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Review

Combination of Fenton processes and biotreatment for wastewater treatment and soil remediation

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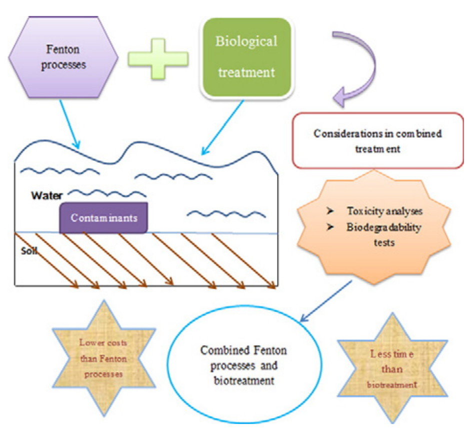
HIGHLIGHTS

- The combination of Fenton process and biotreatment is novel and useful.
- Toxicity and biodegradability tests are significant to design a combined system.
- No matter which technology at first stage, they would be called combined methods.
- Wastewater contains PPCPs or EDCs.
- Use of combination system in wastewater and polluted soil.

GRAPHICAL ABSTRACT



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ABSTRACT

There is a continuously increasing worldwide concern for the development of wastewater and contaminated soil treatment technologies. Fenton processes and biological treatments have long been used as common technologies for treating wastewater and polluted soil but they still need to be modified because of some defects (high costs of Fenton process and long remediation time of biotreatments). This work first briefly introduced the Fenton technology and biotreatment, and then discussed the main considerations in the construction of a combined system. This review shows a critical overview of recent researches combining Fenton processes (as pre-treatment or post-treatment) with bioremediation for treatment of wastewater or polluted soil. We concluded that the combined treatment can be regarded as a novel and competitive technology. Furthermore, the outlook

Abbreviations: AOPs, advanced oxidation processes; BaP, benzo[a]pyrene; BOD, biochemical oxygen demand; COD, chemical oxygen demand; DAF, dry-spun acrylic fiber; DOC, dissoluble organic matters; DCDE, dichlorodiethyl ether; EDCs, endocrine disrupting chemicals; EF, electro-Fenton; EOCs, emerging organic contaminants; HPAM, polyacrylamide; IBR, immobilized biomass reactor; LAB, linear alkylbenzene; LAS, linear alkylbenzene sulfonate; MBR, membrane bioreactor; MBBR, moving-bed biofilm reactor; MPG, α-methylphenylglycine; NXA, nalidixic acid; nZVI, nano zero-valent iron; PAHs, polyaromatic hydrocarbon; PCBs, polychlorinated biphenyl; PF, photo-Fenton; PPCPs, pharmaceuticals and personal care products; PYR, Pyrene; SBR, sequencing batch reactor; SBBR, sequencing batch biofilm reactor; SMBR, submerged membrane bioreactor; SMX, sulfamethoxazole; SOC, soluble organic carbon; SOM, soluble organic matters; TCDD, 2,3,7,8-tetrachlorodibenzo-p-dioxin; TEO, dichloromethane organics; TN, total nitrogen; TOC, total organic carbon; TP, total phosphorus; TPH, total petroleum hydrocarbon; UASB, Upflow Anaerobic Sludge Blanket; UV, ultraviolet; WW, wastewater; WWTP, wastewater treatment plants.

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Biotreatment
Wastewater
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Remediation

for potential applications of this combination in different polluted soil and wastewater, as well as the mechanism of combination was also discussed.

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1. Introduction

Water and soil pollution resulting from illegal discharge and incomplete treatment of waste has caused high concerns (Fu et al., 2014; Franco and Sarria, 2015). Various contaminants persisting in environment include pesticides, dyes, polycyclic aromatic hydrocarbon, polychlorinated biphenyl, heavy metals etc. (Ngah et al., 2011; Brito et al., 2015; Marina et al., 2015). All of these pollutants releasing into the environment pose a huge threat to ecosystem and human health (Freedman, 2008; Murthy and Ramesh, 2009; Trevors, 2010; Xu et al., 2012), making it critical to develop treatment technologies for removal of these pollutants from environment (Malarvannan et al., 2009; Bechmann et al., 2010).

As one of the efficient technologies, advanced oxidation processes (AOPs), have been frequently used for remediation of polluted soil and water in recent years (Huang et al., 2015). AOPs include various technologies (Vilhunen and Sillanpää, 2010; Fernández-Castro et al., 2015; Cheng et al., 2016a) such as Fe^{2+}/H_2O_2 (Kallel et al., 2009), ozonation(O_3) (Esplugas et al., 2007), H_2O_2/UV (Moro et al., 2013), hetero-/homo-geneous Fenton-like process (Wang et al., 2016), photocatalysis (Elghniji et al., 2012), and other treatment technologies may contain electrify, ultrasonic, radiation (Luna et al., 2012; Zhang et al., 2013a; Huang et al., 2014). Fenton processes (Fe^{2+} / H_2O_2) are able to oxidize pollutants by producing hydroxyl radical (•OH) (Maezono et al., 2011; Wang et al., 2016). •OH is the major reactive intermediate responsible for organics' oxidation and is regarded as the dominant oxidant, which has been mentioned by some key publications (Georgiadis, 2008; Rd et al., 2012). On account of short reaction time, high efficiency of pollutants degradation and the wide diversity of target contaminants, Fenton processes have become the representative AOPs. Fe^{2+} is used as the catalyst and hydrogen peroxide is used as the oxidant in Fenton oxidation. On the other hand, biotreatment is regarded as an environmental friendly method for pollutants treatment, but the reaction conditions need to be carefully regulated (Ganigué et al., 2012; Wan et al., 2013; Xiao et al., 2015). In addition, biotreatment technologies might be not effective at high pollution level due to the limited resistance of microorganisms to toxicity (Kundu et al., 2012).

Combination of Fenton processes and biotreatment is developed to overcome the defects of Fenton technology (e.g., consumption of reagents and drastic reaction process, etc.) or the limitations of biotreatment (e.g., strict reaction condition and time-consuming, etc.). The combination system has been used in wastewater and contaminated soil treatment (Sirtori et al., 2009; Venny and Ng, 2012; Jho et al., 2014; Bing et al., 2015). The Fenton process in the combination system can improve the biodegradability of wastewater, which is beneficial to biotreatment, while biotreatment in the same combination system can stabilize the waste and reduce the use of Fenton chemicals. Fenton process or biotreatment is not only a pre-treatment or post-treatment, since the treatment would usually not finish unless the concentration of pollutants drop to 0. So far the review about the combination of Fenton process and biotreatment has not been reported.

This review describes the recent studies in adopting combined or sequential Fenton/biotreatment for wastewater and polluted soil treatment, and highlights the benefits of the combined treatment. The review firstly introduces basic information about single Fenton process and biological treatment, followed by the considerations in the combined treatment method. Meanwhile, the combined method is classified into (1). Fenton processes and then biological treatment, and (2). biological treatment and then Fenton processes. The outlook and future study in this promising field are also discussed. The structure of this review is illustrated in Fig. 1.

2. Fenton processes and biological treatment

Fenton processes (Fenton, Fenton-like and modified Fenton) and biotreatment have been frequently studied for pollutants removal. Below are the basic illustrations of practical application or experimental researches about single Fenton processes and biotreatment.

2.1. Fenton processes

Fenton technology is a promising and alternative method for remediation of soil or wastewater treatment. Laboratory, plant-scale experiments and practical application of Fenton technology were conducted by many researchers. Fenton process uses hydrogen peroxide as oxidant and ferrous ions as catalyst. The catalytic decomposition of H_2O_2 in the presence of Fe^{2+} involves a complex chain reaction. The critical reactions are listed below (Gu et al., 2012):

$$Fe(II) + H_2O_2 = Fe(III) + {}^{\cdot}OH + OH^- \quad (1)$$

$$Fe(III) + H_2O_2 = Fe(II) + H_2O + H^+ \quad (2)$$

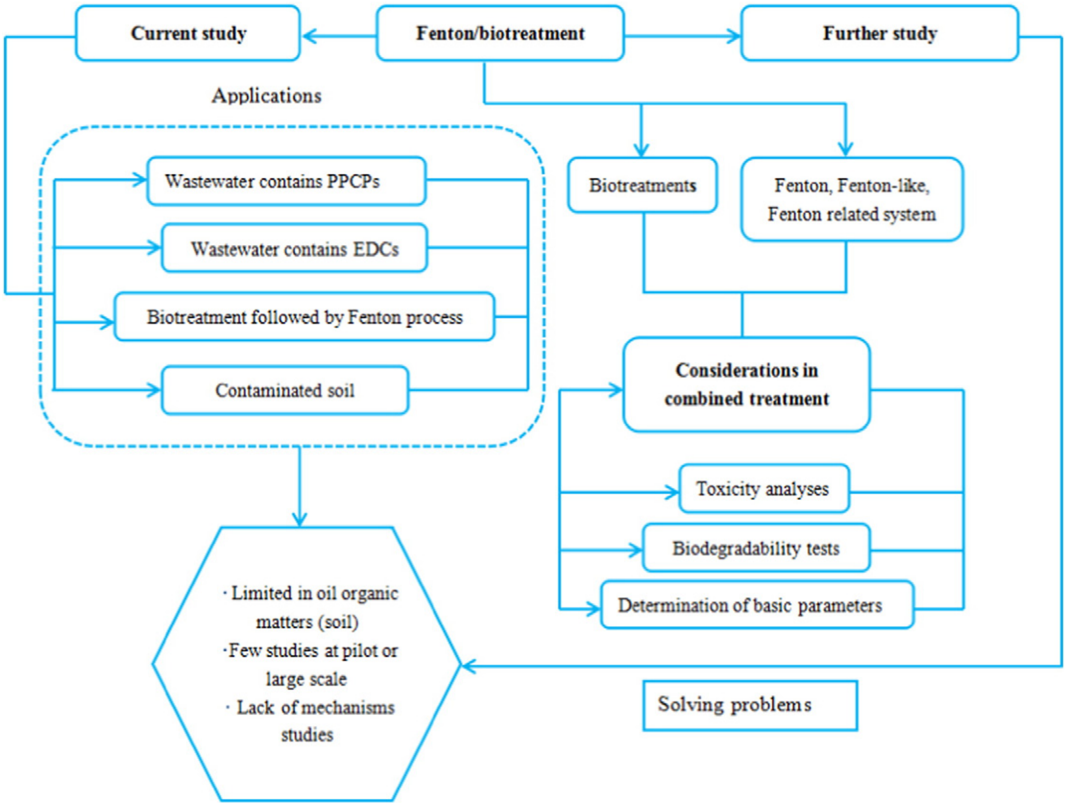

Fig. 1. Overview of review structure.

$$\cdot OH + Fe(II) = Fe(III) + OH^- \quad (3)$$

$$\cdot OH + H_2O_2 = \cdot OOH + H_2O \quad (4)$$

$$\cdot OH + organic = products + H_2O + CO_2 \quad (5)$$

H_2O_2 is decomposed in the presence of Fe^{2+} catalyst, and causes the generation of •OH (Eq. (1)). Afterwards, the active oxidant •OH remove recalcitrant compounds. It is essential that the chemical reaction should be operated at pH of 2.8–3.0, where the Fe^{3+}/Fe^{2+} couple can perform catalytic behavior (Aaron, 2014). The change in pH could diminish the catalytic activity. Only a small amount of Fe^{2+} is required to initiate Fenton reaction because Fe^{2+} could be regenerated (Eq. (2)). Fig. 2 shows the concise features of Fenton and Fenton-like process.

Fenton reaction can be performed at room temperature and ordinary pressure, and possesses high performance and non-toxicity (H_2O_2 is transformed into H_2O and O_2). Therefore, Fenton technology has been extensively applied to the treatment of different wastewater (Bautista et al., 2008) such as pesticides wastewater, pharmaceutical wastewater, laboratory wastewater and fermentation wastewater. However, there are also some limitations in Fenton process, including the relatively high costs of reagents, and the large volume of iron sludge. The various hetero-/homo-geneous catalysts including Fe^{3+}, Cu^{2+}/Cu^{+}, pyrite, and nano zero-valent iron were used to replace the Fe^{2+} for enhancing efficiency. In addition, modified Fenton processes were developed by combining classic Fenton oxidation with the physical methods such as UV, ultrasonic, electric or microwave. Recently, modified Fenton technologies (Bautista et al., 2008; Jiang et al., 2009) have been widely used for its high adaptation and degradation ability, which are reviewed by some authors (Bautista et al., 2008; Wang et

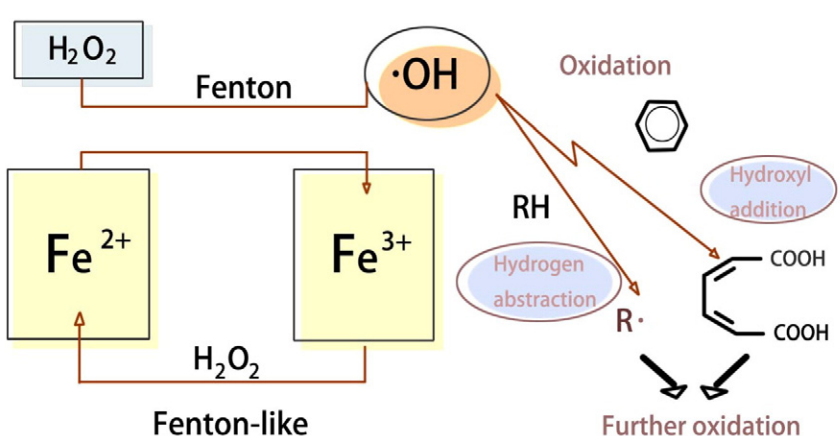

Fig. 2. Characteristic of Fenton and Fenton-like process (modified from Cheng et al., 2016a).

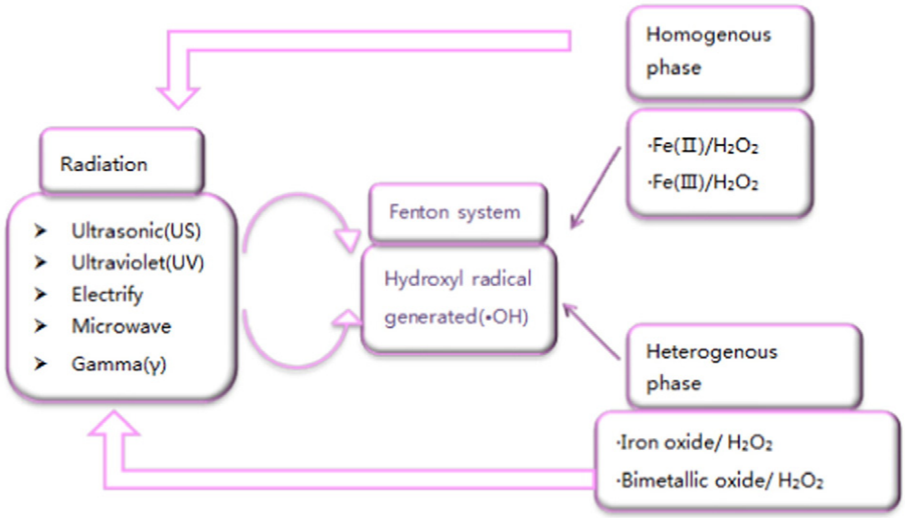

Fig. 3. The basic classification of Fenton system.

al., 2016). The classification of common Fenton technologies was briefly summarized in Fig. 3.

2.2. Biological treatment

Biological treatment is a technology which can remove environmental pollutants by organisms or extracellular enzymes. A simple classification of biotreatment is presented in Fig. 4. Microbe occupied an important position in biological treatment. The essence of biological treatment is organic metabolism that the organisms used substrates as energy and carbon resource, and the substrate matter can be served as the electron donor in redox reaction of organisms' development. The organism, such as bacteria, filamentous fungi, yeast, algae or plant, is used to remove pollutants in biotreatment technologies (Huang et al., 2008, 2010a). Biological treatments include composting, bioreactors, biofilters and biostimulation etc., which possess the advantage of low cost, high removal rate, no secondary pollution and so on (Boopathy, 2000). A study of anaerobic bacterial degradation of crude methanol showed that microorganisms in sediment degraded methanol completely (Yuan et al., 2016). Bustamante et al. (2013) found that composting effectively stabilized digestate, and compost can be used as soil conditioner. The efficiency of biological treatment is affected by environmental conditions, species and quantity of microorganism, and characteristics of pollutants (Haritash and Kaushik, 2009; Huang et al., 2010b). Aziz and Aziz (2011) concluded that the increase in aeration rate resulted in an increase in chemical oxygen demand (COD) concentration during biotreatment of landfill leachate. Joss et al. (2009) found that the presence of toxic substances inhibited the ammonification by microorganisms. In addition, Lu et al. (2010) found that petroleum-contaminated soil after composting needs a further treatment. Therefore, a

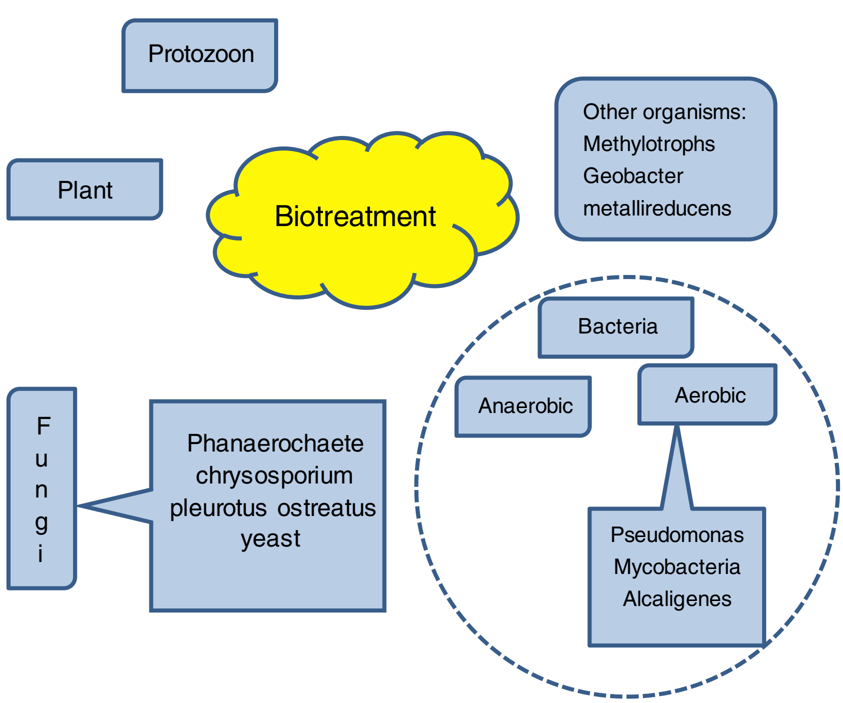

Fig. 4. The simple classification of biotreatment.

combination of biological treatment and other technologies such as Fenton (Padoley et al., 2011), photo-Fenton (Sirtori et al., 2009), electron-Fenton (Fatiha et al., 2013), etc. has attracted great concern.

3. Considerations in combined method of Fenton and biological treatment

The composition of wastewater is complicated, so preliminary analyses of wastewater are always performed for choosing suitable treatment methods. The parameters total organic carbon (TOC), chemical oxygen demand (COD), biochemical oxygen demand (BOD) and toxicity are analyzed. Wastewater with highly toxic should be disposed by Fenton process first. If the COD value of wastewater with good biodegradability cannot meet discharge standards, wastewater could be disposed by further biological method after by Fenton process. Partially toxic wastewater with low biodegradability and relatively high TOC value should be pretreated by Fenton process. Fig. 5 summarizes the different steps necessary to apply Fenton/biotreatment in different wastewater. From this figure, it is observed that the toxicity analyses and biodegradability analyses were significant to apply combined method of Fenton process and biological treatment. Toxicity analysis is usually done by acute toxicity testing using microorganisms such as *D. magna*, *Selenastrum capricornutum*, *Vibrio fischeri*, (Fernández-Alba et al., 2002; Emery et al., 2005), *Pseudomonas* (*fluorescens* or *putida*) (Lange et al., 2006), and *Escherichia coli* (Chatzitakis et al., 2008), etc. In addition, the respirometric assays have been used as an efficient method for the measurement of acute toxicity, since the oxygen demand in assays represents the activity and viability of aerobic microorganisms. Biodegradability can be monitored by some ways which include the simple analysis of BOD_5 and COD and the calculation of BOD_5/COD. The ratio shows the proportion of organic matters that are biodegradable under aerobic conditions. Additionally, Zahn-Wellens test is one of biodegradability assays. In this assay, analyzed samples are considered as biodegradable when the biodegradation percentage is over 70% (Lapertot et al., 2008). Although the steps of applying Fenton and biotreatment in soil remediation are not clear, the significant steps like toxicity and biodegradability analyses used in wastewater treatment could be used as reference.

When applying combined Fenton process and biological treatment, there are several issues should be paid attention to:

(i). The proper dosage of chemical oxidant. For instance, too many oxidizers would induce excessive mineralization of effluent, leading to insufficient carbon source for microbes in the subsequent biotreatment.

(ii). The influence of Fenton reaction on indigenous microbes. For example, the optimal value of pH for Fenton reaction is 2.8–3.0, which might not be unfavourable to the growth of some indigenous microbes.

(iii). The inhibition of matters from previous treatment to the next process. Organic matter might be produced in biotreatment process, which would reduce the production of hydroxyl radicals by reacting with catalysts in the following Fenton process.

(iv). The competition for hydroxyl radicals. Some matters in the environment, such as soil organic matters (SOM) in soil, can compete for hydroxyl radicals with target pollutants. Many researchers found that the competition led to a decline of remediation efficiency (Zapata et al., 2008).

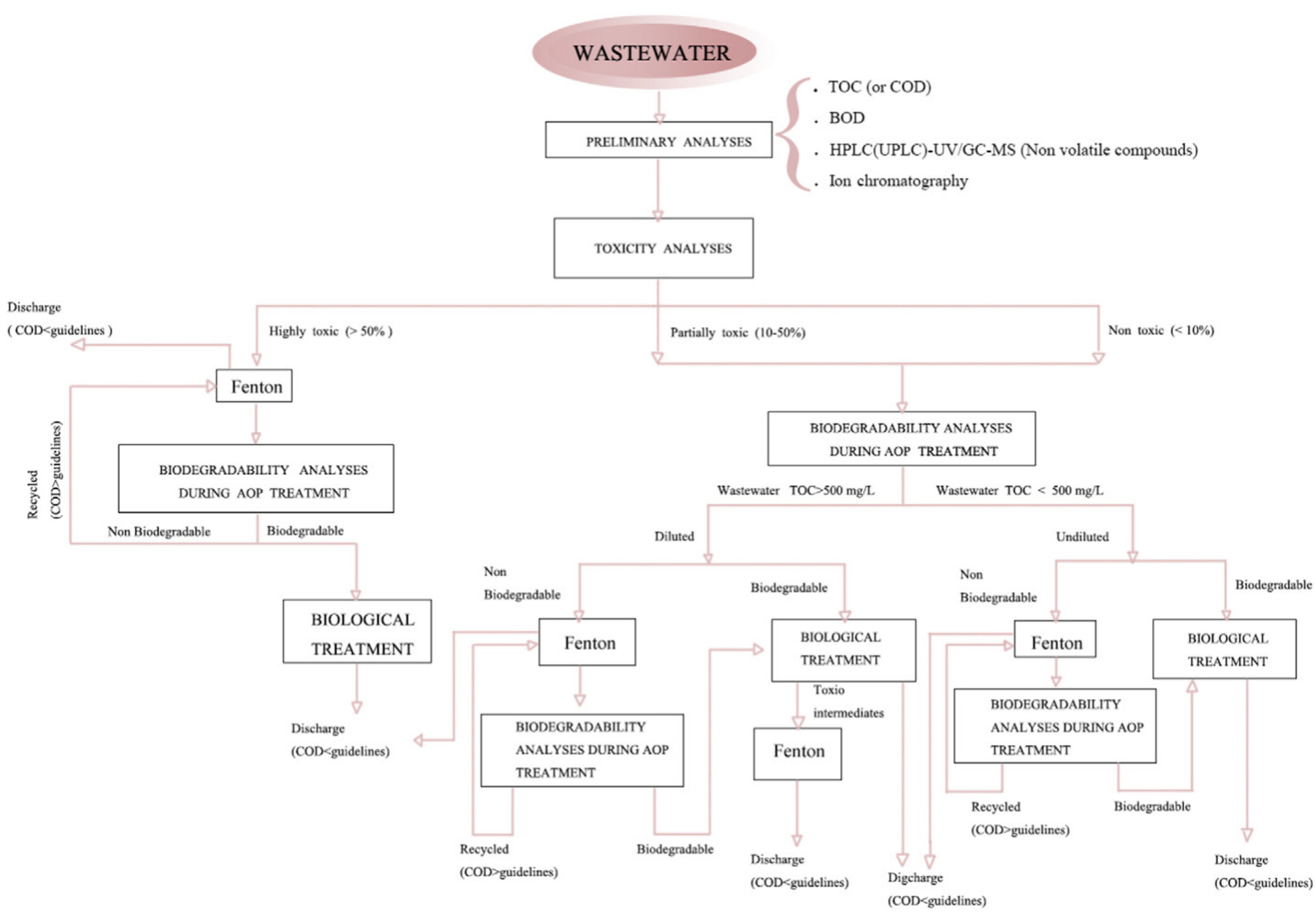

Fig. 5. Selection for the best treatment option of a specific toxic and/or non-biodegradable wastewater (modified from Fig. 1. in Oller et al., 2011).

(v). The analysis of normal parameters including volatile solids, total suspended solids, TOC, pH, temperature and nutrients elements. The growth of microbe is affected by nutrients elements (Sirtori et al., 2009).

In practically, for the remediation of contaminated soil, there is another consideration that hydroxyl radicals may react with soil humus due to the nonselective nature of hydroxyl radicals. In this case, the limitations must be taken into account when infer the reaction mechanisms, pathway and kinetics.

In general, analyses of the above different parameters could provide useful information for applying Fenton/biotreatment. And the combined method has been successfully applied in wastewater treatment and contaminated soil remediation.

4. Wastewater treatment by Fenton processes followed by biological treatment

Researchers focused on the application of Fenton processes followed by biological treatment in wastewater treatment. Using Fenton process as a pre-treatment method can improve the efficiency of biotreatment, which is because the intermediates in the process are more biodegradable than original ones. The combined method of Fenton process and biological treatment could meet more stringent regulations, and its applications in the treatment of pharmaceuticals or personal care products (PPCPs) and endocrine disrupting compounds (EDCs) wastewater are highlighted in this review.

4.1. Wastewater containing pharmaceuticals or personal care products

Nowadays, the treatment of wastewater containing pharmaceuticals and personal care products (PPCPs) has attracted attention (Matamoros et al., 2015). PPCPs, such as sulfamethoxazole (SMX) (Gonzalez et al., 2007), tetracycline (Wu et al., 2010) and alkanolamines (Klare et al., 2000), are used commonly as pharmaceuticals or personal care products and resistant to biodegradation. Fenton process followed by biotreatment has been introduced for the removal of PPCPs from wastewater (Gonzalez et al., 2009; Harimurti et al., 2010).

Padoley et al. (2011) used Fenton process to improve the biodegradability of pyridine and 3-cuanopyridine wastewater, because the wastewater was characterized by high COD concentration and low biodegradability. Then biotreatment with the isolated *Pseudomonas* (*pseudoalcaligenes*-KPN) were carried out for further treatment. It was observed that the removal rate of pyridine and 3-cuanopyridine reached to 84% and 99%, respectively. Photo-Fenton reaction was optimized by researchers to improve the biodegradability of SMX wastewater for the subsequent biological treatment (Gonzalez et al., 2009). They found that SMX was degraded completely in a relatively short time in the sequencing batch biofilm reactor, and aeration benefited the mineralization of organic carbon. Other researchers used the combined treatment to treat amoxicillin and ampicillin wastewater (Elmolla and Malay, 2011; Elmolla and Chaudhuri, 2012). In addition, the combined treatment of Fenton and activated sludge process was developed to treat monoethanolamine (MEA) wastewater (Harimurti et al., 2010). The combined system may similar to the process shown on Fig. 6. Results showed that the removal rates of MEA and COD in wastewater treated by the combined method were higher than those by activated sludge process without Fenton pretreatment. Biodegradability of MEA wastewater was improved by Fenton pretreatment, which indicated that the combined method is promising in the treatment of MEA wastewater. Fatiha et al. (2013) adopted the electro-Fenton process followed by activated sludge process to remove tetracycline from wastewater, and 69% and 86% of TOC was removed after 2 and 4 h electrolysis, respectively. Moreover, the BOD_5/COD ratio in tetracycline wastewater was increased by using electro-Fenton method, and as a result the mineralization rates of tetracycline increased from 46% in the treatment with electro-Fenton to72% in the treatment with combined method.

4.2. Wastewater containing endocrine disrupting compounds

Endocrine disrupting compounds (EDCs) are contaminants with estrogenic or androgenic activity at quite low concentrations, including pesticides or herbicides (Lindan, Diuron and Linuron, etc.) (Maria et al., 2008), industrial compounds (PCBs, bisphenol A, phthalate esters, etc.) (Lau et al., 2005; Kaneco et al., 2006), and benzene ester (Wu et al., 2013), etc. Physical, chemical and biological technology was used for treating EDCs (Jeon et al., 2013; Komesli et al., 2015), and novel

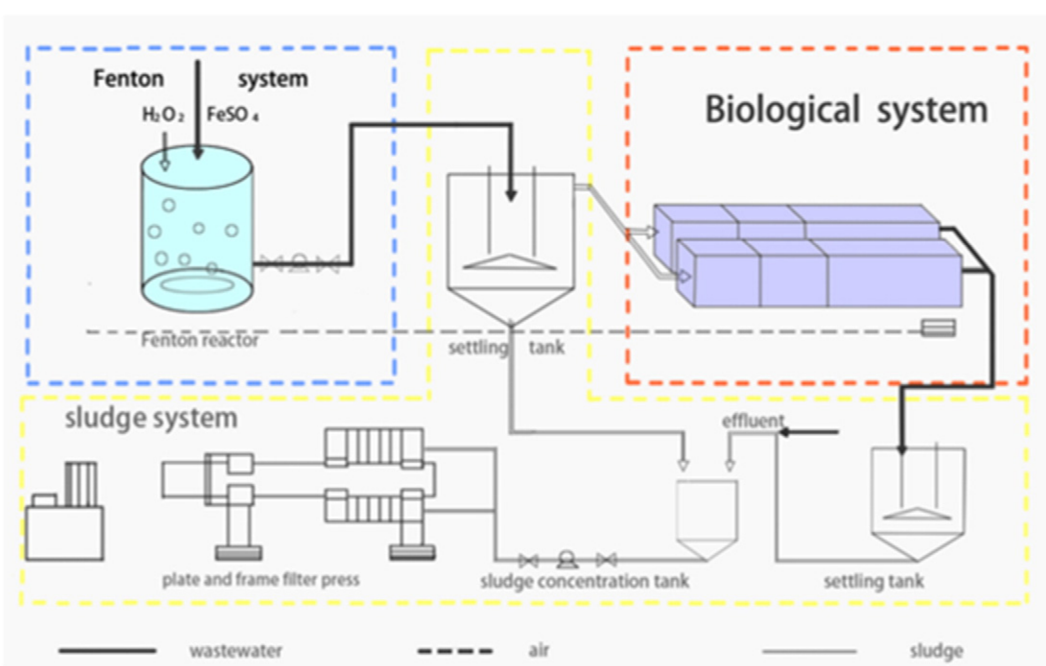

Fig. 6. The whole combined treatment process including sludge treating procedure (some are modified from Zhang et al., 2013c).

combined method of Fenton and biotreatment was proved to be one of the most effective methods (Chen et al., 2009; Vilar et al., 2012).

Pesticides are resistant and many of them were regarded as EDCs (Konstantinou and Albanis, 2003). Oller et al. (2007a) obtained 92% of dissolved organic carbon (DOC) removal rate and 85% of pesticides removal rate by photo-Fenton treatment followed by aerobic biological reactor. Similar conclusions were confirmed in the studies reported by Maria et al. (2008) and Vilar et al. (2012). Recent researches of removing the pesticides belonging to EDCs from wastewater by Fenton/biotreatments are summarized in Table 1.

Combined method of photo-Fenton and fixed bed reactor was used to break down di-(2-ethylhexyl) phthalate (DEHP) in synthetic or practical wastewater (Chen et al., 2009), producing a relatively low-toxic and biodegradable solution. After photo-Fenton oxidation, the BOD_5/COD ratio of prepared effluents increased from 0.19 to 0.45, indicating the enhancement of biodegradability. The removal rate of DEHP was over 80% after 3 days of treatment with the combined method in practical wastewater, but the presence of other organic pollutants in practical wastewater decreased the removal rate.

Bamboo industry in china produces wastewater which contains organic acids, polyphenols, amino acids, flavonoids, dibutylphthalate, etc. Wu et al. (2013) found that after Fenton and sequencing batch reactor (SBR) process, phthalate and derivants of benzene ester were reduced completely. The increase of amide and alkane may be attributed to the existence of by-products from different stage of combined treatment.

4.3. Wastewater containing other pollutants

The combined system has also been used in other wastewater treatment.

Dyeing and textile wastewater was regarded as low-biodegradable as it contains complex hard-degradable organic matters (Zonoozi et al., 2008; El-Desoky et al., 2010). Lucas et al. (2007) found that Fenton process followed by yeast (*C. oleophila*) treatment can remove 95% of Reactive black 5 while Fenton process alone required 5 times reagents to achieve an identical level. The combination treatment obviously relieved financial pressure. Rodrigues et al. (2009) used a Fenton/biotreament in a SBR to remove organic matter and color with the purpose of purifying the synthetic effluent. Global removal rates of DOC, color and BOD_5 are 90.2% 97.3%, 96.1%, respectively. They were higher than those obtained by biologically treated effluent 28.9%, 36.0%, 63.9%, respectively. Wei et al. (2015) observed that Fenton process enhanced BOD_5/COD ratio from 0.35 to 0.69 in dry-spun acrylic fiber (DAF) wastewater, and toxicity was found a notable reduction via the experiments of *Vibrio fisheri* bacteria light loss. After biotreatment, the average removal rates of COD, NH_4^+-N and total nitrogen (TN) reached to 82.5%, 98.4%, and 74.6%, respectively. The combined treatment made final effluents meet the discharge standard.

The Fenton/biotreatments can be a useful treatment for landfill leachates, particularly the aged landfill leachates. Zhang et al. (2013b) adopted Fenton oxidation and submerged membrane bioreactor (SMBR) as appropriate options for old landfill leachate treatment. After Fenton oxidation, the removal rate of COD was 69.0%. The next SMBR process made the COD removal rate increase to 93.1%. The trend of TOC is the same as COD. Silva et al. (2013) found that solar photo-Fenton was also a beneficial pre-oxidation, which improved the biodegradability (higher than 70%) in wastewater according to Zahn-Wellens test.

In other studies, Wang et al. (2008) found that linear alkylbenzene sulfonate (LAS) in surfactant wastewater were decreased from 490 mg L^{-1} to 23 mg L^{-1} after Fenton oxidation and then the immobilized aerobic biomass reactor made LAS removal rate reached to 99%. And the biodegradability has been improved obviously even at a small concentration of H_2O_2. Khoufi et al. (2009) used the treatments that combined electro-Fenton, anaerobic digestion and ultrafiltration to treat olive oil wastewater at pilot scale. The results showed good removal rate of organic matter, and even obtained surplus energy after methanization in this experiment. In leather wastewater treatment, the Fenton process followed by biotreatment (*T. ferrooxidans*) system made the removal rates of COD, BOD and chromium reached to 93%, 98%, and 100%, respectively (Mandal et al., 2010). Other applications of Fenton processes followed by biotreatment to treat different wastewater are listed in Table 2.

5. Wastewater treatment by biological treatment followed by Fenton processes

Many researches confirmed that biological treatment followed by Fenton processes is effective in the removal of pollutants from wastewater (Weiwei et al., 2009; Zhang et al., 2014; Fernandes et al., 2014). For example, combined anaerobic digestion reactors and photo-Fenton process was found to yield good efficiencies (92–97%) for removal of azo dyes, which are well known for its resistance to biological treatment (García-Montaño et al., 2008). Azizi et al. (2015) found that the efficient system combining SBR and Fenton almost completed decolorization of Acid Red 18, whereas the decolorization rate was 88% in SBR. COD removal rate was nearly 97%, which showed an obvious increase compared to the sole treatment with SBR. Punzi et al. (2015) observed that the photo-Fenton process performed after biotreatment increased the removal rate of COD from 60% to 92%. In addition, the decolorization rate of a textile azo dye was nearly 100% in the combined treatment.

Nousheen et al. (2014) took sequential biological and photo-Fenton process for the treatment of mixed domestic and industrial wastewater. The results showed that the biotreatment decreased pollution load in raw influent, and then the photo-Fenton process removed more than 90% of COD and 80% of color from waste samples. The two-step combination also made a progress for the paper mill wastewater treatment (Fernandes et al., 2014). In one study, *Cryptococcus podzolicus* removed 68% of COD after incubation, and the next Fenton oxidation increased the removal rate of COD and TOC to 85% and 90% respectively (Fernandes et al., 2014). Zhang et al. (2014) used electro-Fenton (EF) as a post process after biotreatment (sequencing batch biofilm reactor) for landfill leachates treatment, and obtained remarkable augment in removal of COD and BOD_5. Some researchers obtained the similar results by three steps including biotreatment, coagulation, and EF (Moreira et al., 2015). A novel research applied Fenton oxidation between two biotreatment for landfill leachate treatment. In this research,

Table 1
Fenton processes followed by biotreatment for ECDs-pesticides wastewater treatment.

Target wastewater	Treatments	Main performances	Reference
Diuron and Linuron herbicides	Photo-Fenton/aerobic SBR	Complete TOC, Diuron and Linuron removal	(Farré et al., 2006)
Diuron and Linuron herbicides	Photo-Fenton/aerobic SBR	80% removal of TOC, complete Diuron and linuron removal	(Farré et al., 2007)
Phorate, Terbufos, phoxim Parathion, etc.	Fenton-coagulation/MBBR	72% removal of COD, 98% removal of organophosphorus	(Chen et al., 2007)
Chlorfenvinphos alachlor, atrazine, isoproturon diuron	Photo-Fenton/packed-bed bioreactors	Over 80% of DOC degraded, over 50% of the total carbon converted	(Lapertot et al., 2007)
Diuron and Linuron	Solar photo-Fenton/SBR	87% mineralization of two herbicides, 83% TOC removal	(Maria et al., 2008)
Eighteen pesticides	Photo-Fenton /IBR	86% removal for 18 pesticides, 91% COD was removed	(Vilar et al., 2012)
Perfekthion, Metomur, Couraze, Vydate	Photo-Fenton/MBR	Over 95% removal of DOC and COD	(Pérez et al., 2013)

Table 2
Fenton processes followed by biotreatment for various wastewater treatment.

Target wastewater	Treatments	Main performances	Reference
Containing MPG and DOC	Photo-Fenton /aerobic biological treatment	95% mineralizationg	(Oller et al., 2007b)
Linear alkylbenzene sulfonate(LAS)	Fenton/aerobic biological processes	94% of COD removal,99% of LAS removal	(Wang et al., 2008)
Dichlorodiethyl ether (DCDE)	Fenton/sequential batch reactor	75% of DCDE removed, 94% of TOC, 93% of COD removal	(Christensen and Gurol, 2004)
Di-(2-ethylhexyl) phthalate (DEHP)	Photo-Fenton/biological system	73.6% of mineralization, BOD_5/COD ratio up from 0.19 to 0.45, 80% of DEHP removal	(Chen et al., 2009)
Leather industry wastewater	Fenton/aerobic treatment(Thiobacillus ferrooxidans)	COD, BOD, sulfide, chromium and color removal were93%, 98%, 72%, 62%	(Mandal et al., 2010)
Monoethanolamine	Fenton/aerobic batch bioreactor	100% removal of MEA	(Harimurti et al., 2010)
Acrylic fiber contains sulfate	Fenton-UASB-SBR system	90% of COD removal sulfate removal were and 75%	(Li et al., 2011)
Ammonia formaldehyde	Electro-Fenton/biodegradation	Nearly complete COD and formaldehyde removal	(Moussavi et al., 2012)
Bamboo industry wastewater	Fenton–SBR process	BOD_5/COD from 0.13 to 0.50, TOC, NH_3–N, TN meet rule	(Wu et al., 2013)
Polyacrylamide wastewater	Fenton/anaerobic biological processes	COD_{Cr} and removal were 94.6% and 91.0%	(Pi et al., 2015)

Klauson et al. (2015) found that biological treatment followed by Fenton process removed over 90% of leachate organics, and plug-flow activated sludge process was further applied to remove the residual organics for meeting discharge standard. Other studies involving the applications of biological treatment followed by Fenton process are presented in Table 3.

6. Contaminated soil treatment by Fenton/biotreatment

Soils as well as the water are confronted with various pollutants. Soils are mainly contaminated by polycyclic aromatic hydrocarbon (PAHs), heavy metals, pesticides and fertilizers from industrial activities and agricultural practices etc. (Sayara et al., 2010; Garciadelgado et al., 2015; Cheng et al., 2016b). Soil contamination causes toxic effects on biota, resulting in unacceptable environmental risks (Jaco et al., 2009).

Remediating contaminated soil is a matter of concern because of the potential danger that pollutants posed to local ecosystem. Common ways including soil washing, incineration, land-filling, chemical oxidation or phytoremediation were used in soil remediation (Yeung and Gu, 2011; Beesley et al., 2011; Mao et al., 2015). Fenton oxidation or biotreatment was also considered as a good remediation method. However, there are chemical effects on soil environment resulted from Fenton reaction, and biotreatment was usually unable to remove high-toxic pollutants efficiently (Neyens and Baeyens, 2003; Dibyendu et al., 2005; Mariusz et al., 2009; Chiew et al., 2011). As time went by, researchers developed combined method of Fenton and biotreatment in contaminated soil remediation, showing better treatment efficiency than sole method. Some novel studies on the application of combination of Fenton processes and biotreatments are presented in Table 4.

Combined method was usually used to remove petroleum hydrocarbon from contaminated soils. In India, researchers found that 57% of aliphatic fraction (C_{14}-C_{28}) was removed in Fenton process and removal rate of C_{14}-C_{28} reached to75% after the next treatment by *Fusarium solani* (exogenous microbes), whereas sole biotreatment only gained a removal rate of 61% (Buragohain et al., 2013). Jho et al. (2014) used Fenton process and bioaugmentation for successful remediation of total petroleum hydrocarbon (TPH) contaminated soil. Bioaugmentation means enhancing microbial activity by adding microorganisms to soil. Another biotreatment method called as biostimulation is to enhance indigenous microbial community in soil by adjusting nutrients or providing electron acceptors or electron donors (Kanissery and Sims, 2011; Andreolli et al., 2015). Goi et al. (2006) found that Fenton-like process and biostimulation performed well in remediating oil contaminated soil, and the removal rate of oil was 74%. It was also observed in other study that removal rate of TPH was 88.9% in weathered oil-contaminated soil by combined Fenton process and biostimulation treatment (Gong et al., 2012). In this study, the amount and activity of indigenous microbes was increased in Fenton process, so the total removal rate of TPH after combined method treatment was obviously higher than that in biotreatment without pretreatment of Fenton.

Use of combined Fenton and biotreatment in creosote polluted soil, removal rate of total PAH was 75%, with 30% increase to the biotreatment (aerobic SBR) alone (Valderrama et al., 2009). Meanwhile, more drastic Fenton oxidation did not favor better biological treatment. The Fenton/biological treatment was also used for polycyclic aromatic hydrocarbon degradation. In the study, Rafin et al. (2009) found that the degradation efficiency of Benzo[*a*]pyrene increased by half and two times, respectively, by comparing to alone Fenton oxidation and alone biotreatment (by *Fusarium solani*). In addition, Composting was a treatment for solid waste but sometimes the compost needs further treatment. Lu et al. (2010) carried out a laboratory study using Fenton-like process and biotreatment for remediating petroleum-contaminated soil after composting. At the end of Fenton-like process, removal rate of total dichloromethane-extractable organics (TEO) was 32.7%. The next biotreatment destroyed 17.9% of TEO. And toxicity analyses showed that the toxicity was decreased obviously.

The use of chelator and interesting operation in Fenton/biotreatment experiment achieved satisfactory results (Xu et al.,

Table 3
Biotreatment followed by Fenton processes for various wastewater treatment.

Target wastewater	Treatments	Main performances	Reference
Green table olive processing wastewater	Biotreatment (Aspergillus nige)/Electro-Fenton Anaerobic–aerobic	96% removal of COD, 65% removal of selected phenols	(Kyriacou et al., 2005)
WW contains Cibacron Red FN-R azo dye	Anaerobic–aerobic biotreatment/photo-Fenton	92–97% of decolourisation, 83% of mineralization	(García-Montaño et al., 2008)
Swine WW contains veterinary antibiotics	Sequencing batch reactor (SBR)/Fenton	Over 95% removal of COD,SS, TN and NH_3–N, 89% removal of TP	(Weiwei et al., 2009)
Pharmaceutical WW contains nalidixic acid	Solar photo-Fenton/ biological treatment	96% removal of DOC, 50% removal of NXA	(Sirtori et al., 2009)
Landfill leachate	Sequencing batch biofilm reactor/electro-Fenton	TOC, COD, and BOD_5 removal were 40.5%, 71.6%, and 61.0%	(Zhang et al., 2014)
Naphthalene aqueous solution	Biodegradation/Fenton-like (nZVI) oxidation	91.6% removal of COD, 99.0% removal of naphthalene	(Bing et al., 2015)
WW contains textile azo dye	Anaerobic treatment/photo-Fenton	96% color removal, 92% removal of COD	(Punzi et al., 2015)

Table 4
Fenton/biotreatment for polluted soil treatment.

Target polluted soil	Treatments	Main performances	Reference
PCB Congeners polluted soil/sediment	Fenton/biotreatment (*Pseudomonas*, *Alcaligenes eutrophus*)	95% degradation of 2-chlorinated biphenyl	(Aronstein and Rice, 1996)
TCDD-contaminated soils	Fenton/aerobic biological treatment	2,3,7,8-tetrachlorodibenzo-p –dioxin degraded 99%	(Kao and Wu, 2000)
Manufacturered gas plant soil	Modified Fenton/biodegradation(bacteria) /surfactants	98% removal of 2-or 3-ring hydrocarbons,70%–85% removal of 4 or 5-ring	(Nam et al., 2001)
PAHs contaminated soils	Fenton/biotreatment(P. *testosterone*)	80–85% removal of PAHs	(Nadarajah et al., 2002)
Creosote polluted soil	Fenton-like/biotreatment	88.5% removal of PAHs	(Niina et al., 2006)
Oil contaminated soil	Fenton-like/biotreatment (no additional microbe)	74% removal of oil	(Goi et al., 2006)
Benzo[*a*]pyrene polluted soil	Fenton/biotreatment (*Fusarium solani*)/ cyclodextrins	High removal than chemical or biological alone	(Rafin et al., 2009)
Creosote-comtaminated soil	Fenton/aerobic sequencing batch reactor	Maximum PAH removal was 80%	
Petroleum-contamina ted soil	Fenton-like/biodegradation (inoculum isolated from oil polluted soil)	50.6% of total dichloromethane organics (TEO) removed	(Lu et al., 2010)
Oil contaminated soil	Modified Fenton/ bioremediation	93%removal of tank oil,C_{10}–C_{40} became more biodegradable	(Xu et al., 2011)
Weathered petroleum oil-contaminated soil	Modified Fenton/ biostimulation(indigenous , microbe)	88.9% removal of total petroleum hydrocarbons (TPH), microbial increased	(Gong, 2012)
PAH contaminated soil	Modified Fenton(chelator)/ biostimulation	85.76% and 77.46% removal of PAH in different column	(Venny and Ng, 2012)
Crude oil contaminated soil	Fenton/biological treatment(*Fusarium solani*)	75% degradation of aliphatic fractiotns (C_{14}–C_{28})	(Buragohain et al., 2013)
Diesel contaminated soils	Fenton and modified Fenton/ bioremediation	59% and 57% removal of TPH	(Sutton et al., 2013)
TPH polluted soil(model or field) Linear alkylbenzene polluted soil	Fenton/Bioaugmentation (TPH degrading mirobial)	Over 90% of TPH degraded	(Jho et al., 2014)
Pyrene-contaminated soil	Electro-Fenton/bioremediation (PYR-degrading bacteria)	91% removal of PYR	(Xu et al., 2015)
Linear alkylbenzene polluted soil	Modified Fenton/biostimulation *(Rhodococcus*, *Ochrobactrum* etc.)	65% degradation of LAB	(Martínez-Pascual et al., 2015)

2011). Xu et al. used citric acid as an iron chelator and added H_2O_2 intermittently to prevent the sharp increase of the temperature. The C_{10}-C_{40} compounds became more biodegradable after Fenton oxidation. The tank oil removal rate was 93%, and increased by 31% compared with biotreatment alone. Researchers also adopted bio-acidification with Fenton oxidation to remove heavy metals in sewage sludge (Ren et al., 2014). Bio-acidification used *Thiobacilli* bacteria that can oxidize sulfur into sulfuric acid, and then the acid could create a good pH condition for next Fenton oxidation. It was clear in the combined treatment that the solubilization efficiency of Cu^{2+}, Pb^{2+} and Cd^{2+} increased obviously. Xu et al. (2015) used the direct electro-Fenton process before biotreament. They found the removal rate of pyrene increased from 50% (individual electro-Fenton and biotreatment) to 91.0% (combined treatment). The combined treatment has been confirmed as a useful method for the remediation of contaminated soil.

7. Conclusions and future research needs

This review paper firstly introduces brief information about Fenton processes and biological treatment, and then the considerations such as toxicity and biodegradability tests in constructing a combined system are discussed. Furthermore, this review mainly talks about extensive applications of Fenton/ biotreatment in wastewater treatment and contaminated soil remediation. In these applications, authors were dedicated to adjust chemical and biological operating conditions for cost reduction, time saving and efficiencies improvement. So far, lots of efforts have been made for the combination system, and the research findings have brought about good progress, promoting the applications of these attractive combination treatments.

Based on the review above, the future research is recommended. Firstly, the applications of Fenton/biotreatment for the remediation of contaminated soil are explored currently, and the pollutants mainly are petroleum organic matters. Additional researches are needed into different pollutants to improve the understanding of the potential of Fenton/biotreatment for soil remediation. Secondly, there are insufficient instances of the new combination treatment in practical water and soil remediation, as well as pilot-plant and large scale operations. Thirdly, the interaction between two treatments needs to be further studied because of its undefinition, and so is the degradation mechanism of the combined treatment. In addition, since some substances may compete for chemical oxidant against the pollutants in Fenton process, it is necessary to research the solutions. Lastly, the whole expenditure should be taken into account for economic plan, to assuring the original technology will be cost-competitive.

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