



## Review

## Principle and application of hydrogen peroxide based advanced oxidation processes in activated sludge treatment: A review



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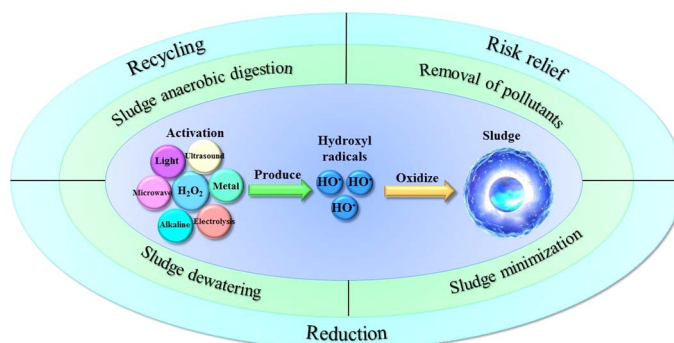
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## HIGHLIGHTS

- Sludge dewatering and minimization can be achieved by H<sub>2</sub>O<sub>2</sub> based AOP.
- Methane yield in sludge anaerobic digestion can be improved prominently by H<sub>2</sub>O<sub>2</sub> based AOP.
- Pollutants retained in sludge can be degraded effectively via strong oxidative hydroxyl radical.
- Cost estimation of H<sub>2</sub>O<sub>2</sub> based AOP in sludge treatment has been materialized.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

## Keywords:

Activated sludge  
H<sub>2</sub>O<sub>2</sub>  
Advanced oxidation processes  
Sludge treatment

## ABSTRACT

The disposal of excessive activated sludge by advanced oxidation processes (AOP) to avoid its potential environmental risk has received extensive attention. As an efficient and clean oxidant, hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) has been used widely in AOP. In H<sub>2</sub>O<sub>2</sub> based AOP, strong oxidative hydroxyl radical (HO<sup>•</sup>) produced by different kinds of activation methods plays an important role in sludge treatment for sludge reduction, recycling and risk relief. This review summarizes current studies and breakthroughs of H<sub>2</sub>O<sub>2</sub> based AOP in sludge treatment. Meanwhile, the possible mechanisms of H<sub>2</sub>O<sub>2</sub> based AOP in sludge dewatering, sludge minimization, anaerobic digestion and the removal of pollutants are clarified systematically. Furthermore, the cost estimation of H<sub>2</sub>O<sub>2</sub> based AOP in sludge treatment is materialized. Finally, the possible focus of future study in sludge treatment via H<sub>2</sub>O<sub>2</sub> based AOP is present. This review can provide a theoretical basis for applying H<sub>2</sub>O<sub>2</sub> based AOP in sludge reduction, recycling and risk relief.

## 1. Introduction

With the wide application of activated sludge process in wastewater treatment, the generation of excessive activated sludge is inevitable [1]. The high water content (exceed 95%) of sludge makes its transportation, storage and handling become inconvenience [2]. Besides, sludge is

a heterogeneous and colloidal complex with high concentration of solid and organic matters, which can be utilized as a renewable source for energy recovery [3]. Moreover, hazardous pollutants such as endocrine disrupting compounds (EDC), tetracyclines (TC), bis (2-ethylhexyl) phthalate (DEHP), nonylphenol (NP), pathogens (PG) and heavy metals are retained in sludge, which may lead to serious environmental risks

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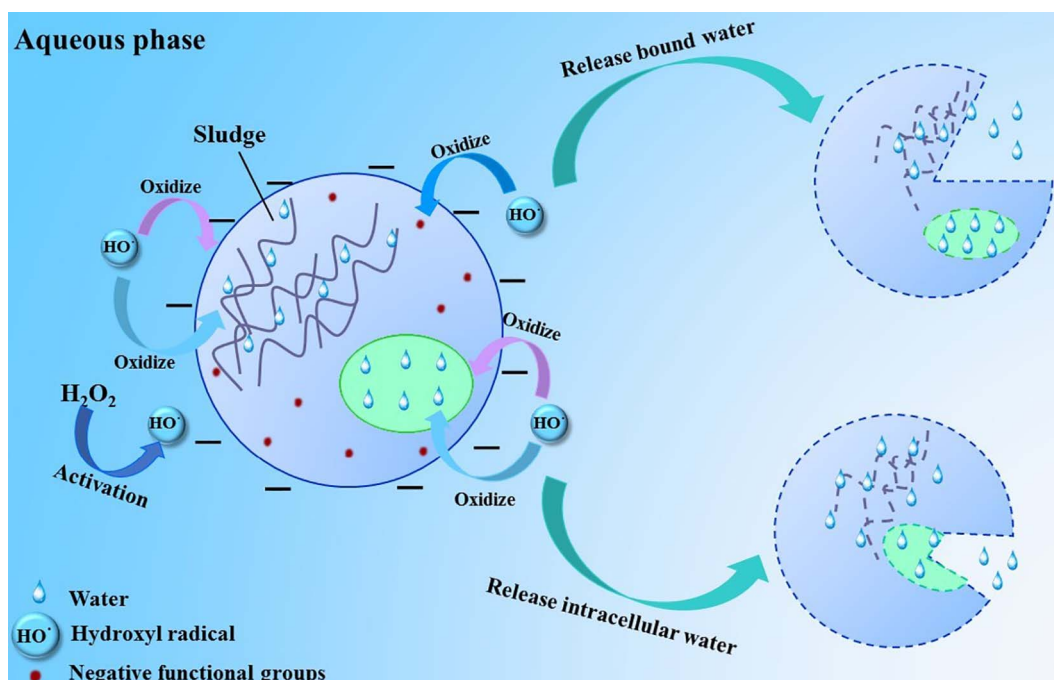


Fig. 1. Mechanism of H<sub>2</sub>O<sub>2</sub> based AOP in sludge dewatering.

[4–8]. In addition, the elimination of odors released from sludge to protect the air environment is notable. Hence, taking corresponding measurements for sludge disposal is urgently [9,10].

In order to realize sludge reduction, recycling and risk relief, different kinds of methods such as advanced oxidation processes (AOP), physical treatment, biological treatment and pyrogenic decomposition have been employed in sludge dewatering [11], sludge minimization [12], thermal drying [13], anaerobic digestion, removal of pollutants [14,15], pelletization for fuel preparation [16,17] and liquefaction for bio-oil refining [18,19]. Among these, AOP have presented excellent performance and achieved extensively attention. In AOP, a variety of oxidants such as hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), persulfate, permanganate and ozone (O<sub>3</sub>) have been used in sludge treatment. Herein, H<sub>2</sub>O<sub>2</sub> has received widely attention according to its high efficiency and environmental friendliness [20–22].

After activation by transition metals, microwave (MW), ultrasound (US), electrolysis and light irradiation, H<sub>2</sub>O<sub>2</sub> can be converted into high oxidative hydroxyl radical (HO·) [23–26]. In sludge dewatering, HO· generated in H<sub>2</sub>O<sub>2</sub> based AOP leads to the destruction of sludge floc and cell membranes. Hence, interstitial water and intracellular water are released from sludge, which contribute to the improvement of sludge dewaterability [27]. Besides, extracellular polymeric substances (EPS) and some recalcitrant organic compounds are converted into soluble organics or mineralized to carbon dioxide (CO<sub>2</sub>) and water (H<sub>2</sub>O), thus improving sludge minimization [28]. In anaerobic digestion, HO· can oxidize recalcitrant macromolecular organics to smaller soluble organics, which is important for improving sludge hydrolysis efficiency. Then, abundant soluble organics can be utilized by bacteria and enzymes in acidification and methanogenesis phase, and methane yield is improved significantly [29]. Besides, H<sub>2</sub>O<sub>2</sub> based AOP have great performances in the removal of pollutants. Refractory organic pollutants retained in sludge can be degraded effectively by oxidative HO· [30–32]. In addition, the destruction of sludge floc by HO· can promote the transformation of heavy metals from binding state to free state. Hence, heavy metals are released from sludge into aqueous phase [33]. Meanwhile, odors released from sludge can be eliminated by H<sub>2</sub>O<sub>2</sub> based AOP.

The potential of H<sub>2</sub>O<sub>2</sub> based AOP in sludge treatment to realize sludge reduction, recycling and risk relief have been demonstrated

thoroughly. Therefore, this review concludes breakthroughs in the application of H<sub>2</sub>O<sub>2</sub> based AOP for sludge treatment. Besides, possible mechanisms of H<sub>2</sub>O<sub>2</sub> based AOP in sludge reduction, recycling and risk relief are clarified comprehensively. Moreover, the possible focus of future research in H<sub>2</sub>O<sub>2</sub> based AOP for sludge treatment is presented. The cost estimation of H<sub>2</sub>O<sub>2</sub> based AOP in sludge treatment is materialized. This review can provide a theoretical basis and guidance for the application of H<sub>2</sub>O<sub>2</sub> based AOP in sludge treatment.

## 2. H<sub>2</sub>O<sub>2</sub> based AOP in sludge dewatering and minimization

### 2.1. Sludge dewatering

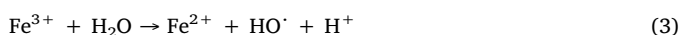
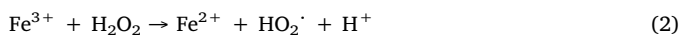
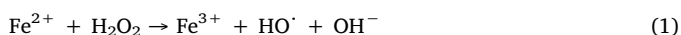
Sludge dewatering is a bottleneck in sludge treatment and disposal process [34]. Sludge dewatering is very difficult due to EPS playing an important role in binding water. EPS are metabolic products consisted of proteins, polysaccharides and small amounts of DNA and lipids [35,36], which accumulate on the bacterial cell surface and form a protective gel-like reservoir for water preservation [37]. The negatively charged EPS network formed by the ionization of negative functional groups such as carboxyl, hydroxyl and amino could maintain hydrated sludge structure and prevent the release of water. Besides, high negative surface charge density can prevent sludge destabilization and flocculation via electrostatic repulsion [38]. H<sub>2</sub>O<sub>2</sub> based AOP can improve sludge dewatering effectively. Strong oxidative HO· formed in H<sub>2</sub>O<sub>2</sub> based AOP can oxidize sludge floc and EPS. The interstitial water entrapped inside the sludge flocs and water combined with EPS are released from sludge because of the destruction of sludge floc and EPS. Besides, functional groups in sludge EPS are oxidized by HO·, which reduces the degree of  $\pi$ -electron systems, decreases the number of aromatic rings and conjugated bonds in a chain structure and converses a linear ring system to a non-linear ring system [39,40]. Moreover, the collapse of EPS results in the destruction of sludge matrix, and the oxidation of microorganism cells becomes easier [36]. The HO· penetrates the microorganism walls, improves the cell membranes permeability and breaks the cell walls uniformly, thus intracellular water in sludge cell is released [27]. All these contribute to the improvement of sludge dewaterability. Mechanism of H<sub>2</sub>O<sub>2</sub> based AOP in sludge dewatering is shown in Fig. 1. The effects of H<sub>2</sub>O<sub>2</sub> based AOP in sludge

**Table 1**  
The effects of H<sub>2</sub>O<sub>2</sub> based AOP in sludge dewatering.

Methods	Dosage				Raw sludge Wc (%)	Dewatering parameters			Ref
	H <sub>2</sub> O <sub>2</sub>	Catalyst	pH	Other		Wc of sludge (%)	CST reduction (%)	SRF reduction (%)	
Fenton process	5 g/L	Fe <sup>2+</sup> : 6 g/L	4	–	97.92	–	48.52	93.29	[55]
Fenton process	21 mg/g DS	Fe <sup>2+</sup> : 105 mg/g DS	6	–	–	–	48 ± 3	–	[56]
Fenton-skeleton builders process	33.8 mg/g DS	Fe <sup>2+</sup> : 40.3 mg/g DS	5	OPC: 350.0 mg/g; Lime: 450.0 mg/g	95.9–96.6	49.54	–	–	[34]
Fenton-skeleton builder process	50 mg/g DS	Fe <sup>2+</sup> : 30 mg/g DS	3	Lime: 50.0 mg/g DS	96.2 ± 0.6	55.1 ± 0.6	–	96.0	[46]
Fenton-skeleton builder process	47.9 mg/g DS	Fe <sup>2+</sup> : 34.3 mg/g DS	7	Lime: 43.2 mg/g DS	96.8	55.8 ± 0.6	–	–	[57]
Fenton-skeleton builder process	31.9 mg/g DS	Fe <sup>2+</sup> : 33.7 mg/g DS	5	Red mud: 275.1 mg/g DS	96.4–97.2	59.8 ± 0.4	–	–	[58]
Fenton-surfactant process	40 mg/g DS	Fe <sup>2+</sup> : 40 mg/g DS	4	DDBAC: 60.0 mg/g	95.11 ± 0.24	57.17	90.47	–	[48]
Fenton-like process	373 mg/g DS	Fe <sup>3+</sup> : 288 mg/g DS	2	–	80.0 ± 1.5	66.1	–	74.88	[2]
Fenton-like process	250 mg HP/L	ZVI: 500 mg ZVI/L	2	–	–	–	50	–	[59]
US-Fenton-process	0.25 g/L	Fe <sup>2+</sup> : 0.2 g/L	3	US time: 1.5 h	98.8	–	84.95	97.04	[53]

dewatering are summarized in Table 1.

Capillary suction time (CST), specific resistance to filterability (SRF) and water content (W<sub>c</sub>) are main parameters in the evaluation of sludge dewaterability. Fenton process can enhance sludge dewatering effectively. In Fenton process, Fe<sup>2+</sup> ions catalyze the decomposition of H<sub>2</sub>O<sub>2</sub> for producing HO· (Eq. (1)). The dissolution and decomposition of EPS were happened synchronously by the oxidation of HO· [41]. Meanwhile, Fe<sup>3+</sup> ions can initiate a Fenton-like process, following the formation of Fe<sup>2+</sup> ions for Fenton process (Eq. (2)). Besides, Fe<sup>2+</sup> and HO· are produced in the reaction between Fe<sup>3+</sup> and H<sub>2</sub>O (Eq. (3)) [42]. Liu et al. investigated the effect of Fenton process in sludge dewatering. The W<sub>c</sub> of sludge cake decreased from 86.2% to 75.2% at pH 3 with 6 mg/L of Fe<sup>2+</sup> and 3 mg/L of H<sub>2</sub>O<sub>2</sub> [43]. Besides, the change of pH can influence the effect of Fenton process in sludge dewatering. As shown in the research of Zhang et al., the CST value at pH 4 was 157 s [41]. However, when pH increased from 4 to 7, CST value increased from 157 s to 200 s. This increasing trend reflected the deterioration of sludge dewaterability, which was corresponded with the tendency of excitation-emission matrix (EEM) profile of soluble EPS (S-EPS) (Fig. 2). According to the research of He et al., a Fenton-like process via Fe<sup>3+</sup> activating H<sub>2</sub>O<sub>2</sub> has achieved great results in sludge dewatering [2]. The specific reactions are shown as Eqs. (2) and (3). The oxidative radicals generated in Fenton-like process leads to the dissolution of EPS and lysis of sludge cells, thus bound water is transformed into free water and released from sludge eventually. The W<sub>c</sub> of sludge cake decreased from 80.0% to 66.1% at pH 2 with 288 mg/g dry solid (DS) of Fe<sup>3+</sup> and 373 mg/g DS of H<sub>2</sub>O<sub>2</sub> [2].



Besides, skeleton builders such as lime, cement and red mud have been applied in H<sub>2</sub>O<sub>2</sub> based AOP to assist sludge dewatering. After the addition of skeleton builders, the caducity and death of microorganism are enhanced owing to the constantly-changing environment. The plate-like structure of sludge disappears significantly, whereas irregular holes are formed. The conversion of bound water into free water and the reduction of sludge particle size are improved by skeleton builders inlayed or pierced in microbial cells [34]. Moreover, a permeable and rigid lattice structure is formed in sludge cake under high pressure, which leads to the improvement of sludge dewaterability [44,45]. Mo

et al. studied the effect of Fenton-lime process in sludge dewatering. In Fenton process at pH 3 with 50 mg/g dry solids (DS) of Fe<sup>2+</sup> and 50 mg/g DS of H<sub>2</sub>O<sub>2</sub>, the reductions of CST and SRF were 37% and 21%, respectively. After the addition of lime, the cooperation of lime and Fenton process was favorable for improving sludge dewatering. When the dosage of lime was 50 mg/g DS, the reductions of CST and SRF were increased to 68% and 96%, respectively. Besides, the W<sub>c</sub> of sludge cake decreased from 96.2 ± 0.6% to 55.1 ± 0.6% [46]. In the study of Liu et al., the reduction of SRF reached 93% distinctly after the addition of ordinary portland cement (OPC) in Fenton process at optimum condition [10]. Surfactants also have great performances in assisting H<sub>2</sub>O<sub>2</sub> based AOP for sludge dewatering. Surfactants can break the binding between the loosely bound EPS (LB-EPS) and tightly bound (TB-EPS) of sludge floc by neutralizing the negative charge in sludge surface. Bound water combined with EPS is released from sludge into aqueous phase. Besides, LB-EPS and TB-EPS are turned into S-EPS by the high hydration ability of surfactants. Hence, interstitial water entrapped inside the sludge flocs and the water held inside the sludge are also released [47]. The sludge dewaterability could be improved effectively by the cooperation of oxidative HO· and surfactants. In the research of Chen et al., the effects of Fenton and Fenton-surfactant process in sludge dewatering were compared. In Fenton process at pH 4 with 40 mg/g DS of Fe<sup>2+</sup> and 40 mg/g DS of H<sub>2</sub>O<sub>2</sub>, the W<sub>c</sub> was decreased to 63.36% and approximately 48% bound water was released. After the addition of 60 mg/g DS of dodecyl dimethyl benzyl ammonium chloride (DDBAC; A kind of surfactants), the W<sub>c</sub> was decreased to 57.17% and approximately 68% bound water was removed [48].

US can cooperate with Fenton process for assisting the activation of H<sub>2</sub>O<sub>2</sub> and sludge dewatering. In US-Fenton process, the decomposition of H<sub>2</sub>O<sub>2</sub> and H<sub>2</sub>O are promoted by US, which are favorable to the production of HO· (Eqs. (4) and (5) [49]. The Fenton process is accelerated by the cavitation effect, which promotes the transformation of Fe<sup>3+</sup> to Fe<sup>2+</sup>. Consequently, the generated Fe<sup>2+</sup> continue to initiate Fenton process for producing HO· [50]. Besides, local extra high temperatures, sharp discharging, high pressure and superspeed jet flows are generated to destroy microbial cells and alter sludge characteristics for sludge dewatering [51,52]. In US-Fenton process, the dosages of Fe<sup>2+</sup> and H<sub>2</sub>O<sub>2</sub> and reaction time required in Fenton process can be reduced about 66.7%, 75.0%, and 75.0%, respectively. After 1.5 h reaction at pH 3 with 0.4 g/L of Fe<sup>2+</sup> and 0.25 g/L of H<sub>2</sub>O<sub>2</sub>, the reductions of CST and SRF were 84.95% and 97.04%, respectively [53]. In Fenton process, the similar results were achieved after 6 h reaction with 0.6 g/L of Fe<sup>2+</sup> and 1.0 g/L of H<sub>2</sub>O<sub>2</sub>. O<sub>3</sub> can be used to promote the

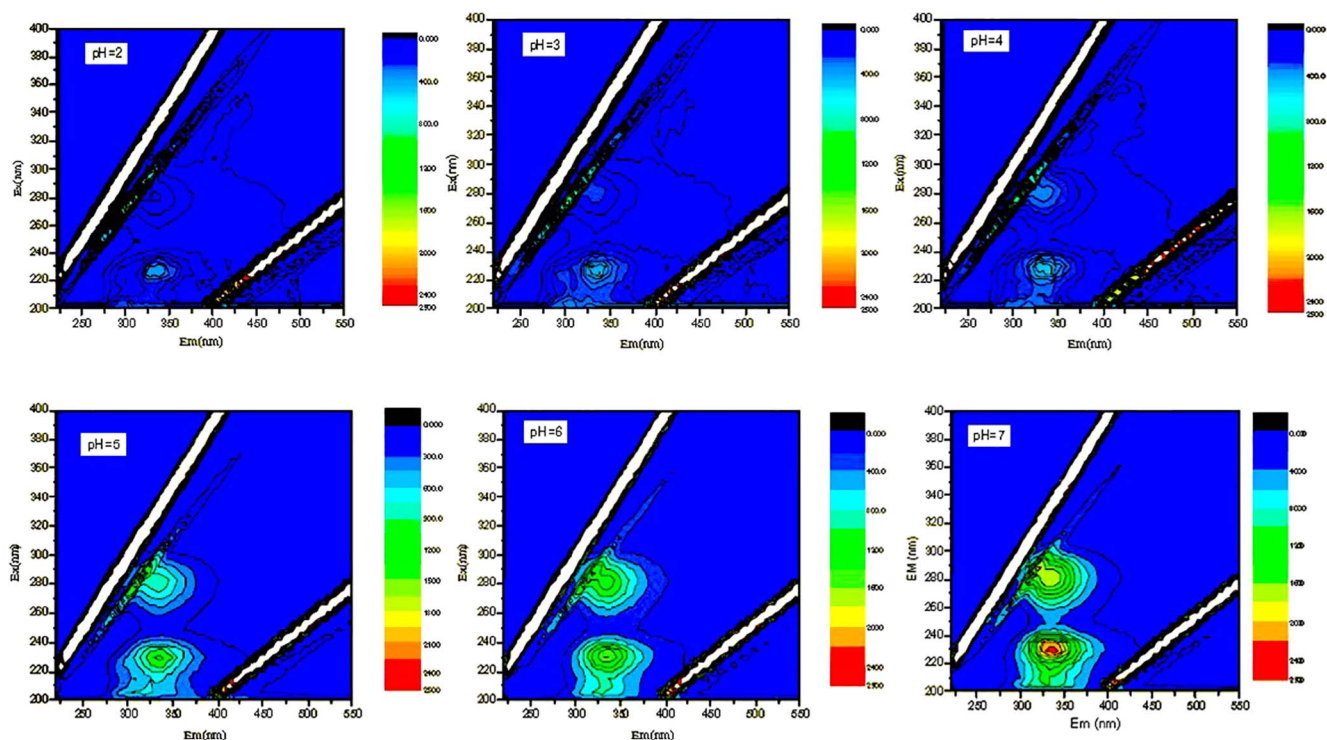


Fig. 2. Influence of pH on EEM profile of soluble EPS after Fenton conditioning ( $[\text{Fe}^{2+}]/[\text{H}_2\text{O}_2]$ (mol/mol) = 0.1; 30%  $\text{H}_2\text{O}_2$  = 0.3% (v/v) [41]. Copyright 2015 Elsevier.

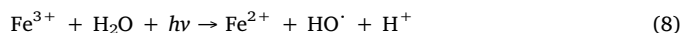
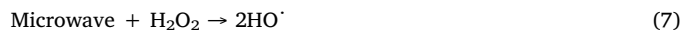
decomposition of  $\text{H}_2\text{O}_2$  and the production of  $\text{HO}^\cdot$  (Eq. (6)) [54]. However,  $\text{O}_3$ - $\text{H}_2\text{O}_2$  process only slight improved sludge dewaterability, and the filterability of sludge was deteriorated. The  $W_c$  decreased from 94.6% to 89.8% and the CST value increased from 17.2 s to 32.9 s when the dosage of  $\text{O}_3$  was 83 mg/g mixed-liquor suspended solids (MLSS) [20].



## 2.2. Sludge minimization

Sludge minimization can be achieved by  $\text{H}_2\text{O}_2$  based AOP through sludge disintegration, solubilization and mineralization [60]. In  $\text{H}_2\text{O}_2$  based AOP, sludge structure is destroyed by oxidative  $\text{HO}^\cdot$ , which leads to sludge disintegration. Under the oxidation of  $\text{HO}^\cdot$ , EPS are degraded and transformed to soluble organics. Besides,  $\text{HO}^\cdot$  kills most micro-organisms and destroys cell walls. Intracellular contents are released from cells into aqueous phase. Meanwhile, some recalcitrant organic compounds contained in sludge are oxidized into soluble organics or mineralized to  $\text{CO}_2$  and  $\text{H}_2\text{O}$ . These jointly contribute to sludge minimization [28]. Mechanism of  $\text{H}_2\text{O}_2$  based AOP in sludge minimization is shown in Fig. 3. In sludge minimization, the oxidation of  $\text{HO}^\cdot$  leads to the decrease of total suspended solids (TSS), volatile suspended solids (VSS) and total chemical oxygen demand (TCOD), whereas soluble COD (SCOD) shows an increasing trend according to the increase of soluble organics. The effects of  $\text{H}_2\text{O}_2$  based AOP in sludge minimization are summarized in Table 2. In the study of Cho et al., Fe/MgO catalyst was synthesized for  $\text{H}_2\text{O}_2$  activation [61]. Fe/MgO catalyst can provide  $\text{Fe}^{2+}$  ions for  $\text{H}_2\text{O}_2$  activation, which is similar to Fenton process. In  $\text{H}_2\text{O}_2$  process with 1:10 of sludge: $\text{H}_2\text{O}_2$  ratio, the SCOD content was 900 mg/L, and the reductions of TCOD, TSS and VSS were 2.1%, 7.8% and 7.7%, respectively. In Fe/MgO- $\text{H}_2\text{O}_2$  process with 1:10:5 of sludge: $\text{H}_2\text{O}_2$ :Fe/MgO ratio, the SCOD content increased to

1480 mg/L, and the reductions of TCOD, TSS and VSS both reached 27.4%, 33.9% and 33.9%, respectively [61]. Wang et al. investigated the effect of MW- $\text{H}_2\text{O}_2$  process in sludge minimization [62]. MW can activate  $\text{H}_2\text{O}_2$  and assist sludge minimization. Heat and irradiation in this process not only decompose  $\text{H}_2\text{O}_2$  to  $\text{HO}^\cdot$  (Eq. (7)), but also promote sludge solubilization [12]. EPS are destroyed and detached from sludge under MW irradiation. Meanwhile,  $\text{HO}^\cdot$  reacts swiftly with EPS, which is decomposed to soluble organics or  $\text{CO}_2$  and  $\text{H}_2\text{O}$ . The original SCOD content in sludge was 37.77 mg/L. When temperature was 373 K and the dosage of  $\text{H}_2\text{O}_2$  was 23.4 g/L, the SCOD content increased to 2082.25 mg/L, and the TCOD reduction was 47.56% prominently [62]. Tokumura et al. studied the photo-Fenton process in sludge minimization [63]. In photo-Fenton process, light is used to assist Fenton process for the production of  $\text{HO}^\cdot$ . Photo-Fenton process contains two reactions. The first reaction is the typical Fenton reaction, in which  $\text{H}_2\text{O}_2$  is activated by  $\text{Fe}^{2+}$ . In the second reaction,  $\text{Fe}^{3+}$  reacts with  $\text{H}_2\text{O}$  to produce  $\text{HO}^\cdot$  under the irradiation of light. The  $\text{HO}^\cdot$  is produced and  $\text{Fe}^{3+}$  is reduced to  $\text{Fe}^{2+}$  for activating  $\text{H}_2\text{O}_2$  (Eq. (8)) [64]. In photo-Fenton process at 60 mg/L of Fe and 4000 mg/L of  $\text{H}_2\text{O}_2$ , 40% reduction of MLSS was achieved after 2.5 d reaction [63]. However, the dosage of  $\text{H}_2\text{O}_2$  need to be controlled. Excessive  $\text{H}_2\text{O}_2$  could consume  $\text{HO}^\cdot$  to produce hydroperoxyl radical ( $\text{HO}_2^\cdot$ ) (Eq. (9)). Although  $\text{HO}_2^\cdot$  also can oxidize EPS, its oxidation capability is much lower than  $\text{HO}^\cdot$  [65,66].



Alkaline condition can promote sludge dissolution. In alkaline- $\text{H}_2\text{O}_2$  process, the oxidation of  $\text{HO}^\cdot$  and the hydrolysis of alkaline condition leads to sludge destruction and dissolution. Hence, sludge particles decreased and soluble organics in aqueous phase increased, which contribute to the improvement of sludge minimization [67]. In the study of Kim et al., the TS reduction was 33% and SCOD/TCOD ratio was 54.7% at pH 6.6. When pH was adjusted to 11, the TS reduction increased to 49% and SCOD/TCOD ratio increased to 57.4% [67].

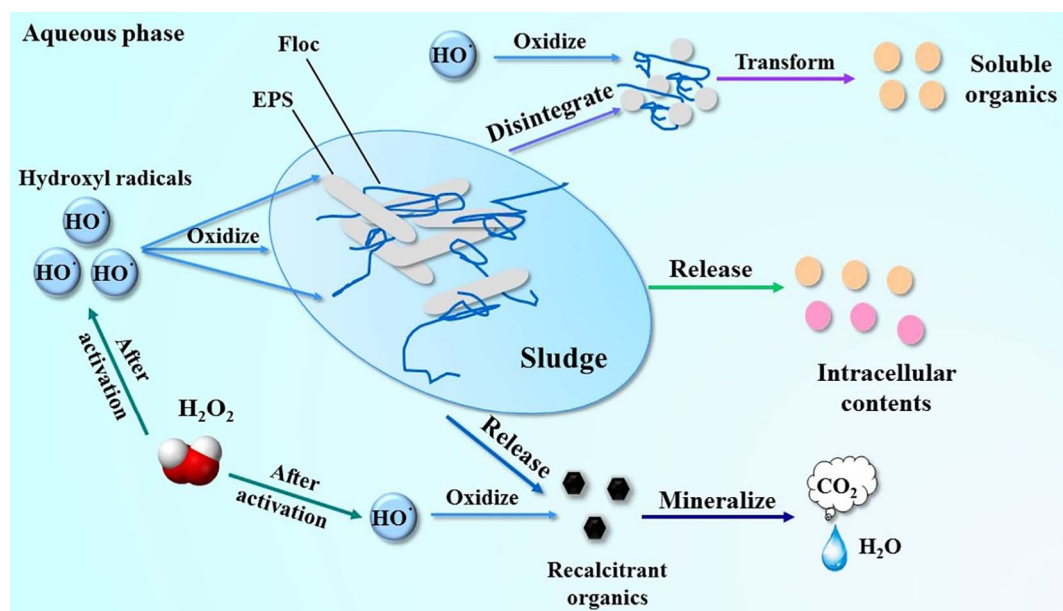


Fig. 3. Mechanism of activated  $H_2O_2$  in sludge minimization.

**Table 2**  
The effects of  $H_2O_2$  based AOP in sludge minimization.

Method	$H_2O_2$ dosage	Change of parameters in sludge minimization	Ref
Alkaline- $H_2O_2$ process	1.6 M	TS: -33%; SCOD/TCOD: +55%	[67]
MW- $H_2O_2$ process	23.4 g/L	TCOD: -47.56%; TSS: -42%	[62]
Fe/MgO- $H_2O_2$ process	3.4 mL	TSS: -34%; VSS: -34%; TCOD: -27.4%	[61]
Fenton process	0.5 g/g SS	COD: -72%; SS: -53%; VSS: -63%	[71]
Electro-Fenton process	127 mmol/L	COD: -72%	[60]
Photo-Fenton process	4 g/L	MLSS: -40%	[63]
US-Fenton process	9 g/L	TCOD: -79%	[50]

The symbol of “+” implies the increase of parameters, whereas the symbol of “-” implies the decrease of parameters.

Zhao et al. reported the great efficient of  $O_3$ - $H_2O_2$  process in sludge minimization [68]. In  $O_3$ - $H_2O_2$  process,  $HO\cdot$  can improve sludge dissolution by decomposing sludge EPS, which promotes the emergency of a large number of soluble organics in aqueous phase. These organics can be utilized as carbon sources to improve the cell decay rate for sludge minimization [69,70]. In  $O_3$  process at 290.5 mg/g of  $O_3$ , the SCOD content was 1460 mg/L. When 436 mg/g TS of  $H_2O_2$  was added, the SCOD content increased to about 1870 mg/L [68].

### 3. $H_2O_2$ based AOP in sludge anaerobic digestion

Anaerobic digestion is regarded as a promising option for sludge recycling [72]. The hydrolysis stage of anaerobic digestion is regarded as the rate-limiting step in sludge degradation and the consequent methane production. In this stage, microorganisms show low performance in hydrolyzing sludge solids such as flocs, EPS and recalcitrant compounds [73]. Hence,  $H_2O_2$  based AOP have been introduced in anaerobic digestion as a pretreatment to accelerate this slow process and improve anaerobic digestion efficiency [74]. The  $HO\cdot$  generated in  $H_2O_2$  based AOP disintegrates sludge and destroys sludge flocs. As a result, the break of the microbial cell membrane leads to the decomposition of cell walls, and both extracellular and intracellular components are released into aqueous phase [75,76]. Besides, EPS and some

macromolecular organic matters are degraded and transformed to soluble organics [77]. All these contribute to the improvement of sludge dissolution. Hence, abundant soluble organics generated in hydrolysis stage can be utilized easily by bacteria in the following acidogenesis and methanogenesis stage for producing methane [67]. Mechanism of  $H_2O_2$  based AOP in sludge anaerobic digestion is shown in Fig. 4. The effects of  $H_2O_2$  based AOP in sludge anaerobic digestion are summarized in Table 3.

$H_2O_2$  can be activated by indigenous iron contained in sludge. After introducing  $H_2O_2$  in activated sludge with high iron content, cell and EPS are destroyed effectively by  $HO\cdot$  generated in indigenous iron- $H_2O_2$  process (Fenton like process). Meanwhile, massive soluble organics are released to aqueous phase, which leads to the increase of SCOD content in aqueous phase. The SCOD content of sludge after pretreatment increased to  $103 \pm 7$  mg/g DS at pH 2 with 50 mg/g DS of  $H_2O_2$ , which was only  $8 \pm 1$  mg/g DS in original sludge. The increase of SCOD content contributed to the improvement of sludge methane potential. When compared with the case without pretreatment, the sludge methane potential with pretreatment was improved 13% after 16 d digestion [78]. Free nitrous acid (FNA), a strong biocidal agent, can destroy microbial species and waste water biofilms to accelerate sludge anaerobic digestion. In FNA- $H_2O_2$  process, oxidative peroxyxynitrite, nitrogen dioxide radical and  $HO\cdot$  are produced in the reaction between  $H_2O_2$  and FNA, which oxidize the cell components and EPS to soluble organics for further utilization [22,79,80]. The SCOD content in FNA- $H_2O_2$  process (50 mg/g DS of FNA and  $H_2O_2$ ) was 0.14 mg COD/mg VS, which were 0.05, 0.04 and 0.10 mg COD/mg VS in original sludge, FNA process and  $H_2O_2$  process [81]. The methane potential after FNA- $H_2O_2$  process was enhanced prominently (59–83%). When compared with original sludge without pretreatment, the methane potentials after  $H_2O_2$  or FNA process were improved 23% and 56%, respectively [81].

In MW- $H_2O_2$  process, MW irradiation causes the destruction of EPS and divalent cation network leading to the improvement of WAS hydrolysis and biodegradation. Besides,  $H_2O_2$  activation is promoted under MW irradiation. Strong oxidative  $HO\cdot$  is produced to oxidize EPS and complex organics to soluble organics. Therefore, sludge hydrolysis efficacy has been improved, which provides the foundation for improving anaerobic digestion efficacy [82,83]. Eskicioglu et al. compared the effects of  $H_2O_2$  and MW- $H_2O_2$  process in anaerobic digestion [82]. In  $H_2O_2$  process, the stable dissolution rates of total solid, COD,

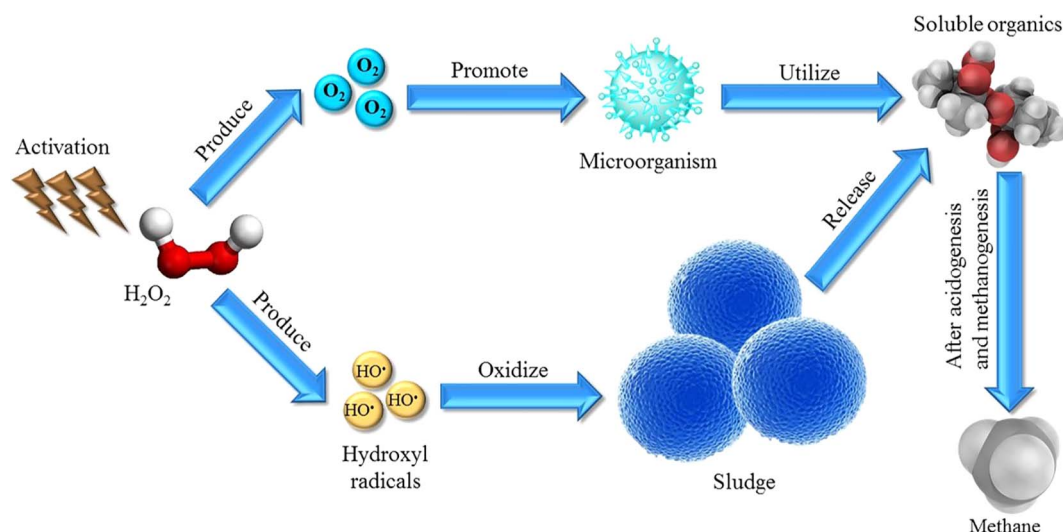


Fig. 4. Mechanism of  $\text{H}_2\text{O}_2$  based AOP in sludge anaerobic digestion.

proteins, sugars and humic acids were only 0%, 2%, 0%, 0% and 1%, respectively. In MW- $\text{H}_2\text{O}_2$  process, the stable dissolution rates of total solid, COD, proteins, sugars and humic acids were increased to 20%, 28%, 35%, 68% and 53%, respectively. When compared with original sludge without pretreatment, the methane yield in MW- $\text{H}_2\text{O}_2$  process after 32 d digestion increased 54%, which increased 25% in  $\text{H}_2\text{O}_2$  process [82]. The effect of pH condition in MW- $\text{H}_2\text{O}_2$  process for sludge anaerobic digestion has been investigated by Eswari et al. [75]. The original pH of sludge was 7.15, and the pH in MW- $\text{H}_2\text{O}_2$ -acid process was adjusted to 3.12–4.18. The COD solubilization in MW, MW- $\text{H}_2\text{O}_2$  and MW- $\text{H}_2\text{O}_2$ -acid process was 30.2%, 50.3% and 56.1%, respectively. The methane yield of original sludge after 30 d digestion was low (33 mL/g VS). In comparison, the methane yields in MW, MW- $\text{H}_2\text{O}_2$  and MW- $\text{H}_2\text{O}_2$ -acid process were 175, 288 and 323 mL/g VS, respectively [75]. Hence, acidic condition was more favorable for sludge anaerobic digestion. In US- $\text{H}_2\text{O}_2$  process, US can promote the dissolution of complex organics and increase the production of  $\text{HO}^\cdot$  by  $\text{H}_2\text{O}_2$  decomposition. The cooperation of US and  $\text{H}_2\text{O}_2$  promotes the increase of SCOD content in aqueous phase. When compared with the case without pretreatment, the SCOD content increased 40% in US- $\text{H}_2\text{O}_2$  process after 60 min sonication [84].

However, the dosage of  $\text{H}_2\text{O}_2$  in sludge anaerobic digestion need to be controlled. The residual  $\text{H}_2\text{O}_2$  in sludge treatment possibly damages the microbial cells or inhibits the microbial metabolism. Besides, residual  $\text{H}_2\text{O}_2$  has toxic effect on enzymes and bacteria participating in anaerobic digestion, which may restrict the whole anaerobic digestion process and result in long lag phase and low methane yield [85].

Furthermore, high  $\text{H}_2\text{O}_2$  dosage also leads to the emergence of refractory compounds causing reaction stagnation and low methane yield. In the research of Liu et al., MW- $\text{H}_2\text{O}_2$  process was introduced in sludge anaerobic digestion [85]. When 0.2 and 0.6 g  $\text{H}_2\text{O}_2$ /g DS of  $\text{H}_2\text{O}_2$  were added in MW- $\text{H}_2\text{O}_2$  process, the sludge dissoluble rates were improved 35% and 45%, respectively. The improvement of methane yield was 29.02% at 0.2 g  $\text{H}_2\text{O}_2$ /g DS of  $\text{H}_2\text{O}_2$ . Nevertheless, no increase of methane yield was observed at 0.6 g  $\text{H}_2\text{O}_2$ /g DS of  $\text{H}_2\text{O}_2$ . Meanwhile, the SCOD removal rate was decreased 30.63%. These results indicated the inhibition of excessive  $\text{H}_2\text{O}_2$  in anaerobic digestion. The utilization of catalase in anaerobic digestion may be a great solution to eliminate the inhibition of  $\text{H}_2\text{O}_2$  according to its high decomposition [85].

#### 4. $\text{H}_2\text{O}_2$ based AOP in the removal of pollutants

The disposal of pollutants such as EDC, TC, DEHP, PG and heavy metals in activated sludge is urgently for environmental protection [87,88]. The application of  $\text{H}_2\text{O}_2$  based AOP in pollutant removal has obtained remarkable effects. The high oxidative  $\text{HO}^\cdot$  in  $\text{H}_2\text{O}_2$  based AOP can degrade organic pollutants effectively. These organic pollutants are oxidized to harmless intermediate products or  $\text{CO}_2$ . Besides, the binding heavy metals can be changed to free state and released from sludge via  $\text{HO}^\cdot$  destroying sludge floc and EPS. The odors released from sludge also can be eliminated by  $\text{H}_2\text{O}_2$  based AOP.

As a class of severe pollutants, estrogens such as estrone (E1), 17  $\beta$ -estradiol (E2), estriol (E3), and 17  $\alpha$ -ethinylestradiol (EE2) have

Table 3  
The effect of  $\text{H}_2\text{O}_2$  based AOP in sludge anaerobic digestion.

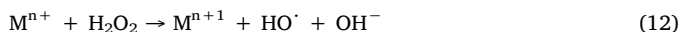
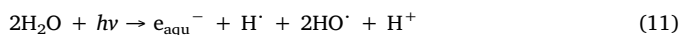
Method	$\text{H}_2\text{O}_2$ dosage	Other condition	Time	SRT (d)	SCOD content		Methane yield		Enhancement of methane potential (%)		Ref
					Before	After	Before	After	Before	After	
FNA- $\text{H}_2\text{O}_2$ oxidation	80 mg/g DS	FNA: 1.54 mg N/L	–	15	0.05 mg COD/mg VS	0.14 mg COD/mg VS	170 L/kg VS <sup>a</sup>	311 L/kg VS <sup>a</sup>	83	[81]	
MW- $\text{H}_2\text{O}_2$ oxidation	0.2 g/g DS	Temperature: 373 K Irradiation: 600 W	–	30	200 mg/L	9000 mg/L	150 mL/g VS	193 mL/g VS	29	[85]	
MW- $\text{H}_2\text{O}_2$ -acid oxidation	0.3 mg/g SS	Temperature: 373 K Irradiation: 1200 W	9 min	30	200 ± 10 mg/L	12.35 g/L	33 mL/g VS	323 mL/g VS	–	[75]	
Electro- $\text{H}_2\text{O}_2$ oxidation	1.8 g/L	Current density: 2.5 A/dm <sup>2</sup>	4 h	35	1.9 ± 0.9 g/L	4.8 ± 3.0 g/L	41.2 mL/g VS	146.8 mL/g VS	78	[86]	
US- $\text{H}_2\text{O}_2$ oxidation	50 g $\text{H}_2\text{O}_2$ /kg DS	Operational frequency :20 kHz	60 min	20	107 mg COD/L	2827 mg COD/L	–	–	–	[84]	

<sup>a</sup> The value is obtained from the figure of authors' study.

potential hazards to humans and wildlife [89]. In Fenton process, HO<sup>·</sup> generated in the reaction between H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup> can effectively oxidize and degrade estrogens retained in sludge. Besides, Fe<sup>3+</sup> can be reduced to Fe<sup>2+</sup> for initiating Fenton process. The oxidation of HO<sup>·</sup> contributes to the degradation and mineralization of EDC. Residue intermediates like ETD-9 can be further converted into less active aliphatic compounds [30]. In the research of Li and Zhang, Fenton process has great performance in the removal of EDC. Under the condition of pH 3 with 15.62 mmol/g of H<sub>2</sub>O<sub>2</sub> and 0.167 of Fe<sup>2+</sup>:H<sub>2</sub>O<sub>2</sub> ratio, the removal rates of E1, E2, EE2 and E3 reached their maximums 70%, 90%, 84% and 98%, respectively [90]. However, excessive H<sub>2</sub>O<sub>2</sub> could promote the reaction between of Fe<sup>3+</sup> and HO<sup>·</sup>, which inhibits the removal of E1, E2, EE2 and E3. When H<sub>2</sub>O<sub>2</sub> dosage increased to 46.86 mmol/g, the removal rates of E1, E2, EE2 and E3 decreased to 64%, 81%, 67% and 87%, respectively. Besides, pH also influences the effect of Fenton process. High pH condition leads to the emergence of massive hydroperoxide anion (HO<sub>2</sub><sup>-</sup>), which acts as an efficient scavenger of HO<sup>·</sup> (Eq. (10)). Meanwhile, Fe<sup>3+</sup> would precipitate as iron hydroxide under alkaline condition [90]. Hence, the cycle of Fe<sup>3+</sup>/Fe<sup>2+</sup> is inhibited and formation of Fe<sup>2+</sup> is restricted, which are unfavorable for the Fenton process. These factors may lead to decrease of the efficiency of Fenton process in the removal of EDC.



Zhang and Li investigated the effect of ultraviolet (UV)-H<sub>2</sub>O<sub>2</sub> process in the removal of EDC [91]. UV can activate H<sub>2</sub>O<sub>2</sub> for improving the production of HO<sup>·</sup>. Under UV irradiation, H<sub>2</sub>O<sub>2</sub> is transformed to HO<sup>·</sup> and H<sub>2</sub>O is transformed to HO<sup>·</sup>, hydrated electrons (e<sub>aq</sub><sup>-</sup>), and H<sup>·</sup> (Eq. (11)). Additionally, metals such as Fe, Ag and Cu retained in sludge also participate in Fenton-type process for producing H<sub>2</sub>O<sub>2</sub> (Eq. (12); M is metal) [92]. EDC in sludge can combine with metals and form EDC-metal complex (Eq. (13)). Then, EDC-metal complex is attacked by molecule oxygen with the formation of an oxygen complex. Eventually, radicals are formed by electron transferring through metal ion to oxygen [93]. In H<sub>2</sub>O<sub>2</sub> process at pH 3 with 2 mol/L of H<sub>2</sub>O<sub>2</sub>, the removal rates of E1, EE2, E3, BPA and NP after 40 min reaction were 73%, 58%, 58%, 54% and 46%, respectively. In UV-H<sub>2</sub>O<sub>2</sub> process with same pH condition and 0.5 mol/L of H<sub>2</sub>O<sub>2</sub>, the removal rates of E1, E2, EE2, E3, BPA and NP in 2 min were 97%, 92%, 95%, 94%, 89% and 67%, respectively [91]. Compared with H<sub>2</sub>O<sub>2</sub> process, the degradation efficiency was improved in UV-H<sub>2</sub>O<sub>2</sub> process, and the required reaction time and H<sub>2</sub>O<sub>2</sub> dosage were reduced. However, alkaline condition may lead to the surface charge of EDC changing from neutral charge to negative charge, which is detrimental to UV-H<sub>2</sub>O<sub>2</sub> process. Besides, precipitations may be formed in this condition. Meanwhile, HO<sub>2</sub><sup>-</sup> may restrict the efficiency of HO<sup>·</sup>. Moreover, excessive H<sub>2</sub>O<sub>2</sub> scavenges could compete with HO<sup>·</sup> and inhibit the removal of EDC [94].



Chlortetracycline (CTC), a kind of TC, can accumulate in sludge according to its strong adsorption to organic matrix and metals [95–99]. US-Fenton process can effectively degrade CTC in sludge. In US-Fenton process, acoustic cavitation and oxidative radicals promote the disintegration of sludge floc and dissolution of sludge. Hence, CTC accumulated in sludge is changed from bound state into free state, and then released from sludge into aqueous phase for further degradation [100]. Pulicharla et al. compared the effects of US, Fenton and US-Fenton process in CTC degradation [101]. Under the optimum condition, the CTC removal rates in US, Fenton and US-Fenton process were 67%, 76% and 82%, respectively. Hence, US-Fenton process was more favorable than US and Fenton process [101].

Besides, Fenton process can assist the biodegradation of DEHP in

anaerobic digestion. In the research of Pham et al., after 20 d anaerobic digestion, the removal rates of DEHP and 2-ethylhexanoic acid (2-EHXA) were 72% and 71%, respectively [102]. After introducing Fenton process as pretreatment, the removal rates of DEHP and 2-EHXA were increased to 85% and 79%, respectively [102]. Chang et al. studied the effect of MW-H<sub>2</sub>O<sub>2</sub> process in the elimination of NP [103]. In MW-H<sub>2</sub>O<sub>2</sub> process, NP compounds were more easily to be extracted and digested by MW irradiation heating sludge particles. Then, NP compounds were oxidized by HO<sup>·</sup> effectively. Under the optimum condition (3 min reaction with 5 mL of H<sub>2</sub>O<sub>2</sub>), over 95% NP was degraded [103]. MW-H<sub>2</sub>O<sub>2</sub> process also has great performance in the degradation of PG. Yu et al. reported that fecal coliforms can be degraded effectively by MW irradiation and oxidative HO<sup>·</sup> [31]. MW temperature and H<sub>2</sub>O<sub>2</sub> dosage were main factors in fecal coliforms degradation. When MW temperature was 328 K and H<sub>2</sub>O<sub>2</sub> dosage lower than 0.04% (w/w), fecal coliforms counts decreased from 5.1 log CFU/L to 3 log CFU/L. When MW temperature exceeded 343 K and more than 0.04% (w/w) of H<sub>2</sub>O<sub>2</sub> was added, fecal coliforms were degraded totally and cannot be detected [31].

H<sub>2</sub>O<sub>2</sub> based AOP also can be applied in the removal of heavy metals. In Fenton process, oxidative HO<sup>·</sup> destroys sludge structure and enhances EPS dissolution, thus heavy metals binding in sludge are released from sludge and dissolved in aqueous phase [33]. Andrews et al. found exceed 70% of copper (Cu) and zinc (Zn) were eluted from sludge after Fenton process at 9.93 g/L of H<sub>2</sub>O<sub>2</sub> and 16 g/L of Fe [104]. Besides, the content of cadmium (Cd) and nickel (Ni) was lower than detection limits [104]. In the research by Zhu et al., Fenton-like process promoted the efficiency of bioleaching in the removal of heavy metals [105]. After 5 d bioleaching, the removal rates of Cu, Zn and Cd all were about 50%, whereas the removal of plumbum (Pb) was only about 19%. When introducing Fenton process as pretreatment, the removal rates of Cu, Zn, Pb and Cd were increased to 75.3%, 72.6%, 34.5% and 65.4%, respectively [105]. Wang et al. investigated the chemical speciation of heavy metals in MW-H<sub>2</sub>O<sub>2</sub> process, which included acid soluble/exchangeable, reducible, oxidizable and residual state [14]. The contents of heavy metals in acid soluble/exchangeable, reducible, oxidizable and residual form were referred as F1, F2, F3 and F4. Metals in form of F4 have low mobility and no toxicity [106]. In original sludge, the F4 of Zn, Cu, Cd and Pb was 13.53%, 20.57%, 12.20% and 48.18%, respectively. After MW-H<sub>2</sub>O<sub>2</sub> process, the F4 of Zn, Cu, Cd and Pb increased about 34.47%, 31.43%, 62.80% and 23.82%, respectively [14]. These results indicated that the potential metal risk has been reduced effectively.

The odor released from sludge has a high risk to atmospheric environment and human health [107,108]. Sulfur-containing compounds are the main contributors to odors. In US-Fenton process, the release of odors from sludge is promoted by cavitation effect [109]. Dissolved sulfur (S<sup>2-</sup>) can be oxidized to odorless dissolved sulfate (SO<sub>4</sub><sup>2-</sup>) by H<sub>2</sub>O<sub>2</sub> (Eq. (14)). Besides, hydrogen sulfide and organic sulfur-containing compounds can be transformed to SO<sub>4</sub><sup>2-</sup> by HO<sup>·</sup> generated in US-Fenton process [110]. Liu et al. investigated the change of S<sup>2-</sup> content in US, Fenton and US-Fenton process [111]. The original S<sup>2-</sup> content was approximately 18 mg/L. After 60 min reaction in US, Fenton and US-Fenton process, the S<sup>2-</sup> content was reduced to 8.1, 12.3 and 16.2 mg/L, respectively. Meanwhile, the SO<sub>4</sub><sup>2-</sup> content in US-Fenton process increased from 200 to 390.6 mg/L, which was 95.3% and 5% higher than that in US and Fenton process [111]. Mechanism of H<sub>2</sub>O<sub>2</sub> based AOP in the removal of pollutants is shown in Fig. 5. The effects of H<sub>2</sub>O<sub>2</sub> based AOP in the removal of pollutants are summarized in Table 4.



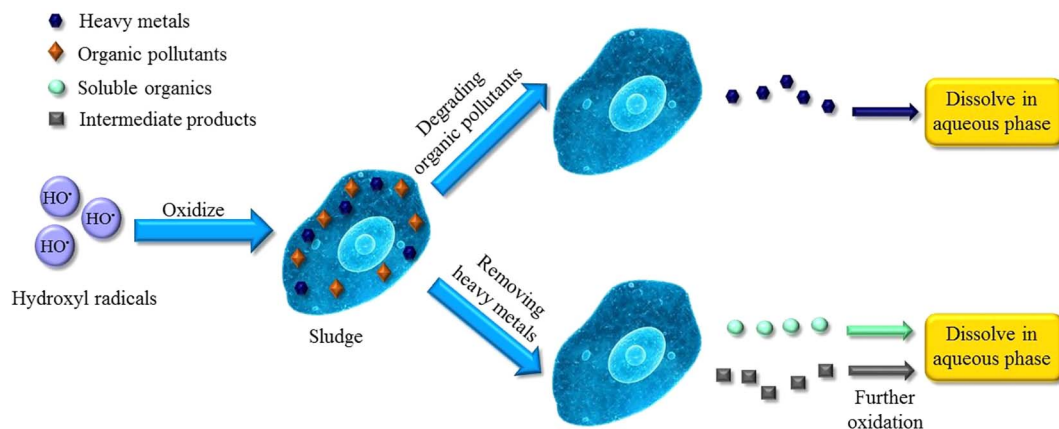


Fig. 5. Mechanism of H<sub>2</sub>O<sub>2</sub> based AOP in the removal of pollutants.

Table 4  
The effect of H<sub>2</sub>O<sub>2</sub> based AOP in removing pollutants.

Pollutant	Method	Condition		Removal rate (%)	Ref
		H <sub>2</sub> O <sub>2</sub> dosage	Other		
Chlortetracycline	Fenton process	0.45 mL	Fe <sup>2+</sup> : 30 mg/g DS; pH: 7	Chlortetracycline: 76	[101]
DEHP	Fenton process	30% v/v	FeSO <sub>4</sub> : 0.01 g/L; pH: 7	DEHP: 85; 2-ethylhexanoic acid: 79	[102]
EDC	Fenton process	15.62 mmol/g	Fe <sup>2+</sup> : 3.52 mmol g <sup>-1</sup> ; pH: 3	E1:70; E2: 90; EE2: 84; E3: 98	[90]
EDC	UV-H <sub>2</sub> O <sub>2</sub> process	0.5 mol/L	pH: 3; UV wavelength: 253.7 nm; UV fluence rate:0.069 mW cm <sup>-2</sup>	E1: 97; E2:92; EE2: 95; E3: 94; BPA: 89; NP: 67	[91]
NP	MW-H <sub>2</sub> O <sub>2</sub> -acid process	5 mL	Temperature: 423 K; HNO <sub>3</sub> : 5 mL	NP: over 95	[103]
Odors	US-H <sub>2</sub> O <sub>2</sub> process	0.5 g/L	Fe <sup>2+</sup> : 0.2 g/L	S <sup>2-</sup> : 53.45	[111]
Toxic metals	Fenton process	3.97–9.93 g/L	Fe: 10–16 g/L	Cu: 80; Zn: 80; Cd: 100; Ni 100	[104]
Toxic metals	Fenton-like process combined with bioleaching	5 g/L	5 d bioleaching	Cu: 75.3; Zn: 72.6; Pb: 34.5; Cd: 65.4	[105]

Table 5  
Economic analysis of different H<sub>2</sub>O<sub>2</sub> based AOP in sludge dewatering.

Method	Dosage (t/t DS)			Reaction time (min)	Total cost (\$/t DS)	Water content (%)	Ref
	H <sub>2</sub> O <sub>2</sub>	Catalyst	Other				
Fenton process	0.13	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 0.18	H <sub>2</sub> SO <sub>4</sub> : 0.06	41	44.6	63.65	[58]
Fenton-lime process	0.13	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 0.24	Lime: 0.43	43	77.2	55.80	[57]
Fenton-red mud process	0.13	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 0.18	Red mud: 0.28	41	46.0	59.80	[58]
Fenton-OPC-lime process	0.15	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 0.18	H <sub>2</sub> SO <sub>4</sub> : 0.06 Lime: 0.28 OPC: 0.28	41	85.0	49.50	[58]
Fenton-DDBAC process	0.04	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 0.20	H <sub>2</sub> SO <sub>4</sub> : 0.06 DDBAC: 0.06	47	~19797.5	57.17	[48]
Fenton-like process	0.38	Fe <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> : 2.06	H <sub>2</sub> SO <sub>4</sub> : Unknow	30	~170.8	66.10	[59]

Note: The chemical cost was calculated based on per ton of dry sludge in related studies. The prices of H<sub>2</sub>O<sub>2</sub>, FeSO<sub>4</sub>·7H<sub>2</sub>O, H<sub>2</sub>SO<sub>4</sub>, lime, red mud and OPC are 200, 70, 100, 80, 5 and 50 \$/t [57,58]. The prices of Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> and DDBAC were 46 ([www. alibaba.com](http://www.alibaba.com)) and 329,593 \$/t ([www. alibaba.com](http://www. alibaba.com)).

Table 6  
Economic analysis of different H<sub>2</sub>O<sub>2</sub> based AOP in sludge minimization.

Method	Dosage (t/t DS)		Reaction time (min)	Chemical cost (\$/t DS)	Energy cost (\$/t DS)	Total cost (\$/t DS)	Decrease of TCOD (%)	Ref
	H <sub>2</sub> O <sub>2</sub>	Other						
Fenton process	0.34	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 0.03 H <sub>2</sub> SO <sub>4</sub> : 0.73	30	143.1	0	143.1	72	[71]
US-Fenton process	0.88	FeSO <sub>4</sub> ·7H <sub>2</sub> O:0.01 H <sub>2</sub> SO <sub>4</sub> : 1.15	20	291.7	1.76	293.46	79.0	[50]
Electro-Fenton process	0.67	NaCl: 2.00 H <sub>2</sub> SO <sub>4</sub> : Unknown	120	~240.0	4.03	244.3	72	[60]

Note: The chemical cost and energy cost were calculated based on per ton of dry sludge in related studies. The prices of H<sub>2</sub>O<sub>2</sub>, FeSO<sub>4</sub>·7H<sub>2</sub>O, H<sub>2</sub>SO<sub>4</sub>, NaCl and electricity are 200, 70, 100, 53 \$/t ([www. alibaba.com](http://www. alibaba.com)) and 0.15 \$/kWh [57,58,60].



**Table 7**  
Economic analysis of different H<sub>2</sub>O<sub>2</sub> based AOP in sludge anaerobic digestion.

Method	Dosage (t/t DS)		Reaction time	Increase of Methane yield (m <sup>3</sup> /t VS)	Output energy (kWh/t)	Benefit of methane (\$/t)	Chemical cost (\$/t DS)	Energy cost (\$/t DS)	Net benefit (\$/t)	Ref
	H <sub>2</sub> O <sub>2</sub>	Other								
Indigenous iron-H <sub>2</sub> O <sub>2</sub> process	0.05	NaOH: 0.002	30 min	34	400	60	10.6	–	49.40	[78]
FNA-H <sub>2</sub> O <sub>2</sub> process	0.08	HCl: 0.033	24 h	141	1617	242.55	21.0	–	221.55	[81]
MW-H <sub>2</sub> O <sub>2</sub> process	1.17	–	9 min	255	2925	438.75	234.0	1620	–1415.25	[75]
MW-H <sub>2</sub> O <sub>2</sub> -acid process	1.17	H <sub>2</sub> SO <sub>4</sub> :0.002	9 min	290	3327	499.05	234.2	1620	–1355.15	[75]
Electro-H <sub>2</sub> O <sub>2</sub> process	0.06	–	4 h	105.6	1211	181.65	10.84	23,301	–23130.19	[86]

Note: The chemical cost, energy cost and benefit of methane were calculated based on per ton of dry sludge in related studies. The prices of H<sub>2</sub>O<sub>2</sub>, NaOH, HCl, H<sub>2</sub>SO<sub>4</sub> and electricity are 200, 300, 150 and 0.15 \$/kWh [57,78]. Methane calorific value is 16 kWh/kgCH<sub>4</sub> and power price is 0.15 \$/kwh [78].

**Table 8**  
The economic analysis of H<sub>2</sub>O<sub>2</sub> based AOP in pollutant removal.

Method	Dosage (t/t DS)		Reaction time	Removal of pollutants (%)	Cost (\$/t DS)	Ref
	H <sub>2</sub> O <sub>2</sub>	Other				
Fenton process	0.538	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 3.101	60 min	E1:70%; E2: 90%; EE2: 84%; E3: 98%	324.67	[90]
UV/H <sub>2</sub> O <sub>2</sub> process	1.17	–	60 min	E1: 97%; E2: 92%; EE2: 95%; E3: 94%	1204.5	[91]
Fenton process	0.271	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 2.135	60 min	Cu: 80%; Zn: 80%	270.25	[104]
H <sub>2</sub> O <sub>2</sub> -bioleaching process	0.034	FeSO <sub>4</sub> ·7H <sub>2</sub> O: 1.0	15 d	Cu: 75.3%; Zn: 72.6%	76.8	[105]

Note: The chemical cost and energy cost were calculated based on per ton of dry sludge in related studies. The cost in this table is the sum of chemical cost and energy cost. The prices of H<sub>2</sub>O<sub>2</sub> and FeSO<sub>4</sub>·7H<sub>2</sub>O are 200 and 70 \$/t [57].

## 5. Cost estimation

The cost estimation of H<sub>2</sub>O<sub>2</sub> based AOP is important for measuring the practical application potential. Based on available data from the published literature for sludge treatment, the preliminary cost estimation of H<sub>2</sub>O<sub>2</sub> based AOP for different sludge treatment has been materialized.

Table 5 depicts the chemical cost of different H<sub>2</sub>O<sub>2</sub> based AOP in sludge dewatering. The calculated cost of Fenton process (44.6 USD (\$)/t DS) is less than Fenton-like process (170.8 \$/t DS). When considering the Wc after these process, Fenton process may be a favorable choice. The utilization of skeleton builders such as lime, red mud and OPC in Fenton process can improve sludge dewaterability. The calculate costs of Fenton-lime, Fenton-red mud and Fenton-OPC-lime process were 77.2, 46.0 and 85.0 \$/t DS, respectively. Hence, using red mud combined with Fenton process is economic for sludge dewatering. Besides, the cost of Fenton-DDBAC process is about 19797.5 \$/t DS, which may be too expensive for practical application.

The chemical costs and energy costs of these methods in sludge minimization were calculated for comparison (Table 6). The chemical costs of Fenton, US-Fenton and electro-Fenton process were about 143.1, 291.7 and 240.0 \$/t DS, respectively. Besides, US-Fenton and electro-Fenton process need extra energy consumption, which were 1.76 and 4.03 \$/t DS, respectively. When compared with Fenton process, their higher costs indicate that related study focusing on improving efficiency and saving cost is necessary.

The cost estimation of H<sub>2</sub>O<sub>2</sub> based AOP in sludge anaerobic digestion is shown in Table 7. When compared with typical anaerobic digestion, the application of indigenous iron-H<sub>2</sub>O<sub>2</sub> process and FNA-H<sub>2</sub>O<sub>2</sub> process can bring extra economic benefit, which was 60 and 242.55 \$/t, respectively. After deduction of chemical costs, the net benefits were 49.4 and 221.55 \$/t, respectively. Besides, although the methane yields were improved by MW-H<sub>2</sub>O<sub>2</sub> process, MW-H<sub>2</sub>O<sub>2</sub>-acid process and electro-H<sub>2</sub>O<sub>2</sub> process, their high energy consumptions may lead to their practical application become difficult. Overall, FNA-H<sub>2</sub>O<sub>2</sub> process is a better choice according to its cheap cost and high economic benefit.

Table 8 shows the preliminary costs of H<sub>2</sub>O<sub>2</sub> based AOP in the

removal of pollutants. In the removal of EDC, the costs of Fenton process and UV-H<sub>2</sub>O<sub>2</sub> process were 324.67 and 1204.5 \$/t DS, respectively. The high energy cost (970.5 \$/t DS) in UV-H<sub>2</sub>O<sub>2</sub> process may lead to its practical application become hard. In the removal of heavy metals (Cu and Zn), the cost of bioleaching-H<sub>2</sub>O<sub>2</sub> process was cheaper than Fenton process, which was 76.8 \$/t DS. However, the processing period of bioleaching-H<sub>2</sub>O<sub>2</sub> process was longer than Fenton process. Hence, considering improve its efficiency is necessary before practical application.

## 6. Conclusion

This review provides a general overview of H<sub>2</sub>O<sub>2</sub> based AOP in sludge dewatering, sludge minimization, anaerobic digestion and removal of pollutants. In H<sub>2</sub>O<sub>2</sub> based AOP, strong oxidative HO· is produced for sludge treatment to realize sludge reduction, recycling and risk relief. In sludge dewatering, HO· can destroy EPS and promote the release of sludge bound water into aqueous phase. Meanwhile, HO· stimulates sludge disintegration, solubilization and mineralization to realize sludge minimization. In sludge anaerobic digestion, sludge EPS and other recalcitrant organics are transformed into soluble organics by oxidative HO· to improve the efficiency of anaerobic digestion. Furthermore, organic pollutants retained in sludge can be oxidized and removed by HO·. In addition, the destruction of sludge floc and EPS by HO· promotes the transition of heavy metals from binding state to free state for metal removal. Besides, the odor released from sludge also can be eliminated.

Although H<sub>2</sub>O<sub>2</sub> based AOP have achieved attracting effects in sludge treatment, there still exist some deficiencies. Adjusting pH to optimum is necessary for H<sub>2</sub>O<sub>2</sub> based AOP to obtain best effects in sludge treatment. Besides, H<sub>2</sub>O<sub>2</sub> dosage also need to be controlled to avoid the inhibition to sludge anaerobic digestion. The relationship of H<sub>2</sub>O<sub>2</sub> and microorganism need to be illuminated distinctly. Moreover, in Fenton-type process, excessive metal ions may consume a part of HO· which is unfavorable for the process. The mechanism and reaction kinetics of HO· in sludge treatment still lack specific analysis to offer a better understanding. After the cost estimation, the costs of using MW,

electrolysis, US and UV in H<sub>2</sub>O<sub>2</sub> based AOP for sludge treatment process are high. Hence, the practical application of these process may be very hard currently. That how to improve their efficiency and reducing their energy consumption need to be considered. The utilization of sunlight in H<sub>2</sub>O<sub>2</sub> based AOP seems to be a nice choice. Future research can focus on catalyst with high photocatalytic performance and cheap cost to improve the photocatalytic efficiency in H<sub>2</sub>O<sub>2</sub> based AOP.

## Acknowledgments

The authors gratefully acknowledge the financial support provided by the Key research and development project of Hunan Province, China (No. 2016SK2015) and National Natural Science Foundation of China – China (No. 51739004, No. 51521006, No. 21776066).

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