt	Chlorpyrifos, lambda-cyhalothrin, diazinon	COD removal (photo-Fenton process) = 90.7% COD removal (TiO_2 photocatalytic reaction = 79.6%	Alalm et al. (2015)

Table 2 (continued)

No.	Region	Pollutants	Reaction conditions and comments	References
13.	Wastewater obtained from civilian explosive industry, South-west, China	Dinitrodiazophenol (DDNP)	Treatment: combined Fe^0/air and Fenton process COD removal = 78% Chromaticity removal = 98% chromaticity	Yuan et al. (2016)
14.	Beverage industrial effluent, Spain	Different complex compound	Photo-Fenton process): 53% mineralization (2 h) Photo-Fenton/persulfate: 76% mineralization (4 h)	Exposito et al. (2016)
15.	Winery wastewater, Cyprus	Polyphenols, tannins, and lignins	(a) Fe(II)] = 10 mg/L, $[H_2O_2]$ = 100 mg/L COD removal = 35% and DOC removal = 26% (120 min) (b) $[Fe_{SBA\text{-}15}]_0$ = 100 mg/L, $[H_2O_2]$ = 100 mg/L COD removal = 48% and DOC removal = 48% in 180 min	Loannou et al. (2013)

Table 3 Examples of degradation of phenols, pesticides, and surfactants in water by the homogeneous Fenton reaction

No.	Pollutants	Reaction conditions	Results and comments	References
1.	Phenol	$[H_2O_2]$ = 300–600 mg/L, Fe(II) = 10 mg/L; pH = 3–3.5, room temperature; reaction time = 6 h	Degradation = 100% Mineralization = 60%	Yalfani et al. (2009)
2.	2,4-dichlorophenol	[2,4-Dichlorophenol] = 200 mg/L, $[H_2O_2]$ = 300–580 mg/L, [Fe(II)] = 10–20 mg/L; pH = 2.5–7.0	Removal = ~ 70% within 2 h	Ranjit et al. (2008)
3.	Bisphenol A (BPA)	[BPA] = 10 µg/L, pH = 3, [Fe(II)]/$[H_2O_2]$ = 10	Degradation resulted in various intermediates, benzenediols, monohydroxylated BPA with molecular weight ranged from 94 to 500 Da	Poerscchmann et al. (2010)
4.	Dichlorvos or 2,2-dichlorovinyl dimethyl phosphate	$[H_2O_2]$ = 15 mg/L, [Fe(II)]/$[H_2O_2]$ = 3.0, pH = 3.0, room temperature; ultrasonic probe frequency = 20 kHz	Degradation = 100%	Golash and Gogate (2012)
5.	Methyl parathion (Phosphate pesticides)	[Methyl parathion] = 20 mg/L, $[H_2O_2]$ = 200 mg/L; [Fe(II)]/$[H_2O_2]$ = 3.0	Degradation (ultrasonic horn process) = ~ 98.5% TOC removal = 73.7% Degradation (ultrasonic bath process) = ~ 96.5% TOC removal (ultrasonic bath process) = 75%	Shriwas and Gogate (2011)
6.	Carbofuran (carbamate pesticides)	[Carbofuran] = 20 mg/L, $[H_2O_2]$ = 100 mg/L,[Fe(II)] = 20 mg/L	Degradation = ~ 99% Mineralization = 46% after 30 min	Ma et al. (2010)

Treatment of antibiotics

Human and veterinary antibiotics are considered to be of prime importance as emerging micro-pollutants due to their high consumption rate. These micro-pollutants are being generated through household, industry, hospital service, poultry, livestock, and aquatic activity which get deposited and leached into the environment. The fate of antibiotics after their purposive use is not being monitored. Most of the antibiotics are not fully eliminated from the body, and some of them may remain unchanged in the environment (Hirsch et al. 1999; Brown et al. 2006). Thus, in order to bring awareness among the people, world antibiotics week is being organized since November 2015, with the theme "antibiotics: Handle with care." Antibiotics

Table 4 Examples of degradation of dyes by heterogeneous Fenton reaction processes

No.	Dyes	Catalyst	Reaction conditions	Results	References
1.	Acid black 1	Pillared laponite clay-based Fe	[Dye] = 0.2 mM; [Catalyst] = 1 g/L, $[H_2O_2]$ = 6.4 mM, Light intensity = 8 W UVC	Removal = 100% in 60 min	Sum et al. (2004)
2.	Acid blue 185	Natural and ball-milled magnetite nanostructures	[Dye] = 20–120 mg/L [Catalyst] = 1.5 g/L, pH = 3.0	Removal = 80–99% 6-h ball-milled magnetite showed highest efficiency	Acisli et al. (2017)
3.	Acid blue 74	Fe-ZSM5 zeolite	[Dye] = 8.56×10^{-5} mol/L, $[H_2O_2]$ = 21.2 mmol/L, [Catalyst] = 0.5 g/L, pH 5,0, UV irradiation	Removal = 100% in 120 min	Kasiri et al. (2008)
4.	Acid blue 92	Natural martite prepared by ball milling	[Dye] = 10 mg/L [Catalyst] = 2.5 g/L pH = 7.0, 2.5 g/L Ultrasonic power = 150 W	Removal = 100% in 30 min achieved	Dindarsafa et al. (2017)
5.	Acid orange 7	Graphene oxide-iron oxide nanocomposites	[Dye] = 35 mg/L [Catalyst] = 0.2 g/L $[H_2O_2]$ = 2 mM pH = 3.0	Removal = 80% in 20 min Removal = 98% in 180 min	Zubir et al. (2014a, b)
6.	Acid orange 7	Graphene oxide-iron oxide nanocomposites	[Dye] = 0.1 mM [Catalyst] = 0.2 g/L $[H_2O_2]$ = 22 mM pH = 3.0	Removal = 96% on 90 min	Zubir et al. (2014a, b)
7.	Anthraquinone dye	Pyrite nanorods synthesize by oxygen and nitrogen nonthermal plasma	[Dye] = 20 mg/L [Catalyst] = 0.6 g/L pH = 5.0 Ultrasonic power = 300 W	Removal = 100% in 40 min	Khataee et al. (2016)
8.	Brilliant orange X-GN	Iron-pillared montmorillonitic via pillaring	[Dye] = 100 mg/L [Catalyst] = 0.6 g/L catalyst, $[H_2O_2]$ = 4.9 mmol/L H_2O_2 are pH = 3.0	Removal = 98.6% in 140 min Under UV light Removal = 80% in 140 min under visible light	Chen et al. (2009)
9.	1-Diazo-2-naphthol-4-sulfonic acid dye	Mesoporous carbon–Fe	[Dye] = 250 mg/L [Catalyst] = 0.5 g/L pH = 5.0	Removal = 94% in 120 min	Gu et al. (2013)
10.	Methylene blue	Fe_3O_4–MWCNT	[Dye] = 10 mg/L [Catalyst] = 0.3 g/L $[H_2O_2]$ = 0.4 M pH 5.5	Removal = 97% in 720 min	Wang et al. (2014)
11.	Orange II	Fe/ZSM-5 zeolite	[Dye] = 0.1 mM [Catalyst] = 200 mg/L $[H_2O_2]$ = 6 mM Temperature = 53 °C pH = 5.2	Removal = 100% in 240 min	Duarte et al. (2009)
12.	Orange II	Transition metal on carbon aerogels	[Dye] = 0.1 mM [Catalyst] = 0.2 g/L $[H_2O_2]$ = 6.0 mM pH = 3.0	Removal = 100% in 180 min	Duarte et al. (2009)
13.	Orange II	a. Nanocomposites Fe supported on laponite clay b. Nanocomposites Fe supported on bentonite clay	[Dye] = 0.2 mM [Catalyst] = 1.0 g/L $[H_2O_2]$ = 10 mM pH = 2.10–6.60	Removal = 100% in 60 min	Feng et al. (2006)
14.	Orange II	Plasma-synthesized hematite and goethite	[Dye] = 25 mg/L [Catalyst] = 0.2 g/L $[H_2O_2]$ = 5.0 mM pH = 3	Removal = 100% by hematite Removal = 78% by goethite	Djowe et al. (2014)

Table 4 (continued)

No.	Dyes	Catalyst	Reaction conditions	Results	References
15.	Reactive brilliant red X-3B	Fe-pillared bentonite	[Dye] = 10^{-4} mol/L [Catalyst] = 0.5 g/L [H_2O_2] = 10^{-2} mol/L pH = 3.0	Removal > 98% in 100 min	Li et al. (2006)
16.	Rhodamine B	Rice hull-based silica-supported iron catalyst	[Dye] = 5.0 mg/L [Catalyst] = 1.0 g/L [H_2O_2] = 0.98 mmol pH = 5.0	Removal = 99% in 120 min	Gan and Li (2013)
17.	Rhodamine B	Cationic cyclopentadienyl iron silica	[Dye] = 25 mg/L [Catalyst] = 5.0 g/L [H_2O_2] = 1 M pH = 3.0	Removal = 99% in 30 min	Chen et al. (2013)

and antibiotics resistance bacteria present in water have become problems of global reach (Walter and Vennes 1985).

Various oxidation methods are nowadays available to remove antibiotics from environment (Watkinson et al. 2007; Adams et al. 2002; Oturan et al. 2013; Sharma 2013; Feng et al. 2016; Anquandah et al. 2011; Gulkowska et al. 2008). Advanced oxidation process have gained much higher attention in comparison to other oxidation methods (An et al. 2010; Elmolla and Chaudhuri 2010a, b; Alaton and Dogruel 2004; Alaton et al. 2004; Kassinos et al. 2011; Zhang et al. 2006). Examples of degradation of antibiotics by different reaction processes are presented in Table 5.

Removal of amoxicillin by a mixture of Fe(II) and H_2O_2 could be observed up 100% in ≤ 90 min when the antibiotic concentration was in the range from 10 to 500 mg/L. When the level of amoxicillin was in 1 g/L, removal of the antibiotic was in hours. Degradation of amoxicillin also resulted in a decrease in TOC, generally more than 50%. When photo-Fenton reaction using potassium ferrioxalate was applied, a decrease in toxicity of amoxicillin was seen (Table 5). Comparatively, degradation of cefradine was much less than amoxicillin under same conditions of homogeneous Fenton reaction system. Similar to amoxicillin, degradation of ampicillin and cloxacillin was up to 100%, which was also in conjunction with removal of COD and TOC. When homogeneous Fenton reaction process was applied to antibiotic fermentation wastewater, significant removal of COD was observed in 60 min (i.e., 72.4%).

Complete removal of ciprofloxacin by Fenton's reagent was observed in less than 2 min at an initial concentration of the antibiotic as 10 mg/L (Table 5). Degradation of chloramphenicol under heterogeneous Fenton system using FeS_2–H_2O_2 at pH 8.0 was more than 80%. Chlortetracycline could be removed by homogeneous Fenton reaction processes in which a combination of Fenton with sonication gave removal of 82%, a slightly higher than either Fenton reaction or sonication processes (67–76%). Removal of fluoroquinolone was low (i.e., 40%) in use of Fenton's reagent. However, degradation of flumequine increased to 94% under photo-Fenton reaction conditions. Removal of levofloxacin was sought by electro-Fenton reaction process that showed a complete elimination of the antibiotic, and a decrease in TOC was more than 50%. Other fluoroquinolones, moxifloxacin, norfloxacin, and ofloxacin also had the complete removal by the Fenton reaction systems.

Degradation of sulfonamide antibiotics (e.g., sulfamethoxazole, sulfamethazine, sulfanilamide, and sulfasalazine, and sulfamonomethoxine) and trimethoprim has been investigated by Fenton reaction processes. Almost complete elimination of sulfonamides was observed (Table 5). Mineralizations of sulfamethoxazole and trimethoprim were also high (70–90%). When Fenton's reagent was applied to degrade antibiotics in swine wastewater, removal of TOC was 40%. Several studies have been performed on degrading tetracycline by Fenton reaction processes (Table 5). In an hour or less, removal of tetracyclines was in the range of 79–100%, depending on the reaction conditions. In a longer time of 2 h, mineralization up to ~ 4% could be achieved (Table 5).

Conclusion

In the homogeneous Fenton reaction, the conditions of should be optimized in order to evaluate full potential of ·OH radicals to oxidize contaminants in water. These conditions include dosages of Fe(II) and hydrogen peroxide, pH, and temperature. Excess concentrations of Fe(II) precipitate out as ferric hydroxide, and COD value increases by excess amount of hydrogen peroxide. Homogeneous Fenton reaction is limited to acidic pH that results in unwanted sludge of iron-oxy hydroxides. Heterogeneous Fenton reaction systems are being developed by applying catalysts (e.g., ferric oxides) and photolysis to enhance the effectiveness to eliminate contaminants in water. However, a more research is needed on advancing application of catalysts which are

Table 5 Treatment of antibiotics containing wastewater by different Fenton reaction processes

S. no.	Antibiotics	Reaction conditions	Results	References
1.	Amoxicillin	Box-Behnken-statistical experiment design [Amoxicillin] = 10–200 mg/L [H_2O_2] = 10–500 mg/L [Fe(II)] = 0–50 mg/L Fe(II)	Removal = 100% degradation in 2.5 min Mineralization = 37% in 15 min	Ay and Fikret (2010)
2.	Amoxicillin	Central composite factorial design method [Amoxicillin] = 450 µg/L [H_2O_2] = 3.50–4.28 mg/L [Fe(II)] = 254–350 µg/L pH = 3.5, temperature = 20–30 °C	Removal = 100% in 30 min	Homem et al. (2010)
3.	Amoxicillin	[Amoxicillin] = 10 mg/L [Potassium ferroxalate] = 0.1 g/L pH 6.0–8.0 UV light irradiation	TOC removal = 53% in 90 min Decrease in toxicity to 65% to 5%	Trovo et al. (2011)
4.	Ampicillin	[Ampicillin] = 20 mg/L [H_2O_2] = 400 µmol/L [Fe(II)] = 87 µmol/L Fe(II) pH = 3.5	Removal = 100% in 3 min by photo-Fenton Removal = 100% in 10 min by Fenton process	Rozas et al. (2010)
5.	Amoxicillin, cefradine	[Amoxicillin] = 1 g/L [H_2O_2] = 166.5 mg/L [Fe(II)] = 166.5 mg/L pH = 3.0	Removal (amoxicillin) = 97.4% in 48 h Removal (cefradine) = 22.5% in 48 h	Li et al. (2015)
6.	Amoxicillin, cloxacillin	[Amoxicillin] = 150 mg/L [Cloxacillin] = 150 mg/L H_2O_2:COD = 2 Fe(II):H_2O_2 = 76at pH = 3	COD removal = 79% TOC removal = 73% Ammonia–nitrogen removal = 81.9% in 90 min	Affam and Chaudhuri (2013)
7.	Amoxicillin, cloxacillin	[Amoxicillin] = 150 mg/L [Cloxacillin] = 150 mg/L [H_2O_2]:[COD] = 2.0, FeGAC = 3.5 g/L	COD removal = 87.5% TOC removal = 78.0% NH, -N removal = 98.2% in 90 min, pH 3.0	Augstin et al. (2014)
8.	Amoxicillin, cloxacillin	[Amoxicillin] = 138 mg/L, [Cloxacillin] = 84 mg/L [H_2O_2]/[COD] = 2.5, [Fe(II)]:[H_2O_2] = 2.0	COD removal = 89% Degradation = 100% in 1 min	Elmolla and Chaudhuri (2012)
9.	Azithromycin, clarithromycin	[Azithromycin]/[Clarithromycin] = 200 mg/L [Fe(II)] = 0.45 mmol/L, [Fe^0] = 0.3 mmol/L [H_2O_2] = 0.16/0.3 mmol/L, pH = 7.0	COD removal (azithromycin) = 83% COD removal (clarithromycin) = 76% Removal (azithromycin) = 90% Removal (clarithromycin) = 76% in 1 h	Mohammadi et al. (2013)
10.	Amoxicillin, ampicillin, cloxacillin	[Amoxicillin] = 103 mg/L [Ampicillin] = 104 mg/L [Cloxacillin] = 105 mg/L [Zinc oxide] = 0.5 g/L pH = 11.0	Degradation = 100% COD removal = 23.9% TOC removal = 9.7% in 180 min	Elmolla et al. (2010)
11.	Amoxicillin, ampicillin, cloxacillin	[Amoxicillin] = 104 mg/L [Ampicillin] = 105 mg/L [Cloxacillin] = 103 mg/L [COD]/[H_2O_2]/[Fe(II)] = 1:3:0.3 pH = 3.0	Degradation = 100% in 2 min COD removal = 81.4% DOC removal = 54.3% in 60 min BOD/COD ratio improvement = 10 min (amoxicillin), 20 min (ampicillin), 40 min (cloxacillin)	Elmolla and Chaudhuri (2009a)

Table 5 (continued)

S. no.	Antibiotics	Reaction conditions	Results	References
12.	Amoxicillin, ampicillin, cloxacillin	[Amoxicillin] = 104 mg/L [Ampicillin] = 105 mg/L [Cloxacillin] = 103 mg/L $[H_2O_2]$:[COD] = 1.5 [Fe(II)]:$[H_2O_2]$ = 20 pH = 3.0	Degradation = 100% in 2 min BOD/COD ratio improved = 0 to 0.4 COD removal = 80.8% DOC = 58.4% in 50 min	Elmolla and Chaudhuri (2009b)
14.	Amoxicillin, ampicillin, cloxacillin	[Amoxicillin] = 100 mg/L [Ampicillin] = 220 mg/L [Cloxacillin] = 500 mg/L $[H_2O_2]$:[COD] = 3.0 pH = 3.0	COD removal (all three antibiotics) = ~ 80%	Elmolla et al. (2010)
15.	Antibiotic fermentation wastewater	COD = 3279 mg/L TOC = 1296.3 mg/L Color = 3000 $[H_2O_2]$ = 150 mg/L [Fe(II)] = 120 mg/L pH = 4 .0	Color removal = 66.6% COD removal = 72.4% after 60 min	Xing and Sun (2009)
17.	Ciprofloxacin	[Ciprofloxacin] = 10 mg/L $[H_2O_2]$ = 2.5 mM [Fe(II)] = 2.0 mg/L pH = 2.8	Degradation = 80% in 1.8 min	Lima et al. (2014)
18.	Cefalexin	[Cefalexin] = 200 mg/L [Fe(II)] = 1 mM pH = 3.0 Current density = 6.66 mA/cm^2	TOC removal = 49% Mineralization = 100%	Estrada et al. (2012)
19.	Chloramphenicol	[Chloramphenicol] = 50 mg/L [GLDA] = 100 µmol/L $[FeS_2]$ = 100 mg/L $[H_2O_2]$ = 1 mmol/L pH = 8.0	Degradation = 83.3%	Wu et al. (2016)
20.	Chlortetracycline	[Sludge] = 34 g/L [Fe(II)] = 30 mg/L $[H_2O_2]$ = 30 mg/L pH = 3.0	Removal (ultrasonication process) = 67% Removal (fenton oxidation process) = 76% Removal (ferro-sonication) = 82%	Pulicharla et al. (2017)
21.	Ciprofloxacin, sulphamethoxazole, trimethoprim	[Ciprofloxacin]/[sulphamethoxazole]/[Trimethoprim] = 4.24×10^{-2} $mg_c m^2 W^{-2} L^{-1}$ $[H_2O_2]$ = 2.5 mM pH = 2.8	Removal = ~ 60% removal after 240 min	Lima et al. (2017)
22.	Clarithromycin, sulfamethoxazole	[Clarithromycin]/[sulfamethoxazole] = 100 ppb $[H_2O_2]$ = 50 mg/L [Fe(III)] = 5 mg/L pH = 4.0	Removal (clarithromycin) = 70% Removal (sulfamethoxazole) = 95%	Karaolia et al. (2014)
23.	Chloramphenicol, ciprofloxacin, dipyrone	[Chloramphenicol]/[ciprofloxacin]/[dipyrone] = 0.15 mM $[H_2O_2]$ = 22.5 mM [Fe(II)] = 2.25 mM pH = 3.5	Removal (chloramphenicol) = 52% Removal (ciprofloxacin) = 42% Removal (dipyrone) = 47% in 45 min	Giri and Golder (2015)
24.	Enoxacin	[Enoxacin] = 50 mg/L [Fe(II)] = 0.2 mmol/L Current intensity = 300 mA	Residual yields = 54% and 43% after 120 min Fluorine = 77%, nitrate = 5%, ammonia = 24% after 180 min	Annabi et al. (2016)
25.	Flumequine	[Flumequine] = 500 µg/L [Fe(II)] = 0.5 mmol/L $[H_2O_2]$ = 2.0 mmol/L	Degradation (Fenton) = 40% Degradation (photo-Fenton) = 94% after 60 min	Silva et al. (2013)

Table 5 (continued)

S. no.	Antibiotics	Reaction conditions	Results	References
26.	Levofloxacin	[Levofloxacin] = 200 mg/L [Na_2SO_4] = 0.05 M [Fe(II)] = 1 mM pH = 3.0 Current intensity = 6.67 mA cm^{-2}	Removal = 100% in 120 min TOC removal = 60% after 360 min	Gong et al. (2016)
27.	Levofloxacin	[Levofloxacin] = 0.23 mM [Na_2SO_4] = 0.05 M [Fe(II)] = 0.2 mM pH = 3.0 Current intensity = 300 mA	Mineralization = 100% TOC = 95% for 8 h	Barhoumi et al. (2015)
28.	Moxifloxacin	[Moxifloxacin] = 0.15 mM [Fe(II)] = 0.5 mM pH = 3.0	Several intermediates were formed for the degradation of moxifloxacin, which was confirmed by LC–MS analysis	Yahya et al. (2017)
29.	Norfloxacin	[Norfloxacin] = 100 mg/L [Fe_0] = 100 mg/L [H_2O_2] = 10 mmol/L pH = 4.0	Degradation = 95% within 40 min Mineralization = 50%	Zhang et al. (2017)
30.	Norfloxacin	[Norfloxacin] = 0. 25 mM [Na_2SO_4] = 0.05 M [Fe(III)] = 0.1 mM pH = 3.0 Current intensity = 60 mA	Mineralization = 97.7% after 5 h	Özcan et al. (2016)
31.	Norfloxacin	[Norfloxacin] = 15 mg/L [H_2O_2] = 2.1 mmol/L pH = 7.0 (UV/H_2O_2)	Degradation = 100% degradation Mineralization = 55% mineralization	Santos et al. (2015)
32.	Ofloxacin	[Ofloxacin] = 10 mg/L [Fe(II)] = 2 mg/L [H_2O_2] = 2.5 mg/L	Degradation = 100%	Michael et al. (2013)
33.	Oxacillin	[Oxacillin] = 203 µmol/L [Fe(II)] = 90 µmol/L [H_2O_2] = 10 µmol/L	Mineralization = 100% after 5 min	Giraldo-Aguirre et al. (2017)
34.	Ofloxacin, trimethoprim	[Ofloxacin]/[trimethoprim] = 100 µg/L [Fe(II)] = 5 mg/L [H_2O_2] = 75 mg/L in solar light	Removal = 100%	Michael et al. (2012)
35.	Sulfasalazine	[Sulfasalazine] = 100 mg/L [Fe(II)] = 0.20 mM [H_2O_2] = 16 mM	Removal = ~ 99.5% TOC removal = 82.4% COD removal = 41% in 60 min	Fan et al. (2011)
36.	Sulfamethoxazole	[Sulfamethoxazole] = 200 mg/L [Fe(II)] = 1 mM pH = 3.0	TOC removal = 80% (photo-electro-Fenton process) in 6 h Mineralization = 63% (electro-Fenton process) TOC removal = 25% (electro-Fenton process)	Wang et al. (2011)
37.	Sulfamethoxazole	[Sulfamethoxazole] = 50 mg/L [Fe(II) = 2.6 mg/L [H_2O_2] = 120 mg/L	Toxicity reduction = 80% to 20%	Trovo et al. (2009)
38.	Sulfamethoxazole	[Sulfamethoxazole] = 200 mg/L [Fe(II)] = 10 mg/L [H_2O_2] = 300 mg/L pH = 2.8	Removal = 100%	Gonzalez et al. (2007)
39.	Sulfamethazine	[Sulfamethazine] = 50 mg/L [Fe(II)] = 40 mg/L [H_2O_2] = 600 mg/L	Degradation = 100% in 2 min	Moya et al. (2010)

Table 5 (continued)

S. no.	Antibiotics	Reaction conditions	Results	References
40.	Sulfamethazine	[Sulfamethazine] = 20 mg/L [Fe(II)] = 3.5–28 mg/L [H_2O_2] = 10–400 mg/L Current intensity = 2mWCm^{-2}	Mineralization = 70% after 120 min	Kitsiou et al. (2014)
41.	Sulfamethazine	[Sulfamethazine] = 20 mg/L [Ce–Fe-graphene] = 0.5 g/L [H_2O_2] = 8 mM pH = 7.0	Degradation = 99% TOC removal = 73%	Wan et al. (2016)
42.	Sulfanilamide	[Sulfanilamide] = 0.6 mM [Na_2SO_4] = 0.05 M [Fe(II)] = 0.20 mM pH = 3.0 Current intensity = 300 mA	Mineralization = 100%	Ghenymy et al. (2014)
43.	Sulfamonomethoxine	[Sulfamonomethoxine] = 4.53 mg/L [H_2O_2] = 0.49 mmol/L [Fe(II)] = 19.51 µmol/L pH = 4.0	Degradation = 98.5% COD removal = 99% after 120 min	Hui et al. (2012)
44.	Sulfonamide	[Sulfonamide] = 0.19 mM Fe(II):H_2O_2 = 1.5 pH = 2.5	Degradation = 99.9% COD removal 64.7–70.7% in 15 min	Dehghani et al. (2013)
45.	Sulfamethoxazole, trimethoprim	[Sulfamethoxazole]/[trimethoprim] = 20 mg/L [H_2O_2] (sulfamethoxazole) = 2.6 mM [H_2O_2] (trimethoprim) = 3.0 mM [Fe(III)] = 0.5 mg/L pH = 5.0	Mineralization (sulfamethoxazole) = 70% Mineralization (trimethoprim) = 90%	Dias et al. (2014)
46.	Sulfamethoxazole, erythromycin, clarithromycin	[Substrate] = 100 µg/L [Fe(III)] = 5 mg/L [H_2O_2] = 50 mg/L pH = 4.0	Removal = 100%	Karaolia et al. (2017)
47.	Swine wastewater (5 sulfonamide + 1 macrolide	[Antibiotics] = 1 mg/L [Fe(II)] = 0.91 mmol/L, [H_2O_2] = 1.37 mmol/L [Fe(II)]:[H_2O_2] = 0.66, pH = 5.0	TOC removal = 40% As removal = 78%, Cu removal = 36% Pb removal = 18%, Toxicity removal = 25%	Ben et al. (2009)
48.	Tetracycline	[Tetracycline] = 20 mg/L [Fe_3O_4 C] = 0.15 g/L, [H_2O_2] = 3 mM pH = 3.0	Removal = 79% in 44 min Mineralization = 43.7% in 120 min	Kakavandi et al. (2016)
49.	Tetracycline	[Tetracycline] = 100 mg/L [Fe(II)] = 1.0 g/L, [H_2O_2] = 150 mmol/L pH = 3.7	Removal = 93.6% in 60 min Mineralization = 31.8% after 60 min	Hou et al. (2016)
50.	Tetracycline	[Tetracycline] = 40 mg/L [Fe_3O_4 void TiO_2] = 0.25 g/L [H_2O_2] = 0.377 M, pH = 3.0, 9.0	Degradation = 100% at pH 3.0 Degradation = 75% at pH 9 within 6 min	Du et al. (2017)
51	Tetracycline	[Tetracycline] = 40 mg/L [Fe(II)] = 5 mg/L, [H_2O_2] = 71.5 mg/L	Mineralization = 100%	Turbay et al. (2013)

applicable in a wide range of pH and can recycle iron in the Fenton reaction system.

Fenton oxidation process can efficiently remove a range of contaminants in water. For example, antibiotics such as ofloxacin, trimethoprim, sulfonamide and sulfamethoxazole could be completely using the Fenton reaction system. However, Fenton reactions process needs to be combined with other methods to achieve thorough mineralization. Significantly, most of the studies in the literature on removing contaminants by Fenton reaction processes are on the bench scale, and a very little is known on performing at a pilot scale with polluted water. It is important Fenton reaction system should be demonstrated by using contaminated water containing nitrate, bromide, and natural organic matter. These constituents of water scavenge $^{\cdot}OH$ radicals and thus decrease the effectiveness of the advanced oxidation processes. Furthermore, pilot and field scale testing of the selected Fenton reaction is required to demonstrate the capabilities, possible limitation, and reaction conditions of Fenton reaction processes to produce drinking water from contaminated source water.

Acknowledgements Dr. Bhawana Jain (postdoctoral fellow, No. F.15-1/2013-14/PDFWM-2013-14-GE-CHH-18784 (SA-II)) is thankful to UGC, Delhi, India, for Research Project grants. Professor Hyunook Kim appreciates the financial support by Korea Ministry of Environment (MOE) (Project ID: 2015001790002). We thank reviewers for their comments, which improved this paper greatly.

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