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Research progress on remediation of total petroleum hydrocarbons in soil by chemical oxidation - a review

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Chemical oxidation remediation is a technique that involves the addition of chemical oxidants to contaminated soil to degrade total petroleum hydrocarbons (TPHs), with the goal of reducing soil organic matter content or eliminating organic contamination. This paper reviews the research progress of several major chemical oxidation remediation technologies, including hydrogen peroxide (H₂O₂)-based Fenton and Fenton-like systems, calcium peroxide (CaO₂)-based Fenton and Fenton-like systems, and persulfate-activated oxidation systems (e.g., Na₂S₂O₈). Among these, the persulfate-activated oxidation system has recently emerged as a research hotspot due to its potential in eliminating TPHs from soil. The efficiency of TPHs degradation depends significantly on the activation method employed and the oxidative capacity of the system. Consequently, future research should focus on two critical directions: (1) the development of highly efficient, cost-effective, and environmentally sustainable activation methods; and (2) the enhancement of oxidative performance in existing systems, such as Na₂S₂O₈/CaO₂ and Na₂S₂O₈/H₂O₂. In discussing the advancements in these major chemical oxidation remediation technologies, this paper specifically examines various persulfate activation methods and their corresponding treatment efficiencies. The aim is to provide insights and references for the development of efficient, cost-effective, and environmentally friendly persulfate-activated oxidation systems, thereby promoting the application of chemical oxidation remediation technologies in the treatment of petroleum hydrocarbon-contaminated soils.

KEYWORDS

total petroleum hydrocarbons, chemical oxidative remediation technologies, fentonbased reaction, activated persulfate, advanced oxidation

1 Introduction

With the continuous development of industry, the demand for fossil fuels is constantly increasing (Dai et al., 2024; Zhou et al., 2024), consequently, the issue of environmental pollution is becoming increasingly severe (Liu et al., 2024; Liu et al., 2023b). The exploitation of crude oil, the use of petroleum products, industrial production activities, discharges of oily wastewater, and natural spills have all contributed to the increasingly severe phenomenon of environmental pollution caused by TPHs (Ashjar et al., 2021; Wu et al., 2020). The leakage of Total Petroleum Hydrocarbons (TPHs) into the environment has altered the structure of soil and impacted microbial diversity (Zhang et al., 2023). As a

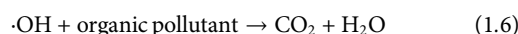
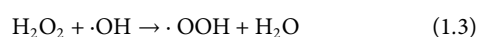
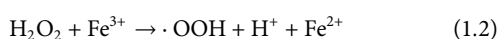
result, the soil becomes a carrier of hazardous substances, thereby posing risks to soil ecology, human health, and the wellbeing of plants and animals (Sayed et al., 2021; Pinedo et al., 2014; Kim et al., 2022). Consequently, the treatment and remediation of soils contaminated with TPHs, and the restoration of soil ecological stability, have emerged as urgent priorities that demand immediate, and targeted, interventions to address the growing environmental and ecological concerns (Yuan et al., 2023; Peng et al., 2017; Kim et al., 2021).

At present, physical restoration (Chang et al., 2022), bioremediation (Bidja Abena et al., 2019; Feng et al., 2021; Othman et al., 2022), and chemical remediation (Li et al., 2022; Effendi et al., 2022; Liu et al., 2023a; Chen et al., 2022) are commonly used to remediate TPHs-contaminated soils. Among these approaches, chemical remediation technology has emerged as a research hotspot, primarily due to its strong pertinence, high treatment efficiency, and relatively short implementation period. Chemical remediation technology mainly encompasses three key methods: chemical flushing technology (Naeem and Qazi, 2020), chemical stabilization technology (Yang et al., 2020), and chemical oxidation technology (Chang et al., 2022; Bu et al., 2023; Li et al., 2024a). The chemical oxidation systems commonly used for TPHs-contaminated soils, such as the H₂O₂ Fenton oxidation system, the CaO₂-based Fenton oxidation system, the activated Na₂S₂O₈ oxidation system and the Na₂S₂O₈/CaO₂ composite oxidation system (Ni et al., 2024).

This paper primarily reviews the advancements in chemical oxidation technology, which aims to eliminate or mitigate soil contamination by introducing chemical oxidizing agents and activators. This process effectively degrades pollutants and restores soil health. The review encompasses the practical applications and current research status of several key chemical oxidation technologies, while also exploring potential future directions for development.

2 H₂O₂ -based fenton system

In 1893, Fenton HJ initially proposed that a mixed solution of H₂O₂ and Fe²⁺ exhibits potent oxidising properties, and is capable of degrading the majority of organic substances into inorganic states. As a result of the extensive research conducted, the Fenton oxidation system has become a more sophisticated and well-developed process. With the depth of research, Fenton oxidation system has been developed more mature. H₂O₂ is used as the oxidant and mixed with Fe²⁺. Under acidic conditions, Fe²⁺ catalyzes the generation of highly oxidative hydroxyl radicals (·OH), which have a standard electrode potential of 2.76 V. The hydroxyl radical (·OH) is strongly electrophilic and highly oxidative, capable of undergoing electrophilic reactions with most organic pollutants. This ultimately leads to the mineralization of these pollutants into H₂O and CO₂. The mechanism of treatment can be described as follows (Equations 1.1–1.6):



The remediation of petroleum hydrocarbons in soils has been the subject of extensive study with regard to Fenton oxidation systems based on H₂O₂ (Apul et al., 2016). It has been demonstrated that the choice of catalysts and the influencing factors have a significant impact on the final degradation. The traditional hydrogen peroxide oxidation Fenton oxidation system employs hydrogen peroxide in conjunction with Fe²⁺, which reacts at low pH to produce ·OH, before undergoing further reaction. The Fenton oxidation system has been the subject of extensive research with the objective of enhancing its efficacy and reducing any potential environmental impact (Priyadarshini et al., 2022).

Chen et al. (2022) employed hydrogen peroxide (Fe²⁺/H₂O₂) oxidation systems, utilising Fe²⁺ as an activator to treat TPHs pollutants, the schematic diagram of the experimental setup was given (Figure 1a). The effects of the oxidant concentration, Fe²⁺ dosage, and initial pH conditions of the removal rate of TPHs from oil-based drilling cuttings as shown in Figure 1b. The findings demonstrated that under room temperature conditions, liquid–solid ratio = 10:1, H₂O₂ = 10 mmol/g, Fe²⁺ = 10 mmol/g, initial pH = 3, the TPHs removal rates were found to be 45.04% for the (Fe²⁺/H₂O₂) system. And the (Fe²⁺/H₂O₂) system removed up to 80% of C₁₀–C₁₃ components. In previous studies, Fernando Pardo (Pardo et al., 2014) and colleagues employed H₂O₂ as an oxidising agent (400–4,000 mmol L⁻¹), ferric ions as a catalyst (5–20 mmol L⁻¹), and trisodium citrate (50 mmol L⁻¹) as a chelating agent. The removal efficiencies of petroleum hydrocarbons (TPHs) were obtained at two different pollutant concentrations (1,000–10,000 mg diesel kg/soil), with efficacies reaching up to 75%.

Akpoveta et al. (2018) achieved up to 87.6% degradation of TPHs (10% diesel) by Fenton oxidation with 350,000 mg/L H₂O₂ and 600 mg/L FeSO₄, in the optimal room temperature range of 27°C–30°C, and at pH = 4.7, and the reaction products are environmentally friendly. Xu et al. (2011) employed three distinct oxidising reagents to address soil contamination resulting from cable insulating oil leaks. The efficacy of three different oxidising agents (H₂O₂, CaO₂, and permanganate) in facilitating the oxidation of various carbon chain lengths, including C₁₀–C₁₂, C₁₃–C₁₆, C₁₇–C₂₀, C₂₁–C₂₄, C₂₅–C₄₀ (Table 1), and TPHs oxidation were evaluated. Among them H₂O₂ is the most effective of the three oxidants for chemical oxidation of cable oil contaminated soils under neutral pH conditions. Therefore, the experiments investigated the optimum concentration of oxidant, the optimum amount of iron catalyst and the addition of hydrogen peroxide for the treatment of TPHs with H₂O₂ as oxidant. The results showed that the optimum removal rate of 46% of cable oil contaminated soil was achieved when Fe(II) = 6.98 mmol/L and H₂O₂ at a concentration of 1,469 mmol/L was added at three times with a minimum time interval of 5 h. Yang et al. (2022) also pointed out that the optimal method of adding reagents is to add the iron catalyst in one step, then the stepwise addition of H₂O₂, which has no effect on the microbiological environment in the soil.

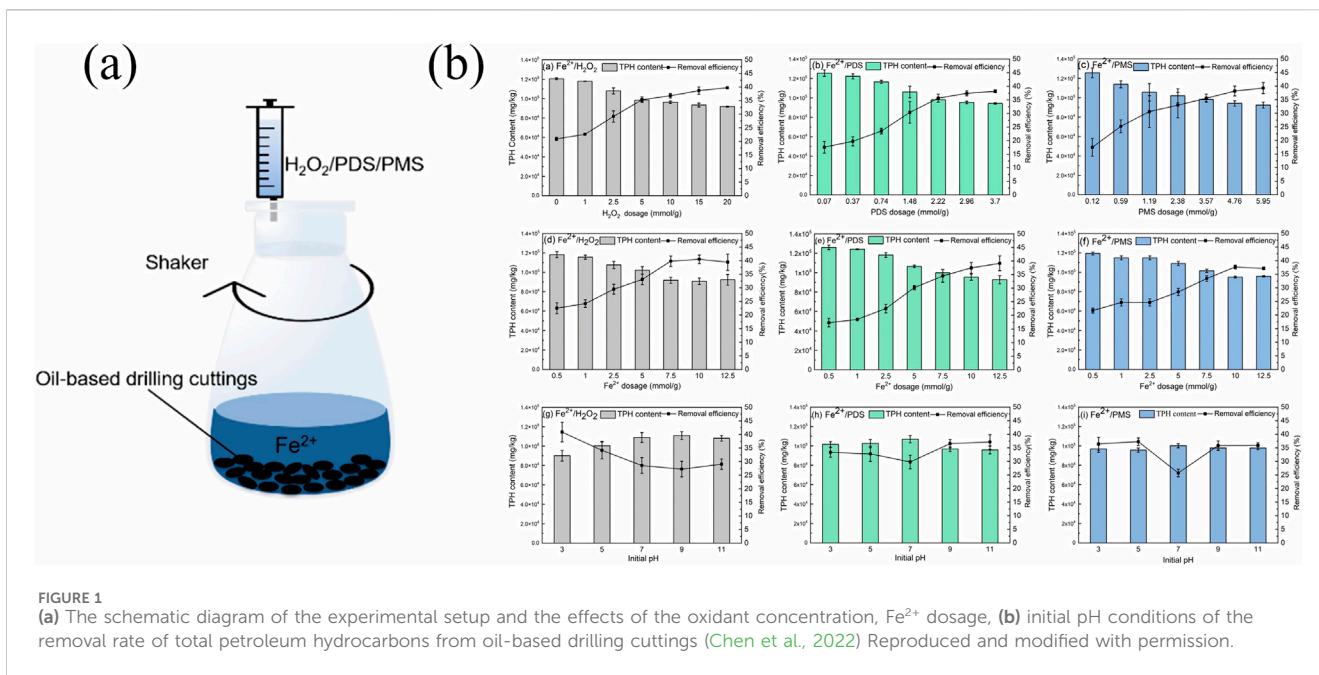


FIGURE 1 (a) The schematic diagram of the experimental setup and the effects of the oxidant concentration, Fe²⁺ dosage, (b) initial pH conditions of the removal rate of total petroleum hydrocarbons from oil-based drilling cuttings (Chen et al., 2022) Reproduced and modified with permission.

TABLE 1 Removal efficiency (%) of cable oil by liquid hydrogen peroxide pH 7.5 (Xu et al., 2011) Reproduced and modified with permission.

Oil fraction	H ₂ O ₂ concentration ^a (mM/v%)			Iron dose ^b (mg FeSO ₄)							Delivery method ^c		
	330 (1.0%)	1,464 (4.5%)	2,928 (9%)	0	27 (94:1)	53 (47:1)	80 (31:1)	106 (23:1)	133 (19:1)	265 (9:1)	M-1	M-2	M-3
C ₁₀ -C ₁₂	6	12	11	7	10	12	75	77	78	79	9	7	7
C ₁₃ -C ₁₆	9	19	8	10	15	23	6	3	7	11	21	47	31
C ₁₇ -C ₂₀	17	23	15	8	13	25	22	12	18	23	18	48	31
C ₂₁ -C ₂₄	17	27	16	6	12	27	16	1	9	19	13	47	30
C ₂₅ -C ₄₀	22	22	7	10	14	33	18	7	11	20	2	37	26
TPH	17	24	14	13	17	24	21	11	17	23	14	46	30

The initial TPH, concentration is 3,171 mg kg⁻¹.

^aCitric acid acts as chelating agent, iron dose is 52.9 mg FeSO₄, pH is 7.5. Delivery method is M-1.

^bCitric acid acts as chelating agent, 4.5% H₂O₂ were applied. pH is 7.5. Delivery method is M-1.

^cCitric acid acts as chelating agent, iron dose is 52.9 mg FeSO₄ 4.5% H₂O₂ were applied. pH is 7.5.

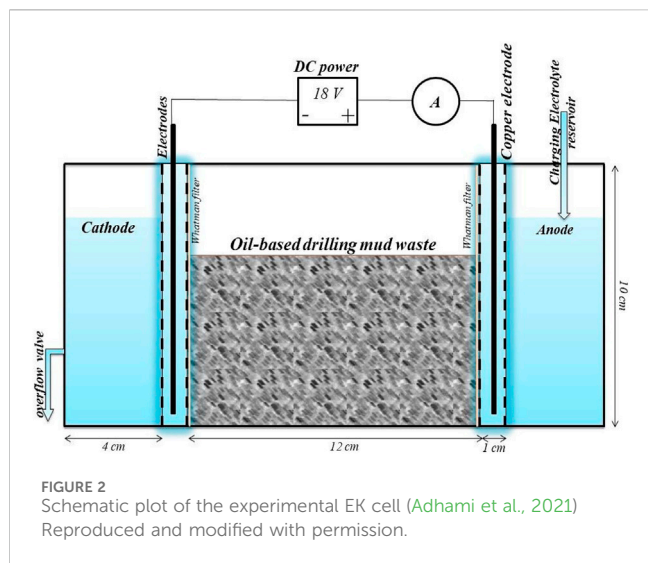
In order to enhance the efficacy of the H₂O₂ Fenton oxidation system in the treatment of TPHs, various new Fenton composite remediation systems have been proposed. In a study conducted by Adhami et al. (2021), an electrokinetic-Fenton oxidation system (Figure 2) was employed to treat oil-based drilling waste. This resulted in a notable reduction in the TPHs content, from 31,600 mg/kg to 72,680 mg/kg, with an impressive degradation rate of 77%, and this electrokinetic-Fenton method has also been widely used in the treatment of organic matter in soils (Paixão et al., 2020).

Sivagami et al. (2019) used the ultrasonic-Fenton method to treat TPHs in oil spill sludge, and under the conditions of pH = 3.0, sludge/water ratio = 1:100, ultrasonic power = 100 W and ultrasonic amplitude = 40–50%, and the weight ratio of H₂O₂/Fe²⁺ = 10:1, the removal of TPHs after ultrasonic treatment for 10 min was up to 84.25%, present excellent results.

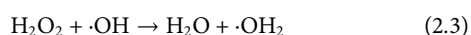
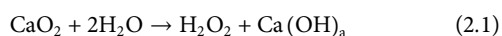
The H₂O₂ Fenton system is distinguished by a minimal environmental impact, a broad spectrum of applications, and straightforward operation. However, the efficacy of TPHs degradation is significantly influenced by the pH value. Therefore H₂O₂-based Fenton systems should require a combination of UV or visible light (Wang et al., 2016a), sono-, electro-, photo-electro-, sono-electro-, heterogeneous electro- and sono-photo- techniques to achieve higher TPHs removal rates (Priyadarshini et al., 2022; Khodaveisi et al. 2011).

3 CaO₂-based fenton system

The traditional H₂O₂ Fenton oxidation system has shortcomings such as dependence on the pH of the soil environment (applicable under acidic conditions) and poor H₂O₂



stability (Xue et al., 2018b). Therefore, to overcome the shortcomings of the traditional H₂O₂-based Fenton technology, researchers have proposed the CaO₂-based Fenton technology, which uses calcium peroxide (CaO₂) to generate H₂O₂ (Wang et al., 2016b) and then participates in the degradation reaction to treat organic pollutants in soil (Xue et al., 2019). Upon dissolution in water, calcium peroxide (CaO₂) generates hydrogen peroxide (H₂O₂), which is then activated by Fe²⁺ to produce reactive radicals (·OH), thereby forming the Fenton reaction. The mechanism of treatment can be described as follows (Equations 2.1–2.4):



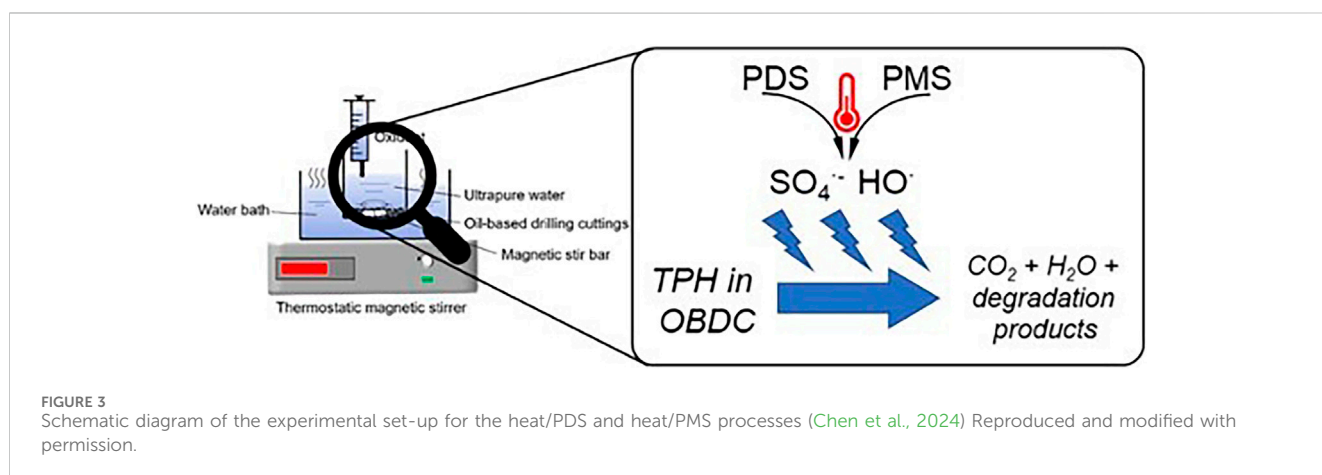
The results of the study (Northup and Cassidy, 2008; Bogan et al., 2003) indicate that the degradation rate of organic matter was increased to some extent when calcium peroxide was used instead of H₂O₂. In a study conducted by Jiang et al. (2021), calcium peroxide

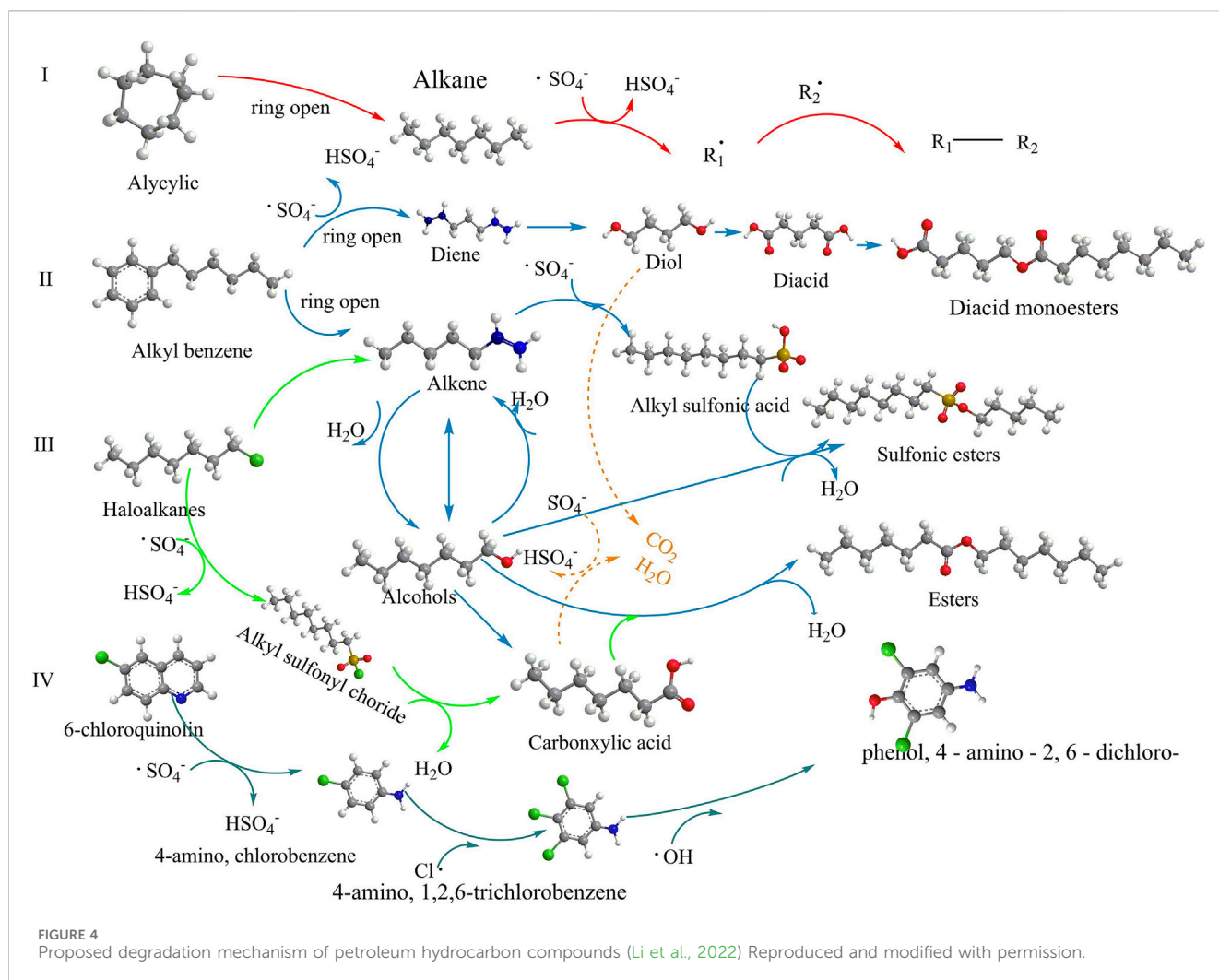
powder with a purity of 89.1% was prepared and used as an oxidising agent for degradation experiments on diclofenac sodium. The results demonstrated that the degradation rate of diclofenac sodium was 97.5% within 180 min, which was more effective than oxidation using H₂O₂-based Fenton’s system.

Ndjou’ou prepared a hydrocarbon-contaminated soil which was treated in laboratory slurry reactors using two types of modified Fenton (MF) chemistry (Ndjou’ou and Cassidy, 2006). The degradation rates of two oxidation systems, namely, liquid hydrogen peroxide (HP) and calcium peroxide (CaO₂) – based oxidant (Cool-Ox™), were compared for TPHs and the four individual fractions (C₆-C₁₀, C₁₁-C₁₆, C₁₇-C₃₄, >C₃₄). The results shown that, in soil with TPHs concentration = 10,604± (850) mg/kg, PH = 8, 40% w/v soil slurry, after 1 week, the removals of liquid HP (100 mL of 50% HP) and CaO₂-based (50 g) treatments were 73.7% and 95.6%, respectively. The results demonstrated that the CaO₂-based treatment exhibited a superior effect compared to the H₂O₂ treatment. This team also investigated the treatment of PAHs in soil by a calcium peroxide system (Gryzenia et al., 2009), which resulted in a degradation rate of 92.3% in 10 days at 21,420 g mg/kg, 500 g Cool-Ox™ powder, 40% w/v soil slurry.

Xue et al. (2018a) used a CaO₂-based Fenton system to treat a mixed system of several petroleum hydrocarbon substances in water, including: benzene, toluene, ethylbenzene, and xylene (marked as BTEX). The oxidation effect of BTEX was investigated by changing the ratio of CaO₂/Fe(II)/BTEX. The results demonstrated that when the CaO₂/Fe(II)/BTEX molar ratio was 5/5/1, BTEX removal was 35%. Furthermore, when the ratio was increased to 40/40/1, the removal rate of BTEX increased significantly, reaching 98% in the same condition. These findings offer insights into the degradation of TPHs in soil.

In water using a Fenton system with CaO₂. The objective was to investigate the oxidation effect of BTEX by varying the ratio of CaO₂/Fe(II)/BTEX. The removal of BTEX was found to be 35% when the molar ratio of CaO₂/Fe(II)/BTEX was 5/5/1. However, when the molar ratio was increased to 40/40/1, the removal rate increased significantly, reaching 98% in the same condition. The findings of this type of investigation may offer insights into the degradation of TPHs in soil. Furthermore, the efficacy of calcium peroxide can be enhanced by preparing it into nanoscale, which increases its specific surface area and consequently accelerates the



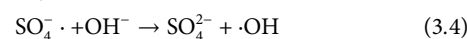
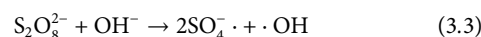
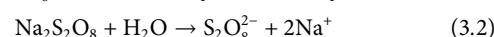
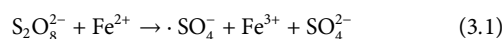


degradation of organic matter (Khodaveisi et al., 2011; Yavari-Bafghi et al., 2022).

4 Activated persulfate oxidation system

The use of persulfate as an oxidising agent for the degradation of organic pollutants has been widely reported (Lominchar et al., 2018; Xu et al., 2023). The stimulated activation of persulfate results in the generation of sulfate-free radicals ($\text{SO}_4^{\cdot -}$) with strong oxidising properties, which exhibit a standard electrode potential of 2.60 V. The most common methods of activation are heat, ultrasound (Darsinou et al., 2015; Deng et al., 2015), electrical current (Chen et al., 2019), ultraviolet (UV) light, and alkali conditions. Sodium persulfate, when activated by ferrous ions, decomposes to generate sulfate radicals ($\cdot\text{SO}_4^-$), which are highly oxidative with a standard electrode potential of 2.60 V. These sulfate radicals oxidize organic pollutants through mechanisms such as electron transfer, addition reactions, and hydrogen atom abstraction. The mechanism of Fe^{2+} activation can be described as (Equation 3.1). Persulfate dissociates in water to produce the $\text{S}_2\text{O}_8^{2-}$, with a standard redox potential of $E^0 = 2.01$ V, and $\text{S}_2\text{O}_8^{2-}$ generates sulfate radicals ($\cdot\text{SO}_4^-$) with a

redox potential of $E^0 = 2.60$ V. In alkaline conditions, these sulfate radicals can further produce hydroxyl radicals ($\cdot\text{OH}$) with a higher chemical oxidation potential ($E^0 = 2.80$ V). These reactive radicals react with pollutants in the soil, thereby facilitating the removal of contaminants, the mechanism of alkaline activation can be described as (Equations 3.2–3.4). The $\text{S}_2\text{O}_8^{2-}$ can be activated under sufficient thermal conditions, leading to the cleavage of the peroxide bond and the formation of $\text{SO}_4^{\cdot -}$, the mechanism of heat activation can be described as (Equation 3.5):



Persulfate was employed for the remediation of TPHs in soil by researchers (Wu et al., 2016). The degradation of TPHs was observed to reach up to 40.8% when the initial TPHs concentration was 14,432.5 mg/kg, and the reaction was conducted for 24 h, the degradation rate was 21% higher than

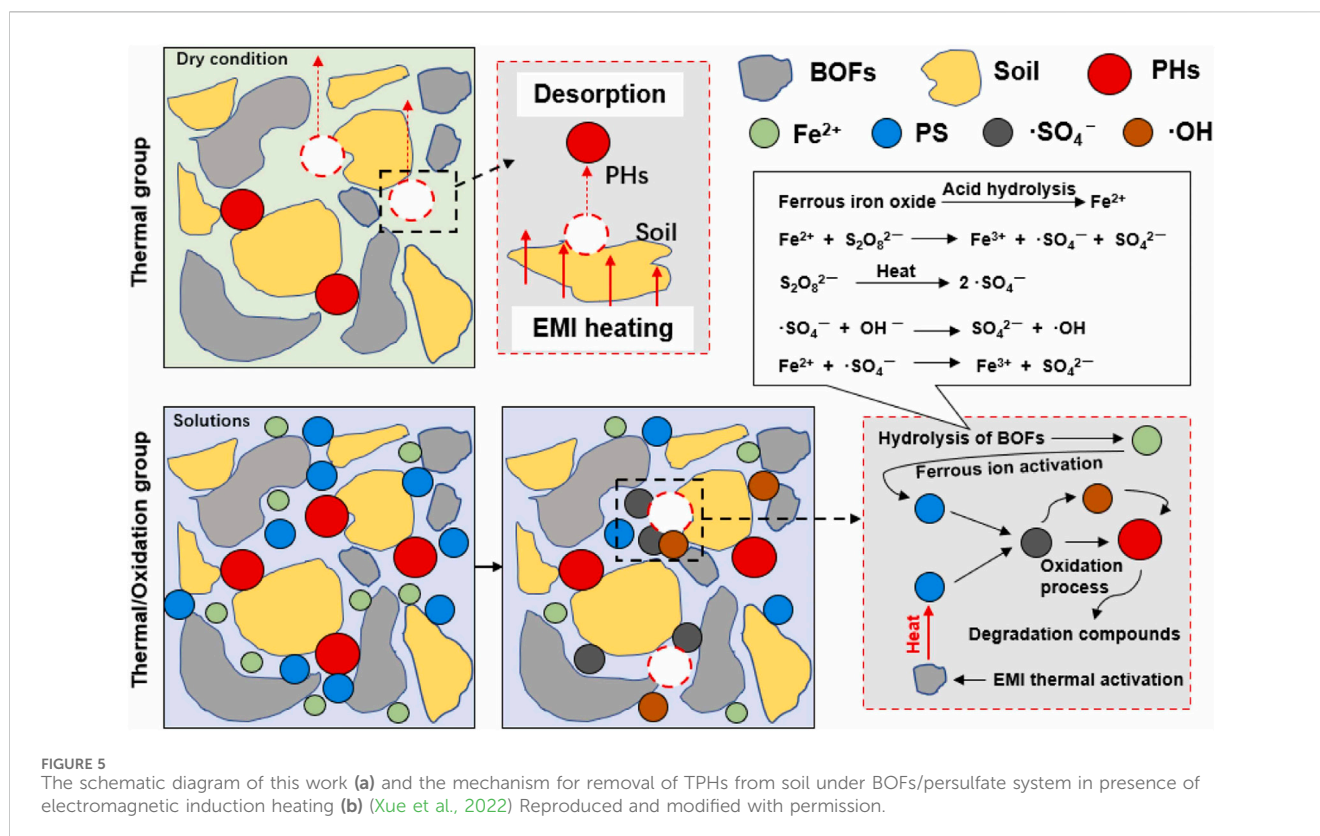


FIGURE 5 The schematic diagram of this work (a) and the mechanism for removal of TPHs from soil under BOFs/persulfate system in presence of electromagnetic induction heating (b) (Xue et al., 2022) Reproduced and modified with permission.

H_2O_2 treatment. The oxidation process was observed to be essentially complete within 60 min, as determined by kinetic experiments. To further enhance the degradation rate, heat-stimulated persulfate activation represents a conventional method for the treatment of TPHs.

Chen et al. employed two different persulfate-based advanced oxidation processes (Figure 3), namely heat/PMS and heat/PDS, for the treatment of TPHs, respectively (Chen et al., 2024). The optimal conditions for the removal of TPHs by the heat/PMS process were found to be 1.48 mmol/g, 80°C, and 120 min, with a degradation rate of approximately 53%. Similarly, the optimal TPHs removal conditions for the heat/PDS process were identified as 3.57 mmol/g, 70°C, and 80 min, which resulted in the degradation of approximately 46%.

Li employed three different activated persulfate methods to treat TPHs in soil (Li et al., 2022), and investigated the effects of dosage, initial pH, temperature, ultrasonic power, and soil water ratio of persulfate on degradation, and the degradation mechanism of petroleum hydrocarbon compounds was also mentioned (Figure 4). After 72 h of treatment. Under the optimal conditions, the degradation efficiency of TPHs by ultrasound/heat activated persulfate (US/Heat/PS) was 78.2%. While the degradation efficiency of TPHs by heat activated persulfate (Heat/PS) system was 72.64%. The degradation efficiency of the single ultrasound activated persulfate (US/PS) system was 56.41%. The findings indicated that the concurrent utilisation of multiple activation methods to facilitate persulfate activation could potentially enhance the degradation rate of TPHs.

Similarly, the use of different novel activation techniques can change the treatment effect of persulfate, also. Xue et al. (2022)

mentioned a novel approach for the remediation of petroleum hydrocarbon-contaminated soil, which is utilising basic oxygen furnace slag (BOFs) and persulfate under electromagnetic induction heating (Figure 5b). Experimental findings demonstrated that persulfate can be effectively activated by BOFs or thermal induction under electromagnetic induction, thereby enhancing the oxidative degradation of BOFs/persulfate and significantly accelerating the TPHs removal rate, the mechanism for removal of TPHs from soil under BOFs/persulfate system in presence of electromagnetic induction heating was given (Figure 5b).

Liu et al. (2023a) investigated the efficacy of a degradation system combining persulfate and microorganisms in the treatment of high concentrations of crude oil ($12,835 \pm 572.76$ mg/kg) contaminated soil. The combined effect of different doses of persulfate (PS) and hydrocarbon-degrading mixed bacteria was investigated as well. The results showed that, use 1% PS oxidation combined with bioremediation, the degradation rate was 80.05% after 180 days of degradation under optimal conditions, and the degradation rate is 4.88% higher than that of single-use biodegradation, 20.94% higher than that of natural attenuation of a single 1% persulfate. The addition of microorganisms has been observed to promote the secretion of enzymes, thereby enhancing the degradation rate of TPHs.

The effectiveness of FeS@BC (iron sulfide@biochar) in activating persulfate (PS) for the remediation of TPHs in petroleum-contaminated soil was systematically investigated (Xia et al., 2022). The optimal TPHs removal efficiency of 61.83% was achieved under the following conditions: a mass ratio of FeS to BC of 1:5, a PS dosage of 0.08 mmol/g, an FeS@BC dosage of 14 mg/g, and

TABLE 2 Chemical oxidative remediation technologies.

Treatment	Hydrocarbon fraction	Removal efficiency	References
H ₂ O ₂ -based Fenton	TPHs and TPHs C ₁₀ -C ₁₃	TPHs, 45.03% and TPHs C ₁₀ -C ₁₃ > 80%	Chen et al. (2022)
H ₂ O ₂ -based Fenton	TPHs (20% biodiesel and 80% diesel)	75%	Pardo et al. (2014)
H ₂ O ₂ -based Fenton	TPHs	87.6%	Akpoveta et al. (2018)
H ₂ O ₂ -based Fenton	TPHs (cable insulating oil)	46%	Xu et al. (2011)
Treatment	Hydrocarbon fraction	Removal efficiency	References
H ₂ O ₂ -based Fenton (electrokinetic-Fenton)	TPHs (oil-based drilling waste)	77%	Adhami et al. (2021)
H ₂ O ₂ -based Fenton (ultrasonic-Fenton)	TPHs (petroleum oil spill sludge)	84.25%	Sivagami et al. (2019)
CaO ₂ -based Fenton	Diclofenac sodium	97.5%	Jiang et al., (2021)
CaO ₂ -based Fenton	TPHs	95.6%	Ndjou'ou and Cassidy (2006)
CaO ₂ -based Fenton	Polycyclic aromatic hydrocarbons (PAH)	92.3%	Gryzenia et al. (2009)
CaO ₂ -based Fenton	BTEX (benzene, toluene, ethylbenzene, and xylene)	CaO ₂ /Fe(II)/BTEX molar ratio of 5/5/1, BTEX removal were 35% CaO ₂ /Fe(II)/BTEX molar ratio of 40/40/1, BTEX removal were 98%	Xue et al. (2018a)
activated persulfate oxidation	TPHs	40.8%	Wu et al. (2020)
activated persulfate oxidation (heat/PMS and heat/PDS)	TPHs	TPHs, heat/PMS = 53%, TPHs, heat/PDS = 46%	Chen et al. (2024)
activated persulfate oxidation (US/Heat/PS and Heat/PS)	TPHs	TPHs, US/Heat/PS = 78.2% TPHs, Heat/PS = 56.41%	Li et al. (2022)
activated persulfate oxidation (PS/microbial)	Crude oil	80.05%	Liu et al. (2023a)
Treatment	Hydrocarbon fraction	Removal efficiency	References
activated persulfate oxidation (FeS@BC/PS)	TPHs	61.83%	Xia et al. (2022)
activated persulfate oxidation (EK-ZVI/PS)	TPHs	27.74%	Li et al. (2024a)
activated persulfate oxidation (Fe/Mn-SOM/PS)	TPHs PAHs	75.74% 80.60%	Cao et al. (2024)

an initial pH value of 5. The study demonstrated that an appropriate amount of BC can enhance the dispersion of FeS and increase the number of active sites. However, an excess of BC can lead to agglomeration, thereby reducing the reaction efficiency. An adequate amount of PS can generate sufficient radicals to degrade TPHs, but an excessive amount of PS can cause radical quenching, which decreases the degradation efficiency. An appropriate amount of FeS@BC can promote the activation of PS, but an excess of FeS@BC can also lead to radical quenching and reduced degradation efficiency. The weakly acidic condition (pH = 5) was found to be the most favorable for TPHs degradation, as it ensures the optimal generation and stability of sulfate radicals (SO₄⁻). Notably, the TPHs removal efficiency of the FeS@BC/PS system was significantly higher than that of the FeS/PS system (47.91%).

Li et al. (2024b) investigated the effectiveness of zero-valent iron (ZVI)-activated persulfate (PS) combined with electrokinetics (EK) for the remediation of TPHs petroleum-contaminated soil. The soil used in the study had a pH of 7.64, an organic matter content of 61.24 g/kg, and a total petroleum hydrocarbon concentration of 12,045 mg/kg. The study explored the impact of ZVI dosage on the

TPHs removal efficiency of the ZVI/PS system, revealing that the optimal removal rate of 11.17% was achieved at a ZVI dosage of 0.95 g (PS:ZVI molar ratio of 10:1). Additionally, the influence of PS injection location on the TPHs removal efficiency of the EK/PS system was examined, with results indicating that simultaneous addition of PS to both the anode and cathode yielded the best performance, achieving a TPHs removal rate of 17.02%. When the EK-ZVI/PS system was employed for TPHs remediation under optimized conditions (0.95 g ZVI, PS added to both electrodes), a TPHs removal rate of 23.72% was obtained. Furthermore, the incorporation of 5% humic acid (HA) into the EK-ZVI/PS system enhanced the TPHs removal rate to 27.74%. The results demonstrated that the EK-ZVI/PS system outperformed the individual ZVI/PS and EK/PS systems in terms of TPHs removal efficiency, highlighting the significant synergistic effect of the combined technology. The optimal TPHs removal rate of 27.74% was achieved under the conditions of 0.95 g ZVI, simultaneous PS addition to both electrodes, and 5% HA addition.

Similarly, for the removal of TPHs from soil (Cao et al., 2024), investigated the effectiveness of *in situ* formed Fe/Mn oxide cross-linked with soil organic matter (Fe/Mn-SOM) complexes in

activating persulfate (PS) for the remediation of TPHs soil. The soil (pH = 7.9) with a TPHs content of 30,421 mg/kg. The Fe/Mn-SOM complexes were formed *in situ* by adding Fe(OH)₂ and Mn(OH)₂ colloidal solutions to the soil, followed by the addition of H₂O₂. After the formation of the Fe/Mn-SOM complexes, PS solution was added to achieve oxidative degradation of TPHs. The study explored the effects of various factors on TPHs degradation efficiency, including the Mn:Fe molar ratio, PS concentration, initial pH, soil-to-water ratio, coexisting anions (Cl⁻, NO₃⁻, HCO₃⁻, H₂PO₄⁻), and organic matter (humic acid, urea, lignin). The results indicated that the highest TPHs removal efficiency of 75.74% was achieved under the conditions of a Mn:F molar ratio of 1:1, PS concentration of 2.0 mmol/L, initial pH of 7, and a soil-to-water ratio of 4:1. This efficiency was significantly higher than that of systems using PS alone (12.52%), Fe²⁺/PS (22.64%), Mn²⁺/PS (18.63%), and Fe²⁺+Mn²⁺/PS (43.46%). Notably, the system also demonstrated a high removal rate for polycyclic aromatic hydrocarbons (PAHs) at 80.60%. The study found that the *in situ* formed Fe/Mn-SOM complexes could directly contact petroleum hydrocarbons in the soil, avoiding the ineffective consumption of free radicals during diffusion. The Fe/Mn-SOM complexes exhibited a certain tolerance to coexisting anions and organic matter, but high concentrations of HCO₃⁻, H₂PO₄⁻, urea, and lignin significantly reduced TPHs removal efficiency. As an *in situ* formed PS activator, the Fe/Mn-SOM complexes are characterized by high efficiency, cost-effectiveness, and environmental friendliness, providing a novel technical approach for the remediation of petroleum hydrocarbon-contaminated soil.

The chemical oxidative remediation techniques in this paper are summarized below (Table 2).

5 Conclusion

The chemical oxidation remediation of total petroleum hydrocarbons (TPHs) in soil primarily relies on the use of strong oxidants, which generate oxidative radicals under various activation conditions. These radicals facilitate the oxidative degradation of TPHs. This paper reviews the mechanisms and remediation efficiencies of hydrogen peroxide-based Fenton oxidation, calcium peroxide-based Fenton and Fenton-like systems, and activated persulfate oxidation in the context of TPH-contaminated soil. The hydrogen peroxide-based Fenton oxidation system is highly sensitive to the pH of the reaction environment and is primarily suitable for soil remediation under acidic conditions. However, its applicability and degradation efficiency can be enhanced by combining it with different activation methods. The calcium peroxide-based Fenton and Fenton-like systems serve as alternative technologies to the hydrogen peroxide Fenton process, offering a broader pH range and addressing the limitations of hydrogen peroxide in neutral to alkaline environments. Notably, activated persulfate oxidation

exhibits advantages such as strong oxidative capacity, long-term stability, and diverse activation methods. To improve the removal efficiency of TPHs in soil and develop effective TPHs oxidation systems, future research should focus on leveraging the high oxidative potential of activated persulfate systems by selecting composite oxidation systems, such as PS/CaO₂ or PS/H₂O₂, and optimizing activation methods, including efficient thermal activation, iron-based composite activation, and alkaline activation. The integration of multiple activation techniques and novel activation methods represents a critical research direction for enhancing TPHs removal efficiency.

Author contributions

YD: Formal Analysis, Funding acquisition, Writing—original draft, Writing—review and editing. ZL: Conceptualization, Data curation, Formal Analysis, Funding acquisition, Writing—review and editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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