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REVIEW ARTICLE

A review on sustainable reuse applications of Fenton sludge during wastewater treatment

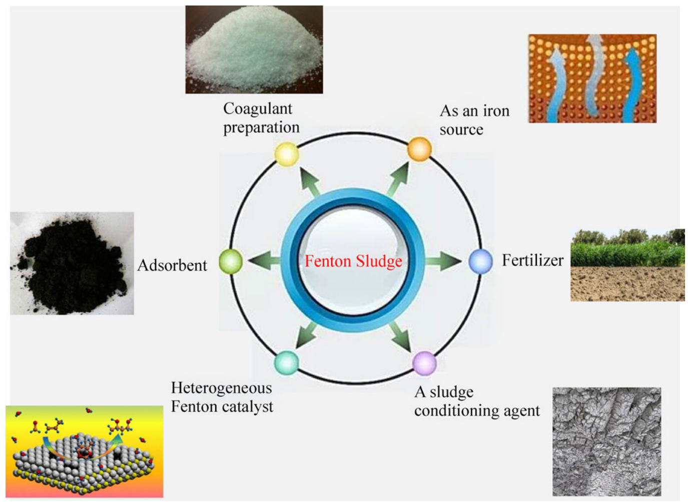
Lihui Gao (✉)[1], Yijun Cao[2,3], Lizhang Wang[1], Shulei Li (✉)[2]

1 School of Environment Science and Spatial Informatics, China University of Mining and Technology, Xuzhou 221116, China
2 National Engineering Research Center of Coal Preparation and Purification, China University of Mining and Technology, Xuzhou 221116, China
3 School of Chemical Engineering and Technology, Zhengzhou University, Zhengzhou 450001, China

HIGHLIGHTS

- The sustainable approaches related to Fenton sludge reuse systems are summarized.
- Degradation mechanism of Fenton sludge heterogeneous catalyst is deeply discussed.
- The efficient utilization directions of Fenton sludge are proposed.

GRAPHIC ABSTRACT



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ABSTRACT

The classical Fenton oxidation process (CFOP) is a versatile and effective application that is generally applied for recalcitrant pollutant removal. However, excess iron sludge production largely restricts its widespread application. Fenton sludge is a hazardous solid waste, which is a complex heterogeneous mixture with $Fe(OH)_3$, organic matter, heavy metals, microorganisms, sediment impurities, and moisture. Although studies have aimed to utilize specific Fenton sludge resources based on their iron-rich characteristics, few reports have fully reviewed the utilization of Fenton sludge. As such, this review details current sustainable Fenton sludge reuse systems that are applied during wastewater treatment. Specifically, coagulant preparation, the reuse of Fenton sludge as an iron source in the Fenton process and as a synthetic heterogeneous catalyst/adsorbent, as well as the application of the Fenton sludge reuse system as a heterogeneous catalyst for resource utilization. This is the first review article to comprehensively summarize the utilization of Fenton sludge. In addition, this review suggests future research ideas to enhance the cost-effectiveness, environmental sustainability, and large-scale feasibility of Fenton sludge applications.

1 Introduction

The sustainable treatment of industrial wastewater has been of increasing importance due to the significant presence of organic refractory and toxic contaminants. However, traditional separation and conversion methods are unable to effectively remove refractory and toxic pollutants, thereby requiring the development of new, effective technologies. In general, various highly loaded, refractory, toxic wastewater undergo advanced oxidation processes (AOP) for treatment, such as the Fenton oxidation process, O_3 oxidation, and photochemical

✉ Corresponding authors
E-mail: lihuigaocumt@163.com (L. Gao); lishuleibgs@163.com (S. Li)

oxidation (Badawy and Ali, 2006; Deng and Englehardt, 2006; Umar et al., 2010; Guvenc and Varank, 2021). As compared to other AOPs, the Fenton process is most significantly applied due to its high performance (fast degradation rate), simplicity (operating at room temperature and atmospheric pressure) and non-secondary pollution (H_2O_2 can be decomposed into environmentally safe substances, such as H_2O and O_2) (Wang et al., 2016).

The British chemist H.J.H. Fenton developed the Fenton oxidation process in 1984 based on the hypothesis of Haber and Weiss. In general, the Fenton process includes more than 20 chemical reactions (Barb et al., 1949; Walling, 1975), of which its core reaction is presented in Eq. (1). In the classical Fenton oxidation process (CFOP), an appropriate transition metal (Fe) is first applied to produce an active species, specifically •OH, which serves as a catalyst in the decomposition of hydrogen peroxide (H_2O_2) and ultimately in the effective mineralization of organic matter in wastewater (Duan et al., 2018; Jain et al., 2018; Sillanpää et al., 2018). In general, highly oxidative hydroxyl radicals (•OH) are acknowledged as the most effective oxidant due to their highly oxidative and non-selective reactions against organic contaminants (Páramo-Vargas et al., 2016; Tang et al., 2019; Mahtab et al., 2021b). Figure 1 presents the oxidation mechanism of the Fenton process. As such, the Fenton process is widely applied in various organic wastewater treatments.

$$Fe^{2+} + H_2O_2 + H^+ \rightarrow Fe^{3+} + H_2O + \bullet OH \qquad (1)$$

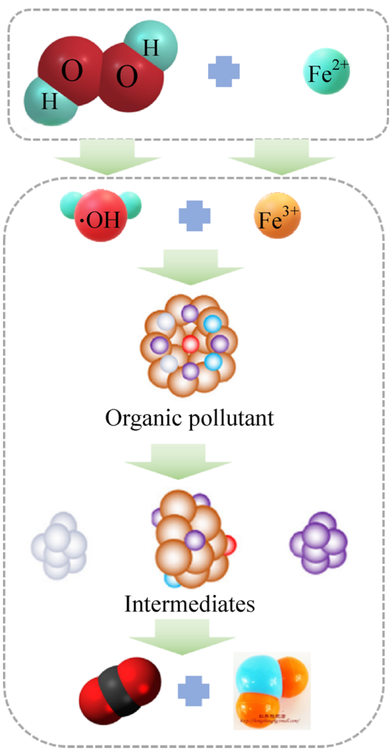

Fig. 1 Reaction mechanism for the Fenton process.

CFOP, however, presents several drawbacks, such as the requirement for pH adjustment, high cost of H_2O_2, and excess sludge production, which significantly limits its application (Bautista et al., 2008; Oturan and Aaron, 2014; Jain et al., 2018; Bello et al., 2019; Zhang et al., 2019b; Ghernaout et al., 2020). These shortcomings hinder the widespread application of this method, particularly due to emerging restrictive legislature on sludge disposal and economic assessments on wastewater treatment processes (Mahtab et al., 2021a). The Fenton sludge yield is largely dependent upon the proportion and volume of the reagents. In addition, the sludge produced by Fenton's treatment of refractory wastewater is usually regarded as hazardous solid waste due to the excessive amount of residual pollutants, which increases the cost of sludge disposal and the risk of secondary pollutants. Although researchers have begun to study the resource utilization and reduction of Fenton sludge, achieving bulk Fenton sludge consumption is still a huge challenge. This present review aims to summarize the physicochemical properties of Fenton sludge and detail the application of various Fenton sludge utilize systems as sustainable solutions.

2 Fenton sludge characterization

Fenton method applications generally produce excess sludge, which is a significant disadvantage. According to literature, approximately 40–180 mL/$L_{treated\ wastewater}$ of Fenton sludge is produced (Guedes et al., 2003; Benatti et al., 2006; Mahiroglu et al., 2009), which covers a sludge treatment cost of approximately 1–2 Euro/$m^3_{wastewater}$ (Di Iaconi et al., 2010). Sludge generation increases the imminent risk of further pollution, which can be resolved by sludge remediation and recycling. However, this sludge remediation increases cost by up to 30%–35% of the total working wastewater treatment cost (Neyens and Baeyens, 2003). Although some studies reported that the generated sludge is easily settleable, can be dewatered due to its high density, and has an efficient settling velocity that does not require further conditioning (Bolobajev et al., 2014; Kattel et al., 2016; Klein et al., 2016), still the physicochemical characterization of Fenton sludge and related research is limited.

Fenton sludge, a new type of complex heterogeneous sludge, is a mixture of $Fe(OH)_3$, organic matter, heavy metals, microorganisms, sediment impurities, and moisture. Fenton sludge properties are largely dependent on its wastewater source as well as the volume and ratio of the added reagents. Table 1 presents the characteristic of Fenton sludge under different wastewater. Fenton sludge contains 20%–30% organic matter, and the organic matter mainly includes organic acids, phenols, and organic substance flocculated and adsorbed during the Fenton process. Fenton sludge organic content is largely composed of 7.36% carbon, 12.52% oxygen, 2.25% hydrogen,

Table 1 Characteristic of Fenton sludge from different wastewaters

Elements	Dye wastewater (Zhang et al., 2018)	Landfill leachate (Dantas et al., 2020)	Papermaking wastewater (Fan et al., 2016)	Bagasse wastewater (Hua, 2017)	Papermaking wastewater (Zhang, 2013)
pH	11.3	–	7.2–7.4	7.46–7.66	7.2
TCOD (mg/L)	230	5500±1100			
SCOD (mg/L)	81	–			
Si (%)	0.32	0.41			
Fe (%)	13.4	19.51			
Ca (%)	0.35	0.47			
Na (%)	12.57	12.81			
Al (%)	0.091	0.07			
Mg (%)	0.44	0.51			
Cu (mg/kg)	12.71	13			
Zn (mg/kg)	358	902			
As (mg/kg)	0.22	–			
Ni (mg/kg)	51	123			
Cr (mg/kg)	0.07	0.05			
H (%)			2.25	–	–
O (%)[a)]			12.52	–	–
N (%)			0.78	–	–
S (%)			0.85	–	–
Organic content (%)			24.51	28.5	16–18
HHV (MJ/kg)			3.54	–	–
CaO (%)			1.17	5.46	3–3.5
Na_2O (%)			0.33	0.68	< 1.7
MnO (%)			0.06	0.17	< 1.7
Fe_2O_3 (%)			56.06	53.2	50–60
K_2O (%)			0.01	0.06	< 1.7
MgO (%)			0.11	0.06	1–1.6

Notes: a) O content = organic content in sludge-C-H-N-S (Fan et al., 2016).

0.78% nitrogen, and 0.85% sulfur (Hua, 2017). Only a small part of the Fenton sludge (after dewatering) is solid matter, such that over 70% is water. The ash from Fenton sludge contains mainly $Fe(OH)_3$, which can be converted to Fe_2O_3 at higher temperatures. Furthermore, some heavy metals such as Mg, Cu, As, Ni, Cr, and Zn can also be detected. Benatti et al. investigated the composition and metals in Fenton sludge, to which their results found metals in Fenton sludge that were mainly constituted of amorphous materials (> 80%) (Benatti et al., 2006). Meanwhile, the metals in Fenton sludge that originated from distinct wastewater presented similar characteristics, specifically exchangeability and the presence of amorphous iron oxide (Benatti et al., 2009). Therefore, Fenton sludge can be categorized as an iron-contained hazardous waste, such that utilization and recovery can be catered around its iron-rich characteristics.

3 Fenton sludge reuse system: Application and mechanisms

Many efforts are being made to find proper and suitable uses for Fenton sludge to minimize the production Fenton process solid waste. However, due to the high content of heavy metals and other harmful organic substances in Fenton sludge, the few studies on its utilization have mainly focused on the aspects detailed below.

3.1 Preparation of coagulant

Ferric ions and polymeric ferric sulfate (PFS) are commonly used flocculants in wastewater treatment (Zhang et al., 2010; Ge et al., 2020). The composition of Fenton sludge is mainly $Fe(OH)_3$, and it can be dissolved as Fe^{3+} in an acidic environment. Therefore, some researchers try to convert solid waste-Fenton sludge into flocculant to realize the reduction and resource utilization of Fenton sludge. Figure 2 summarizes a literature survey of various reuse systems/methods with respect to different flocculants, which can be dived into two categories: One is the sludge directly recycling system, another is synthetic flocculant through acid dissolution-modification (reduction and oxidation) processes. In the sludge recycling system, the solid waste from Fenton oxidation process is generally directly introduced into a coagulation reactor (Yoo et al., 2001). As compared to the sole addition of coagulants, a mixture of Fenton sludge and coagulants as well as coagulation recycling can significantly reduce coagulant dose requirements and sludge production by up to 50% (Yoo et al, 2001). In addition, sludge recycling during coagulation lowers the overall sludge generation during the Fenton oxidation process.

Several efforts have been made to convert Fenton sludge into a low-cost potential synthesized flocculant. The preparation process of flocculants from Fenton sludge mainly includes acid dissolution, reduction, and oxidation. The main oxidation methods include direct oxidation, catalytic oxidation, and microbial oxidation. Among them, the direct oxidation mainly uses strong oxidants such as HNO_3, H_2O_2, $NaClO_3$, and $KClO_3$. Zhang et al. produced a low-cost PFS through sulfuric acid dissolution-iron powder reduction-sodium chlorate oxidation, showing that the coagulation performance of sludge-based PFS is better than that of poly aluminum sulfate, but lower than that of commercial PFS (Zhang, 2013). However, Fenton sludge contains about 20%–30% organic matter, and these organic substances will not only cause the appearance of synthetic PFS to become dark black, but also likely affect the flocculation performance of synthetic PFS. Hua et al. comparatively studied the effect of organic matter on the flocculation performance of synthetic PFS, and the results showed that after calcination at 400°C to remove organics, the flocculation effect of synthetic PFS was significantly improved, and the removal efficiency was increased from 70% to 77% (Hua, 2017). In addition to the above method, alkalization is also a common flocculant synthesis process for Fenton sludge. Fan et al. synthesized a magnetic poly ferric sulfate (MPFS) product through alkalization, of which properties such as functional groups, polymer crystal structure, and surface structure are able to meet the requirements of commercial coagulants (Fan, 2016). Therefore, optimizing the preparation of flocculants may be a promising utilization solution of Fenton sludge.

3.2 Reused of Fenton sludge as an iron source

Previous studies have conducted the use of iron-rich Fenton sludge as a recycled iron source in the Fenton process (Bolobajev et al., 2014; Kattel et al., 2016; Klein et al., 2016), which shows that sludge reused system can be classified into two categories: sludge/H_2O_2 system and Fe^{2+}/sludge/H_2O_2 system. In sludge/H_2O_2 system, the acidic environment generally leads to the dissolution of iron on the surface of Fenton sludge and the dissolution mechanism may be mainly attributed to the following three types: protonation, complexation and reduction. The protonation mechanism is likely that the H^+ in the aqueous solution reacts with the OH bond at the surface of sludge to break the O-Fe bond and Fe^{3+} dissolves. The dissolution of iron by complexation involves the attachment of a complexing ligand onto the ferric oxyhydroxide surface, thereby resulting in iron dissolution (Kattel et al., 2016). Both humic and fulvic acids have been previously reported to effectively conjugate iron ions and thus form ion-ligand complexes (Voelker and Sulzberger, 1996; Paciolla et al., 1999). As a result, the adsorbed complexing ligands offer an electron to the ferric oxyhydroxide surface, thus reducing Fe^{3+} to Fe^{2+} (Kattel et al., 2016). Then the subsequent reaction mechanisms of activating H_2O_2 by dissolved Fe^{3+} are as follows (De Laat and Gallard, 1999; Kattel et al., 2016):

$$Fe^{3+} + H_2O_2 \rightarrow Fe(HO_2)^{2+} + H^+ \quad (2)$$

$$FeOH^{2+} + H_2O_2 \rightarrow Fe(OH)(HO_2)^{+} + H^+ \quad (3)$$

$$Fe(HO_2)^{2+} \rightarrow Fe^{2+} + HO_2^{\cdot} \quad (4)$$

$$Fe(OH)(HO_2)^{+} \rightarrow Fe^{2+} + OH^{-} + HO_2^{\bullet} \quad (5)$$

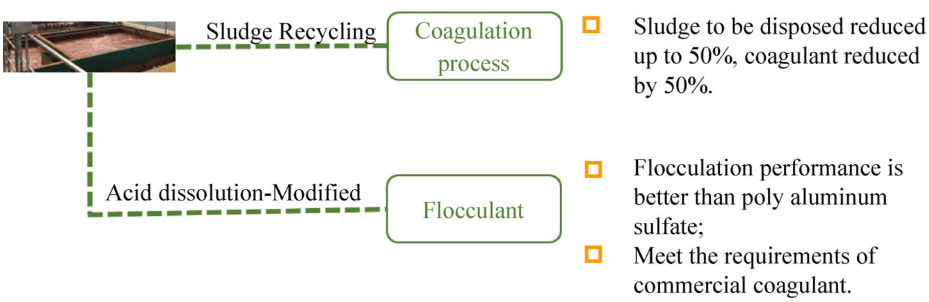

Fig. 2 Flowsheets of Fenton sludge utilization to produce flocculants

According to Eqs. (2) and (3), the reaction of H_2O_2 with Fe^{3+} produces ferric-hydroperoxyl complexes that decompose to generate Fe^{2+} and $HO_2^{\bullet}$, as detailed in Eqs. (4) and (5). However, the reaction rate of Eqs. (2)–(5) is extremely low, 2.7×10^{-3} M^{-1}/s, which is about 10^{-4} of the homogeneous Fenton reaction (Fe^{2+}/H_2O_2) (Ensing et al., 2003). Therefore, the Fenton-like reaction using Fenton sludge as the iron source may be mainly based on heterogeneous catalysis. Heterogeneous Fenton oxidation occur on the ferric oxyhydroxide surface in the H_2O_2/sludge system following a series of reactions (Lin and Gurol, 1998):

$$\equiv Fe^{3+}OH + H_2O_2 \leftrightarrow (H_2O_2)s \tag{6}$$

$$(H_2O_2)s \rightarrow \equiv Fe^{2+} + H_2O + HO_2^{\bullet} \tag{7}$$

$$\equiv Fe^{2+} + H_2O_2 \rightarrow \equiv Fe^{3+}OH + H_2O + HO\bullet \tag{8}$$

$$\equiv Fe^{2+} + O_2 \rightarrow \equiv Fe^{3+}OH + HO_2^{\bullet} \tag{9}$$

$$HO_2^{\bullet} \rightarrow H^+ + O_2^{\bullet-} \tag{10}$$

$$\equiv Fe^{3+}OH + HO_2^{\bullet-}/O_2^{\bullet-} \rightarrow \equiv Fe^{2+} + H_2O/OH^- + O_2 \tag{11}$$

Decantation and centrifugation wastewater treatment often result in the continuous loss of a ferric activator (Kattel et al., 2016), thereby requiring the addition of Fe^{2+} as an iron replacement (Bolobajev et al., 2014). On the contrary, due to the addition of Fe^{2+}, the efficiency of organic contaminants' degradation was improved compared with the sludge/H_2O_2 system. The complete utilization of the oxidant H_2O_2 can also be achieved by adding fresh activator (Fe^{2+}) to the H_2O_2/sludge system. Studies reporting Fenton sludge reuse as iron source systems are listed in Table 2. According to research by Li et al., the Fenton oxidation process using Fe^{2+} produced by electrolysis of Fenton sludge as the iron source can significantly reduce the COD and color of wastewater (Li et al., 2007). A previous study applied 160 mL sludge/L of petroleum refinery wastewater over 180 min, to which the results exhibited 53% mineralization (Diya'uddeen et al., 2015). Cao et al. investigated a sludge reuse system with additional regeneration, to which the results indicated effective wastewater treatment and enhanced biodegradability (Cao et al., 2009). Similarly, another study executed pulping wastewater treatment, of which a ferric salt recovery rate of 74.8% and total solid reduction of 87.3% were observed, which significantly lowered the resulting second environmental pollution (Chen et al., 2011). Previous studies have indicated similarities between the efficiencies of Fenton sludge reuse systems and CFOP within four reuse cycles (Bolobajev et al., 2014; Kattel et al., 2016; Klein et al., 2016), but the actual reuse cycles are significantly dependent on the applied wastewater and reuse methods. Li et al. and Kishimoto et al. conducted an electrolytic tank to reuse the Fenton sludge for Fenton process, indicating that iron recovery rate could reach 100% with the separation batch mode with high treatment efficiency (Li et al., 2007; Kishimoto et al., 2013). Kavitha and Palanivelu reused iron sludge for Fenton oxidation process after solid waste re-dissolution through chemical regeneration with acid and hydroxylamine (reducing agent), to which a contaminant removal of 97% was observed during the iron-containing sludge recycled (Kavitha and Palanivelu, 2004). The repeated use of Fenton solid waste may result in increased rates of total suspended solids (TSS) and turbidity as well as lowered zeta potentials, thereby decreasing sludge dewatering. In addition, high-temperature calcination in the sludge reuse system may result in iron leaching. Therefore, the development of new Fenton sludge reuse technology is of great significance to the efficient utilization of Fenton sludge.

3.3 Synthetic heterogeneous catalyst/adsorbent

Fenton reactions generally follow concise reaction conditions and produce excess iron-containing sludge. As such, many studies have developed catalysts such as iron oxide (Garade et al., 2009; Demarchis et al., 2015; Hou et al., 2017), high polymer material-supported iron-containing catalysts (Fernandez et al., 1998; Sabhi and Kiwi, 2001; Tao et al., 2003; Cheng et al., 2004), and inorganic material-supported iron oxide catalysts (Liao et al., 2009; Shukla et al., 2010) and different oxidants (H_2O_2 and $S_2O_8^{2-}$) that can be applied in Fenton-like systems. With the relatively high content organic matter and iron, Fenton sludge was selected to synthesize heterogeneous catalyst/biochar via pyrolysis or hydrothermal carbonization (Zhang et al., 2017; Shen et al., 2020a; Shen et al., 2020b; Belete et al., 2021). In general, Fenton-like processes all follow adsorption and then catalytic oxidation, thereby requiring optimization of the adsorption performance and application of Fenton sludge-based adsorbent to determine the activating different oxidants in Fenton-like processes.

3.3.1 Adsorption

Organic pollutant and heavy metal removal is generally achieved via adsorption due to its simple operation, good selectivity, and given the vast availability of renewable adsorbents such as carbonaceous materials, mineral materials, and metallic materials (Ighalo and Adeniyi, 2020; Zhu et al., 2020). According to previous studies, the good pore structure, high special surface area, and rich functional groups of biochar suggest their applicability as

Table 2 Previous literature on Fenton sludge reuse as iron source systems

Type	Characteristics	References
Dye wastewater; iron-containing sludge electrolytically generates Fe^{2+} via the Fenton process	1. Enhanced COD and color removal; 2. Significantly higher conductivity; 3. Organic material accumulation can be observed; 4. Negative zeta potential	Li et al., 2007
Synthetic olive wastewater, Fenton process with iron source from baked Fenton sludge	1. High calcination temperature shows better organic depletion, higher levels of iron leaching; 2. Biological oxygen demand in five days was increased;	Rossi et al., 2013
Three different wastewaters; ferric sludge acts as an iron source during the Fenton-based process	1. Behaved similar to the CFOP during four reuse cycles; 2. High iron-containing sludge produced during Fenton-based treatment; 3. Lowered hazardous ferric waste production and overall treatment cost	Bolobajev et al., 2014
Palm Oil Mill Secondary Effluent; solar Fenton oxidation resulted in reusable iron sludge	1. Enhanced COD and color removal after five cycles due to excess iron. 2. Lowered COD and color removal observed between recycles 1 and 5;	Shahrifun et al., 2015
Phenolic contained wastewater, Fenton-based treatment with iron source from ferric sludge	1. Enhanced formation of highly reactive species; 2. A substantial organic contaminant degradation increase	Bolobajev et al., 2016a
Chlorophenols-contained water, ferric sludge with tannic acid served as iron source in the Fenton-based process	1. Tannic acid reduced ferric acid to Fe^{2+}; 2. Higher reactivity and lowered Fe^{2+} addition costs, which resulted in decreased sludge production.	Bolobajev et al., 2016b
Palm oil mill secondary effluent, which underwent solar Fenton oxidation with wet and dried Fenton sludge iron source	1. Recycled wet Fenton sludge treatment demonstrated higher removal of contaminant compared to the recycled dried Fenton sludge treatment; 2. Repeated use of recycle sludge resulted in higher SS and turbidity; 3. high-temperature calcination in the sludge reuse system may result in iron leaching	Shahrifun et al., 2016
Landfill leachate, reused ferric oxyhydroxide sludge-activated hydrogen peroxide	1. Lowered applied Fe^{2+} dosage and solid residue; 2. Optimized addition of Fe^{2+} activator can improve the overall efficacy of reuse cycles;	Kattel et al., 2016
Landfill leachate, a pilot study continuous ferric sludge reuse in Fenton-like process	1. Ferric sludge as a catalyst in Fenton-like oxidation resulted in a lower COD removal efficiency of 32% as compared to CFOP; 2. Consistent Fenton-like process efficiencies were observed throughout the 12 sludge reuse cycles.	Klein et al., 2016
Bisphenol A, CaO_2 oxidation catalyzed by reuse of ferric sludge in the presence of chelating agents	1. Presence of oxalic acid resulted in BPA removal of 95.1%; 2. Lowered ferric sludge production and sludge disposal cost.	Zhou et al., 2017
Crepe rubber wastewater and palm oil mill effluent, Fenton oxidation sludge reuse	1. Complete usage of generated sludge resulted in TOC reduction in sludge systems; 2. Ferrous ions and H_2O_2 enhanced the efficiency of the reused sludge system.	Gamaralalage et al., 2017
Agro-food industrial wastewater; ferrous ions reused as catalysts in Fenton-like reactions	Sludge reuse system exhibited lowered residual sludge production and metal content in the final effluent.	Leifeld et al., 2018

adsorbents for contaminant removal (Yi et al., 2020). Based on the remarkable effects of traditional sludge (such as municipal sewage sludge, paper mill sludge) in wastewater treatment for organic pollutants and heavy metals removal by adsorption (Fan et al., 2016; Yoon et al., 2017), researchers have since began to convert Fenton sludge into adsorbents, particularly by applying thermal treatments to transform $Fe(OH)_3$ and organic substances into iron oxide and carbon material for pollutant removal (Tong et al., 2021). Figure 3 presents the Fenton sludge adsorption mechanisms. Similarly, Chu et al applied magnetic biochar (MC) derived from food waste and Fenton sludge via pre-pyrolysis to adsorb organic dyes from aqueous solution. The result indicated MC exhibited a significant removal efficiency for methylene blue (84%) and methyl orange (95%) (Chu et al., 2020). Tong et al. used Fenton sludge to produce aminated hydro-char with a superior adsorption capacity toward Pb^{2+} (359.83 mg/g), which was significantly dependent on the surface complexation, cation-exchange, and electrostatic attraction of the produced hydro-char (Tong et al., 2021).

In general, Fenton sludge is employed to utilize hazardous solid waste via their conversion to adsorbents. However, high adsorbent saturation limits widespread practical applications due to the difficulty and cost of recovery and/or regeneration. High pollutant concentrations and complex wastewater compositions negatively affect the adsorption performance. As such, adsorption

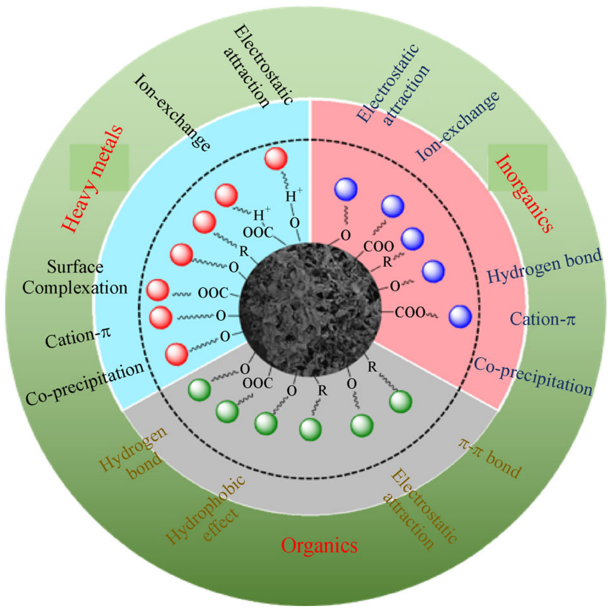

Fig. 3 Adsorption mechanism of Fenton sludge-based adsorbent for pollutants (Ahmad et al. 2014; Leng et al. 2015; Tan et al. 2016; Pan et al. 2021).

alone can not completely remove pollutants from water given the steep stability and reusability requirements of Fenton-based adsorbents. In addition, the adsorption process just transfers pollutants from liquid to solid without reducing its toxicity. As such, further treatment that converts contaminants into harmless matter is necessary for complete contaminant removal from water.

3.3.2 Activation of oxidants

(1) H_2O_2

In general, natural metallic and clay-like matters are widely applied as heterogeneous catalysts during Fenton oxidation. However, these natural metallic substances exhibit instability due to lowered catalytic activity following metal ions release into solution. Similarly, clay-like catalysts have limited active sites and can not completely activate H_2O_2, thus limiting their commercial application. Based on the high iron content in Fenton sludge and efficient catalysis of carbon material on H_2O_2 activation (Ince and Apikyan, 2000; Fang et al., 2014), numerous researchers started to use Fenton sludge as feedstock to convert to heterogeneous catalysts for Fenton oxidation process. Guo et al. amended iron sludge with 0–2 wt% graphene to produce a heterogeneous catalyst. The results indicated that the iron sludge-graphene catalyst exhibited decomposition rates of 99.0%, 98.5%, and 91.8% for rhodamine B, acid red G, and metronidazole, respectively, within 120 min (Guo et al., 2017). Similarly, Zhang et al. developed a heterogeneous, magnetic catalyst ($NiFe_2O_4$) from the co-precipitation of Fenton sludge and $Ni(NO_3)_2$, followed by sintering at 800°C. In addition, the results indicated that an H_2O_2 activated-catalyst with a concentration of 120 mmol/L most effectively produced the highest amount of •OH as well as the highest phenol removal rate of 95% (Zhang et al., 2017). In addition, the catalysts had good reusability and could be regenerated five times. Zhang et al. applied hydrothermal carbonization to mix Fenton sludge and biological sludge, thereby generating a magnetic catalyst with a surface containing Fe_3O_4 particles that were chemically bonded to the carbon structure. Afterward, the magnetic catalyst was applied for methylene blue treatment via Fenton oxidation, to which the results indicated chemical oxygen demand (COD) and total organic carbon (TOC) removal rates of 47% and 49%, respectively (Zhang et al., 2018). Table 3 shows the application of Fenton sludge-based catalysts for H_2O_2 activation.

(2) Activation of persulfate (PS) and peroxymonosulfate (PMS)

In general, $SO_4^{-\bullet}$ exhibits a higher redox potential (E_0 = 2.5–3.1 V) and longer half-life ($t_{1/2}$ = 40 μs) as compared to •OH, which results in effective and sustainable pollutant decomposition (Xiao et al., 2020). Table 4 details the PS and PMS activation methods and radical generation pathways (Wang and Wang, 2018).

In general, PS/PMS activation is achieved via metal activation. However, aqueous solution separation and

Table 3 Application of Fenton sludge-based catalysts for H_2O_2 activation

Feedstock	Synthesis condition	Contaminant	Optimal condition	Optimal removal efficiency	Main ROS	Reusability	Reference
Fenton sludge, graphene	Iron sludge with low amount (0–2 wt%) of graphene	Rhodamine B, acid red G, metronidazole	$[H_2O_2]$ = 10 mmol/L, [Catalyst] = 1 g/L, [pH] = 3.03–9.44	99.0%, 98.5%, 91.8%	•OH	75.8% in the 5th run	Guo et al., 2017
Fenton sludge, $Ni(NO_3)_2$	Co-precipitation, sintering at 800°C	Phenol	$[H_2O_2]$ = 120 mmol/L, [Catalyst] = 2 g/L, [pH] = 3.0, [Phenol] = 250 mg/L	95%±3.4%	•OH	–	Zhang et al., 2017
Fenton sludge, biological sludge	Hydrothermal carbonization	Methylene blue	$[H_2O_2]$ = 1 mL/L, [Catalyst] = 1 g/L, [pH] = 3.0	98%	•OH	85% in the 5th run	Zhang et al., 2018
Fenton sludge, biosolid	Hydrothermal carbonization	Aniline	$[H_2O_2]$ = 60 mmol/L, [Catalyst] = 1 g/L, [pH] = 3.0	77.9%	•OH, $•O_2^-$	–	Zhang et al., 2019a

Table 4 PS and PMS activation methods

Activation method	PS	PMS
Thermal activation	$S_2O_8^{2-} \rightarrow 2SO_4^{-\bullet}$ $SO_4^{-\bullet} + H_2O \rightarrow SO_4^{2-} + HO\bullet + H^+$	$HSO_5^- \rightarrow SO_4^{-\bullet} + HO\bullet$
Base activation	$S_2O_8^{2-} + H_2O \rightarrow 2SO_4^{2-} + HO_2^- + H^+$ $S_2O_8^{2-} + HO_2^- \rightarrow SO_4^{2-} + SO_4^{-\bullet} + O_2^{-\bullet} + H^+$ $SO_4^{-\bullet} + H_2O \rightarrow SO_4^{2-} + HO\bullet + H^+$	$HSO_5^- + H_2O \rightarrow HSO_4^- + H_2O_2$ $H_2O_2 + HO^- \rightarrow H_2O + HO_2^-$ $H_2O_2 \rightarrow 2HO\bullet$ $HO_2^- + H_2O_2 \rightarrow HO\bullet + H^+ + O_2^{-\bullet}$ $HSO_5^- + HO_2^- \rightarrow H_2O + SO_4^{-\bullet} + {}^1O_2$
UV activation	$S_2O_8^{2-} \rightarrow 2SO_4^{-\bullet}$ $H_2O \rightarrow HO\bullet + H\bullet$ $S_2O_8^{2-} + H\bullet \rightarrow SO_4^{-\bullet} + SO_4^{2-} + H^+$	$HSO_5^- \rightarrow SO_4^{-\bullet} + HO\bullet$ $H_2O \rightarrow HO\bullet + H\bullet$ $HSO_5^- + H\bullet \rightarrow SO_4^{-\bullet} + H_2O$
Metal activation	$S_2O_8^{2-} + M^n \rightarrow M^{n+1} + SO_4^{-\bullet} + SO_4^{2-}$	$HSO_5^- + M^n \rightarrow M^{n+1} + SO_4^{-\bullet} + OH^-$
Carbon activation	$S_2O_8^{2-} + e^- \rightarrow SO_4^{-\bullet} + SO_4^{2-}$	$HSO_5^- + e^- \rightarrow SO_4^{-\bullet} + OH^-$

recovery as well as the potential risk of generating a secondary pollution limit metal application in homogeneous activation. Heterogeneous Fenton-like processes have been more recently reported, particularly Fenton sludge-based catalysts generated via continuous thermal treatment and/or modification, which have shown effectiveness in activating PS/PMS (Zhou and Zhang, 2017; Shen et al., 2020b; Shen et al., 2020a). Following thermal treatment, Fenton sludge can transform into Fe_2O_3/γ-Fe_2O_3-containing carbon material/biochar with defective sites, persistent radicals, and highly graphitized carbon structures (Shen et al., 2020b). As such, synthesized Fenton sludge-based heterogeneous catalysts have shown the ability to effectively activate PS/PMS activation due to their high transition metal iron content and abundance of O- and N-containing functional groups (Zhu et al., 2018).

Previous studies have reported the ability of Fenton sludge-based catalyst to adsorb organic pollutants via hydrogen bonding and π-π electron pairing (Guo et al., 2017; Shen et al., 2020a). As a result, free radicals are produced due to oxidant activation, which allows Fenton sludge-based catalyst activation to proceed for simultaneous contaminant adsorption and decomposition. Shen et al. developed a technique to produce Cu-containing Fenton sludge as efficient catalysts for PS activation and the subsequent degradation of tetracycline (TC). The results indicated a TC degradation efficiency of 97.74% after 120 min with 0.2 g/L catalyst and 100 mg/L PS. When using catalyst alone, the TC removal efficiency was 62.89%, indicating the synergistic effect of adsorption and degradation from catalyst plays a dominant role in the PS activation process (Shen et al., 2020a). Shen et al. produced a Fenton-like catalyst from Ni-containing Fe sludge via one-step calcination, to which the catalyst was tested regarding its TC removal efficiency. Low TC removal (maximum removal ≈ 15%) was observed with both the Fenton sludge-based catalyst and PMS. However, the application of the catalyst-activated PMS process resulted in higher removal rates above 70% with 20 mg/L of PMS and 0.2 g/L of catalyst.

The reaction mechanism of the Fenton-based catalyst/oxidants process could be summarized as follows (Fig. 4): 1) pollutants and oxidants are adsorbed on the surface of Fenton-based catalyst; 2) electrons are provided to fracture the relative "weak bond" of oxidant to generate ROS such as $SO_4^{-\bullet}$, •OH; and 3) pollutants are decomposed by radicals in situ. In addition, other ROS such as O_2^- and ${}^1O_2$ were continuously generated in the solution matrix, all of which aided in contaminant degradation (Meng et al., 2020). Previous studies have categorized the catalysts into three types of catalyst reaction sites for oxidant activation: 1) transition metals were the main active centers on Fenton-based catalyst; 2) graphitized structure and unsaturated C on the internal and edge of catalyst; 3) the oxygen vacancies of the Fenton-based catalyst. Shen et al. indicated that the Fenton-based catalyst efficiency was highly dependent on the catalyst transition metals (Fe, Cu) and oxygen vacancies, which enhanced the electron transfer (Shen et al., 2020a). Shen et al. evaluated the removal of tetracycline by a Fenton sludge-based catalyst/PMS process, and reported that carbon structure also played a significant role in the activation (Shen et al., 2020b).

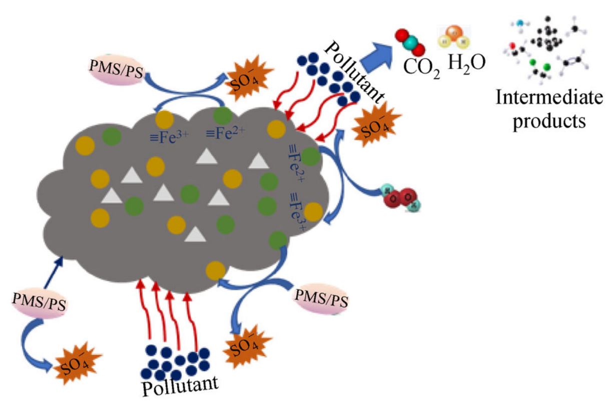

Fig. 4 Illustration of PS/PMS activation.

3.4 Other applications

In addition to participate in Fenton reaction and synthetic heterogeneous catalyst, some researchers found the ferric sludge could also be used as a sludge conditioning agent, electron acceptor during anaerobic digestion, skeleton material for dewatering, and potential phosphorus fertilizer. Xu et al. combined Fenton iron sludge and $NH_3 \cdot H_2O$ to produce a skeleton material, which enhanced sewage sludge dewatering via hydrothermal treatment (Xu et al., 2019). Fe(III) oxides, as an ideal potential for improving the anaerobic digestion performance. Wang et al. introduced the iron-containing sludge in an anaerobic digestion (AD) process to enhance digestion efficiency as well as to reduce organic pollutant in the Fenton sludge (Wang et al., 2018; Wang et al., 2019). Results suggested that the AD performance was significantly enhanced, and more than 70% organic matter was removed from the Fenton sludge. Meanwhile, nearly half of iron ion was reduced to Fe^{2+} through the digestion process, which suggests an Fe recycling between AD and Fenton oxidation process. Wang et al. used iron-rich biochar pyrolyzed from Fenton sludge to adsorb P from the liquid phase of the AD process, then the recovered P with biochar was re-utilized as a P-fertilizer in garden soil (Wang et al., 2020).

4 Recommendation and summary

The present review summarized current Fenton sludge reuse systems that have overcome classical Fenton process restrictions while simultaneously reducing the overall process cost and secondary pollution generation. Previous studies have reported Fenton sludge reuse systems-based studies, though most have been limited to the laboratory stage. This may be because 1) the high water contained in Fenton sludge is a big obstacle to utilization; 2) Fenton sludge has complex composition and small size; 3) preparation of coagulant might be a big step to consume quantities of Fenton sludge, but the presence of heavy metals and radioactivity in Fenton sludge would limit its commercial application; 4) due to the environmental and safety concerns, customers may not want to accept synthetic products from waste material.

Fenton sludge processes benefitted most from a synergistic and synthetic heterogeneous catalyst given that oxidant activation (H_2O_2, PS, and PMS) by the catalyst enhanced the multiple ROS formation through various activation mechanisms. However, the heavy metals in Fenton sludge greatly affect the stability and biotoxicity of the composites. As such, future studies can explore the interaction of synthetic catalysts with the environment as well as its potential ecotoxicity. In addition, current studies have been limited to Fenton sludge-based catalyst applications toward simulated (i.e., artificial) wastewater. Selective contaminant removal by the produced synthetic catalyst must be further examined to address the complexities of pollutants in actual wastewater. In addition, additional processes as pre/post-treatment options may be applied with the heterogeneous catalysis system, such as the combined treatment of a biological (aerobic and anaerobic) treatment system and adsorption process, to produce a real zero liquid discharge.

5 Conclusions

This review presented the Fenton sludge properties and various resource utilization methods, particularly with regards to Fenton sludge applications in heterogeneous catalyst synthesis. Fenton sludge-based catalysts show great promise for the removal of contaminants from aqueous solution. However, the impact of synthetic catalysts on the surrounding environment and practical application for actual wastewater treatment should be further investigated due to the presence of heavy metals in Fenton sludge. This review may be helpful to researchers engaged in Fenton oxidation treatment of refractory wastewater, particularly in the field of Fenton sludge reuse systems.

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