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An overview of bioelectrokinetic and bioelectrochemical remediation of petroleum-contaminated soils

Jun Lan ^{a,1}, Fang Wen ^{b,1}, Yongxiang Ren ^a, Guangli Liu ^c, Yi Jiang ^d, Zimeng Wang ^e, Xiuping Zhu ^{e,*}^a Shaanxi Key Laboratory of Environmental Engineering, School of Environmental and Municipal Engineering, Xi'an University of Architecture and Technology, Xi'an, 710055, China^b Xinjiang Academy of Environmental Protection Science, Urumqi, 830011, China^c Guangdong Provincial Key Laboratory of Environmental Pollution Control and Remediation Technology, School of Environmental Science and Engineering, Sun Yat-sen University, Guangzhou, 510006, China^d Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Kowloon, Hong Kong, China^e Department of Environmental Science and Engineering, Fudan University, Shanghai, 200433, China

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ABSTRACT

The global problem of petroleum contamination in soils seriously threatens environmental safety and human health. Current studies have successfully demonstrated the feasibility of bioelectrokinetic and bioelectrochemical remediation of petroleum-contaminated soils due to their easy implementation, environmental benignity, and enhanced removal efficiency compared to bioremediation. This paper reviewed recent progress and development associated with bioelectrokinetic and bioelectrochemical remediation of petroleum-contaminated soils. The working principles, removal efficiencies, affecting factors, and constraints of the two technologies were thoroughly summarized and discussed. The potentials, challenges, and future perspectives were also deliberated to shed light on how to overcome the barriers and realize widespread implementation on large scales of these two technologies.

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1. Introduction

Petroleum contamination in soils has been reported as one major pollution problem after pesticide pollution, with the intensive development of petroleum prospecting, crude oil exploration, and downstream petroleum industries, such as petrochemical refinery companies [1]. It is mainly caused by the ingredients in oil from the processes of gathering, transportation, and storage, especially from leakages and spills, such as the Exxon Valdez oil spill in 1989, the M/T Haven Tanker oil spill in 1991, and the Deepwater Horizon oil spill in 2010 [2]. Petroleum-contaminated soils contain a lot of petroleum hydrocarbons and their derivatives. Most of them are persistent and harmful [3]. Among them, polycyclic aromatic hydrocarbons (PAHs) are of particular concern because of their diverse structural configurations, low

biodegradability, hydrophobic nature, strong sorption phenomena, and high persistence [4]. Another group of contaminants, halogenated hydrocarbons derived from petroleum hydrocarbons, are also of great concern as irritating odor and toxicity [5]. Besides causing soil pollution, petroleum hydrocarbons and their derivatives may also affect the soil properties, such as mechanical properties, Atterberg limits, shear strength, texture, porosity, permeability, and microbial communities [6]. For these reasons, numerous severe ecological and environmental consequences likely occur. Since the soil is a natural resource that cannot be renewed, remediation of petroleum-contaminated soils should be mandatorily enforced via strict regulations in most countries [7].

Soil remediation technologies can be broadly classified into physicochemical, chemical, thermal, and biological technologies according to the main working principle they are based on. Among these remediation technologies, bioremediation is becoming a hotspot for research and practical application because of some advantages compared to other technologies, such as low expense, easy implementation, and environmental benignity [8]. Bioremediation uses microbes to degrade and detoxify hydrocarbon-

* Corresponding author.

E-mail address: xpzhu@fudan.edu.cn (X. Zhu).¹ These authors contributed equally.

polluted environments, mitigating the risks to human health and ecosystems. It has been playing an increasing role in the treatment of different petroleum-contaminated matrices, such as soil, sludge, sediment, and groundwater [9]. However, the efficiency of bioremediation is limited by many factors, including the activity of microbes, the availability of contaminants, and environmental conditions [10]. Collective implementation of two or more methods was suggested to overcome the disadvantages of a single technology, leading to the development of integrated remediation technologies.

Bioelectrokinetic and bioelectrochemical remediations are two hybrid technologies in which electric fields are used to enhance bioremediation but function in different ways. In bioelectrokinetic remediation, electrokinetics promotes the transportation of microbes, nutrients, and contaminants (e.g., electromigration, electroosmosis, electrophoresis, etc.) by applying a direct-current electric field for bioaugmentation and biostimulation [11]. Some microbes (e.g., *Geobacter* and *Shewanella* species) are capable of constructing direct electrochemical communication with external solids [12]. It makes it possible to enhance bioremediation performance by inserting electrodes into the contaminated soils to serve as electron acceptors or donors in a so-called bioelectrochemical remediation system [13]. The soil microbial fuel cells (soil-MFCs) are a typical form of bioelectrochemical remediation systems in which electricity is generated instead of consumed by converting the chemical energy from contaminant degradation into electricity [14]. Compared to other bioremediation techniques requiring adding chemicals as electron donors/acceptors, the soil-MFCs have several advantages, such as lower energy consumption, less damage to the original structure, and less impact on indigenous microbes [15].

Bioelectrokinetic remediation has drawn increasing attention during the past two decades. It is becoming popular in the treatment of soils contaminated by hydrophobic organic compounds and has been used to treat contaminated soils with low or medium permeability that is hard to be treated by other traditional technologies [16]. There are already some full-scale applications of bioelectrokinetic remediation for petroleum-contaminated soils [17,18]. Bioelectrochemical remediation is relatively new and has been considered a desirable new option for soil remediation, although no full-scale treatment has been performed so far. Several reviews have been reported on bioelectrokinetic or bioelectrochemical remediation for removing contaminants from different matrices [5,14,19]. However, no review has compared these two similar technologies from the same perspective for soil remediation contaminated by petroleum hydrocarbons and their derivatives. Therefore, this review focuses on analyzing the efficiencies, affecting factors, and constraints of bioelectrokinetic and bioelectrochemical remediation for petroleum-contaminated soils and comprehensively understanding the potentials, challenges, and future perspectives of these two technologies. This review aims to provide useful information for researchers and practitioners in this field to overcome the barriers and realize the widespread implementation of these two technologies.

2. Working principles of bioelectrokinetic and bioelectrochemical remediation

2.1. Bioelectrokinetic remediation

Bioelectrokinetic remediation, also called electrokinetic-enhanced bioremediation or electro-bioremediation in some studies, aims to stimulate the migration and transformation of microbes, nutrients, and organic contaminants in the subsurface matrix by applying a weak electric field (less than 1 V cm^{-1}) [20,21].

The electrokinetic processes can enhance the biodegradation of dissolved-phase organic contaminants in the soils by abiotic effects. The specific transport phenomena driven by electrokinetics include electromigration, electroosmosis, and electrophoresis, as explained by the bioelectrokinetic remediation (BIO-EK) model (Fig. 1) [22]. Electromigration is one main mechanism in bioelectrokinetic remediation, which is the movement of charged substances, such as charged contaminants, SO_4^{2-} , NO_3^- , and NH_4^+ ions, toward the electrodes of opposite charges [23]. Electroosmosis is the migration of pore fluids that are capillary water in soil particles combined with free-phase organic contaminants. Generally, the pore fluids are able to transfer from the anode to the cathode by an electric double layer [24]. Electrophoresis refers to the transport of charged biological colloids moving towards the oppositely charged electrode under the action of electric fields. Electromigration often dominates mass transport in electrokinetic processes because its speed ($3.625 \times 10^9 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) is at least one order of magnitude faster than that of electroosmosis and electrophoresis [21]. However, the electromigration and electroosmosis rates are strictly related to soil permeability. Especially in low-permeability soil, electroosmosis plays a leading role [25]. The migration velocities of microbes are mainly driven by electromigration and electroosmosis [26]. The electrokinetic transport will also be affected by other factors. For example, the electromigration of microbes is generally fast in high permeability media, such as sandy soil ($0.5 \text{ cm}^2 \text{ V}^{-1} \text{ h}^{-1}$) [27]. Migration velocities of single inoculum (PAHs-degrading bacteria *Sphingomonas*) were within the scope of $0.05\text{--}0.15 \text{ cm}^2 \text{ V}^{-1} \text{ h}^{-1}$, mainly influenced by cell surface charge and attachment to sediments [28]. Due to electroosmosis transport, hydrophobic contaminants are more mobile in lower permeability clays with migration velocities in the range of $0.16\text{--}0.26 \text{ cm}^2 \text{ V}^{-1} \text{ h}^{-1}$ [29].

The bioavailability of contaminants limits the effect of bioremediation of contaminated soils, especially in clayey soils with a larger specific surface area and electronegativity to absorb organic contaminants. Electrokinetic processes can be used to overcome this kind of limitation [30]. The application of weak electric fields increases the temperature inside soils and promotes the production of natural biosurfactants from microbes [31]. The electrokinetic injection can also realize efficient transport of biosurfactant into low permeability soils, resulting in more feasible interactions between microbes and contaminants. It can also improve the bioaugmentation effect of remediation by directionally injecting

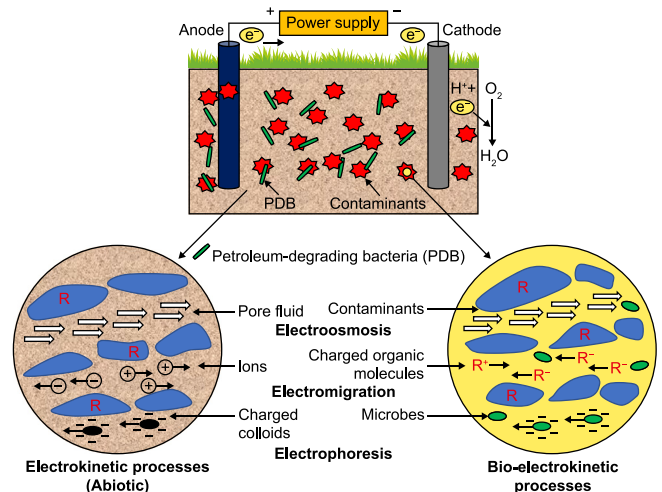


Fig. 1. Schematic diagram of bioelectrokinetic remediation processes [22].

specific bacteria into the polluted zones. Wick et al. investigated the mobility and activity of PAHs degrading bacteria in electric fields. The results showed electroosmosis and electrophoresis could be combined with *in situ* biological treatment to move contaminant-degrading bacteria to highly polluted areas [28]. However, the issue in bioelectrokinetic remediation is that extreme pH conditions and salinity accumulation. This issue is because acid will be formed close to the anode, and base will be produced near the cathode due to the water electrolysis. According to different configurations and operating conditions, bioelectrokinetic systems were classified as single electrode tanks [32], multi-electrode tanks [33], recirculation tanks [34], bio-barriers tanks [35], and polarity-reversal tanks [36], as shown in Fig. 2. All of them have their own advantages and disadvantages, which will be discussed in Section 4.1.1 on affecting factors. In a word, these devices can accelerate the movement of organics, nutrients, bacteria, and pore fluids in the soil matrix and stimulate interactions among organic contaminants and petroleum-degrading bacteria during bioelectrokinetic remediation.

2.2. Bioelectrochemical remediation

Bioelectrochemical systems (BESs) have been extensively developed for wastewater treatment over the past decades as electricity or useful substances (e.g., methane, hydrogen, and formic acid) are simultaneously produced [37,38]. Bioelectrochemical remediation couple bioremediation with electrochemical remediation, in which bioremediation is enhanced by electrochemically active bacteria (EAB) exchanging electrons with the electrodes and generating a bioelectric field [39]. The mechanisms of extracellular electron transfer between electrodes and microbes during bioelectrochemical remediation have three ways: EAB direct contact with the electrodes (giving electrons to the anode or obtaining electrons from the cathode), by indirect mechanisms where a chemical compound acts as an electron shuttle that the microbes can secrete (e.g., pyocyanin and phenazine generation), or added exogenously (e.g., humic substances, sulfate, and nitrate) [40,41]. Some microbes (e.g., *Shewanella oneidensis* and *Geobacter*

sulfurreducens) can also form cellular pili (nanowires) for extra-cellular electron transfer [42]. When electrodes serve as the electron acceptor, electrons produced from contaminant degradation by EAB are transferred to the anode, and electricity is produced by coupling oxygen or other species reduction at the cathode [43]. When electrodes work as the electron donor, electrons produced from the anode can be used by microbes from the bio-cathode.

Based on previous studies, there are four possible removal mechanisms of organic pollutants from soils by bioelectrochemical remediation (Fig. 3) [44]: (a) bio-oxidation at the anode [45], (b) reduction at the cathode [46], (c) contaminants sorption [9], and (d) electrokinetic migration [47]. Biodegradation is the most important mechanism in the bioelectrochemical remediation of petroleum-contaminated soils. Under the bioelectric field, the chemical morphology and distribution of contaminants are changed, and contaminants are adsorbed from the soils onto the electrodes or the anodic biofilm. Relevant research revealed the quantitative profile of soluble ions migration in the bioelectric field. In the case of divalent heavy metal ions in the contaminated soil, heavy metal ions will change chemical morphology and deposit near the cathode under the bioelectric field [48]. Petroleum hydrocarbons and their derivatives can be carbon sources and electron donors for EAB.

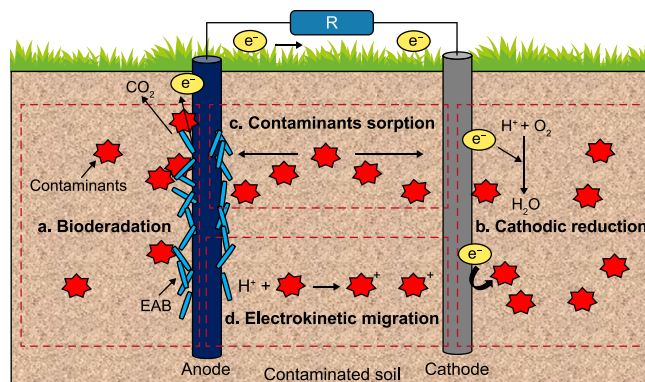


Fig. 3. Schematic diagram of bioelectrochemical remediation processes [44].

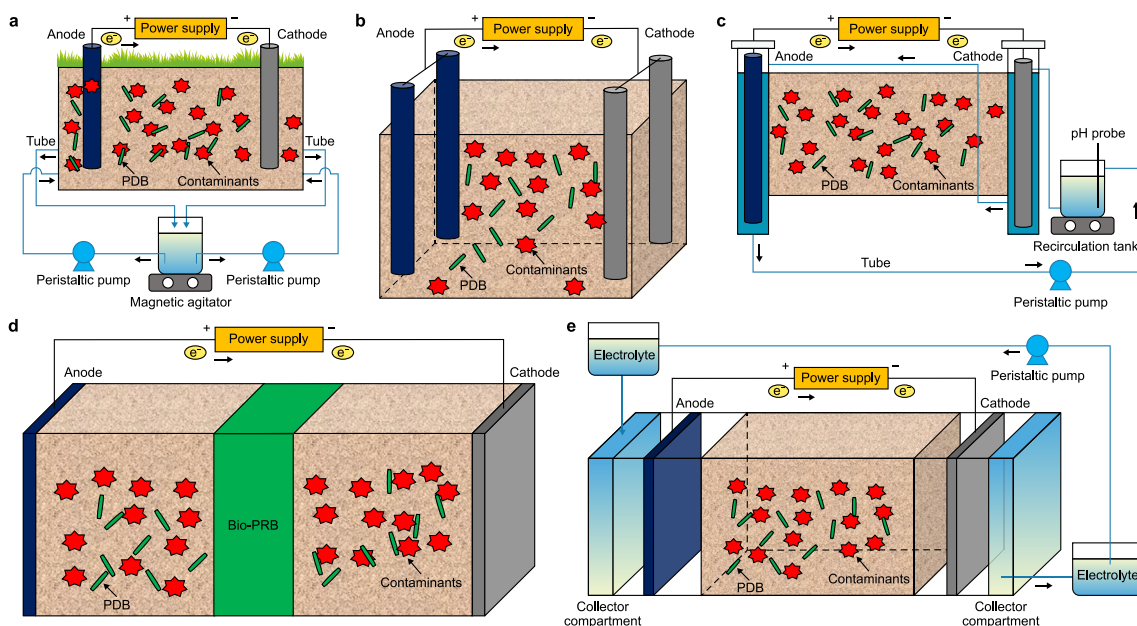


Fig. 2. Schematic diagrams of typical bioelectrokinetic systems. a, Single electrode tank [32]; b, Multi-electrode tank [33]; c, Recirculation tank [34]; d, Bio-barriers tank [35]; e, Polarity-reversal tank [36].

The consortium of different groups of bacteria, including EAB, degrade the contaminants and give electrons to the anode, and the electrons transfer through an external circuit to the cathode, where a final electron acceptor (e.g., O_2) is reduced [49]. For example, under anaerobic conditions, the aromatic hydrocarbons can be degraded to intermediate molecules and linear hydrocarbons with small molecular weight at the anode by iron-reducing bacteria [50].

In recent years, the bioelectrochemical technology used for petroleum-contaminated soil remediation has attracted more and more attention. The soil-MFC is a typical bioelectrochemical remediation system consisting of an anode, a cathode, a separator (optional), and an external circuit. An anode is embedded in the soil, and a cathode is exposed to air. It is very promising for *in situ* soil remediation as no external energy is needed, and even electricity is produced. Theoretically, the open-circuit voltage of soil-MFCs was around 1.1 V with an anode potential of -0.3 V and a cathode potential of 0.8 V (relative to Normal Hydrogen Electrode, NHE) [51]. However, due to the loss of various overpotentials, the actual open-circuit voltage measured in the laboratory is only about 0.7–0.8 V [52]. Soil-MFCs can be designed in different shapes according to the need of a specific application, making the implementation much more convenient. Due to the simple installation and easy maintenance, the tank-type reactor is widely used in petroleum-contaminated soil remediation lab tests (Fig. 4a) [53–55]. Other types, such as column types (Fig. 4b) [56], U-types (Fig. 4c) [45], multi-anodes types (Fig. 4d) [57], and double-chamber types (Fig. 4e) [58], were also used in some studies. They all have their own characteristics, which will be discussed in Section 4.2.1 on affecting factors.

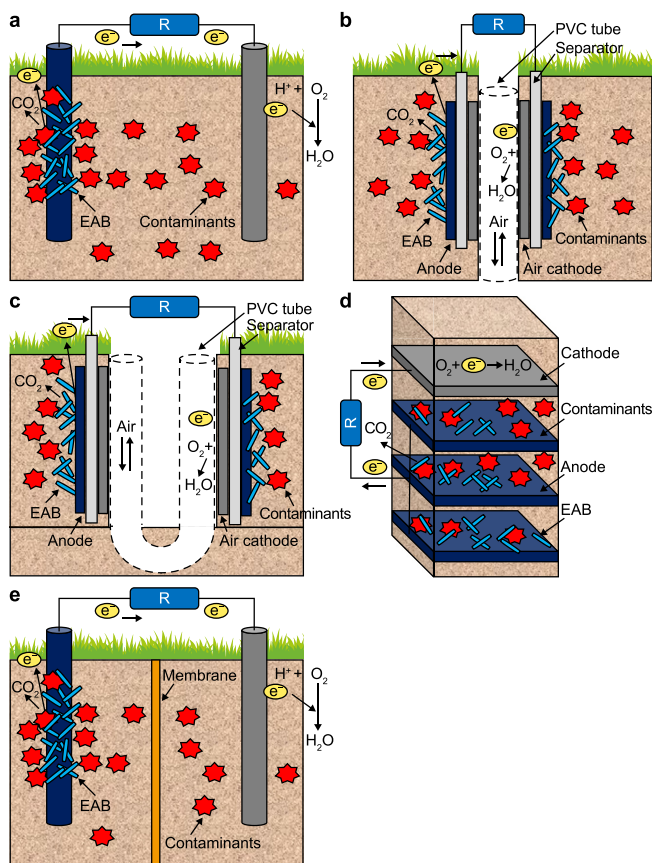


Fig. 4. Typical schematic diagrams of bioelectrochemical remediation systems. a, Tank type [53–55]; b, Column type [56]; c, U type [45]; d, Multi-anodes type [57]; e, Double-chamber type [58].

Except for typical soil-MFCs, other bioelectrochemical remediation systems could also be used for soil remediation, such as microbial electrochemical snorkels (MESs) and microbial electrolysis cells (MECs), that have already been used for the treatment of marine sediments polluted with hydrocarbons [59,60]. MES is a special MFC operated in short-circuited mode, based on removing contaminants with electrochemical reaction, but the system does not produce power [61]. An MES with graphite rod electrodes was conducted on hydrocarbon-contaminated sediments, and the results showed the removal of total petroleum hydrocarbons (TPHs) enhanced by 12% within 200 days [62]. MEC is an emerging technology for efficiently converting multifarious organic wastes to biogases (e.g., hydrogen and methane) by applying an additional voltage between the bioanode and the cathode [63]. In recent reports, MECs were applied to study the removal mechanism and removal efficiency of emerging contaminants (e.g., phenol, quinoline, and tetracycline) in water/sediments [64,65]. Therefore, these two technologies can potentially be applied in the remediation of petroleum-contaminated soil in the future.

3. Removal efficiencies of bioelectrokinetic and bioelectrochemical remediation

3.1. Removal efficiencies of bioelectrokinetic remediation

Bioelectrokinetic remediation has been used to remove different petroleum hydrocarbons and their derivatives from naturally or artificially contaminated soils, such as TPHs, PAHs, diesel, phenol, and 2,4-dichlorophenol, in different soil types with different electrode materials (Table 1).

TPHs and PAHs are used to trace soil contamination by petroleum hydrocarbons in most studies. The removal efficiencies of TPHs ranged 72.8–89% in artificially contaminated soils within the initial TPHs range of 6.8 – 50 $g\ kg^{-1}$ (Table 1). Crognale et al. [66] applied the 1.0 $V\ cm^{-1}$ on bioelectrokinetic remediation of diesel-contaminated soils. After 50 days, the TPHs and PAHs removal efficiencies reached 76% and 79%, with the initial TPHs and PAHs being 0.48 and 0.11 $g\ kg^{-1}$, respectively. The highest removal efficiency of 89% was achieved at an applied electric field of 0.48 $V\ cm^{-1}$ after 700 h in sandy loam with contaminated TPHs [67]. The relationship between the removal efficiency of TPHs with the distance of anode in diesel-contaminated soils was studied by Kim et al. [21]. The results indicated that the removal rate decreased with the increase of electrode distance, and the highest removal efficiency of TPHs was 73% near the anode after 25 days under 1 $V\ cm^{-1}$. Bioelectrokinetic remediation showed satisfactory removal efficiencies on phenanthrene, a typical of PAHs. Over 80% of the initial 200 $mg\ kg^{-1}$ phenanthrene in soils was removed in 20 days with electrokinetic injection as well as electrolyte circulation (2 $g\ L^{-1}$ NH_4NO_3 and 2 $g\ L^{-1}$ KH_2PO_4) and electrode polarity reversal, which were used to control pH and realize uniform distribution of inorganic ions [32]. The phenanthrene removal efficiency increased ten-fold with an applied voltage of 0.6 $V\ cm^{-1}$ in naturally contaminated soils with 78.5 $mg\ kg^{-1}$ phenanthrene, and the maximum removal efficiency was over 98% in 14 days [68]. Total organic carbon (TOC) can also be used to trace the pollution level in petroleum-contaminated soils. A study showed 82% removal efficiency in petroleum-contaminated soils in 25 days with a voltage gradient of 1.0 $V\ cm^{-1}$ compared to 36% without any electric field [69]. Additionally, petroleum and heavy metal co-contaminated soils can be simultaneously treated by bioelectrokinetic remediation. Under 2.0 $V\ cm^{-1}$, the TPH removal efficiencies reached 89% after 30 days, with the initial TPHs being 12.5 $g\ kg^{-1}$ [70].

Bioelectrokinetic remediation of diesel-contaminated soils has been a research hotspot as diesel fuel is a major contamination

Table 1
Removal efficiencies of bioelectrokinetic remediation.

Soil type	Contamination type (artificial/natural)	Targeted pollutants	Initial contamination level (g kg ⁻¹)	Types tank	Electrode materials	Applied electric field	pH control measure	Maximum removal (BEK vs. EK)	Duration (days)	Year	Reference
Clay	Natural	TPHs, PAHs, and phenanthrene	TPHs 2.78, PAHs 4.50, phenanthrene 1.32	Recirculation tank	Stainless steel	0.2 –0.6 V cm ⁻¹	Buffer in both chambers	1.32 mg kg ⁻¹ h ⁻¹ vs. 0.140 mg kg ⁻¹ h ⁻¹	14	2006	[68]
Sandy soil	Natural	TPHs	6.8	Single electrode tank	Graphite	1.0 V cm ⁻¹	No	73% vs. 31%	25	2010	[21]
Sandy, silty, and clayed soil	Artificial	TPHs	12.5	Recirculation tank	Pt-coated titanium anode, carbon plate cathode	2.0 V cm ⁻¹	Electrolyte, pH 8.0	80% vs. No	30	2013	[70]
Sandy soil	Natural	TPHs	59.2	Single electrode tank	Stainless steel	0.48 V cm ⁻¹	No	89% vs. 25%	700 h	2014	[67]
Sandy soil	Natural	TPHs	9	Recirculation tank	Titanium plate	1.0 V cm ⁻¹	Inorganic salt solution	77% vs. 66%	30	2018	[77]
Sandy soil	Artificial	TPHs	50	Recirculation tank	Stainless steel	1 V cm ⁻¹	Inorganic salt solution	72% vs. 48%	98	2020	[78]
Clayey and sandy soil	Natural	TPHs and PAHs	0.48 and 0.11	Polarity-reversal tank	Stainless steel	1 V cm ⁻¹	Polarity reversal	76% vs. 64%, 79% vs. 56%	50	2020	[66]
Silty, clayey, and sandy soil	Artificial	PAHs	150 mL of oil per kg of soil	Recirculation tank	Graphite	1 V cm ⁻¹	Inorganic salt solution	92% vs. 67%	140	2022	[79]
Sandy soil	Artificial	Diesel	4	Recirculation tank	Graphite	0.83 mA cm ⁻²	Buffer and recirculating electrolyte	50–60% vs. 0.5 –2%	8	2001	[74]
Kaolinite clay	Artificial	Diesel	10	Bio-barriers tank	Graphite	1.0 V cm ⁻¹	No	39% vs. No	336 h	2015	[35]
Kaolinite clay	Artificial	Diesel	10	Polarity-reversal tank	Graphite	1.0 V cm ⁻¹	Polarity reversal	29% vs. 12%	14	2015	[36]
Kaolinite clay	Artificial	Diesel	10	Polarity-reversal tank	Graphite	0–1.5 V cm ⁻¹	Polarity reversal	35% vs. 11%	14	2016	[73]
Sandy soil	Artificial	Diesel	10	Polarity-reversal tank	Ti/RuO ₂ anode, stainless steel cathode	1.0 and 2.0 V cm ⁻¹	Polarity reversal	44% vs. 33%, 67% vs. 43%	16	2018	[72]
Silty, clayey, and sandy soil	Artificial	Diesel	20	Polarity-reversal tank	Graphite	1 V cm ⁻¹	Polarity reversal	20–30% vs. 10 –12%	55	2019	[71]
Clay	Artificial	Diesel	10	Single electrode tank	Titanium	2 V cm ⁻¹	No	84% vs. 67%	4	2021	[80]
Sandy soil	Artificial	Petroleum	50	Polarity-reversal tank	Graphite	1.0 V cm ⁻¹	Polarity reversal	46% vs. 28%	100	2010	[81]
Sandy soil	Artificial	Petroleum (expressed as TOC)	156	Single electrode tank	Column-shaped carbon	0.33 –1 V cm ⁻¹	No	82% vs. 36%	15	2017	[69]
Silty, clayey, and sandy soil	Artificial	Engine oil	150	Single electrode tank	Graphite	0.9 V cm ⁻¹	No	50% vs. No	240 h	2020	[18]
Sandy soil	Artificial	Crude oil	Not mentioned	Single electrode tank	IrO ₂ –RuO ₂ –TiO ₂ /Ti anode, Ti plate cathode	2.0 V cm ⁻¹	No	92% vs. 60%	48 h	2021	[82]
Sandy soil	Artificial	Phenol	0.18	Polarity-reversal tank	Graphite	1.0 V cm ⁻¹	Polarity reversal	58% vs. 10%	10	2006	[26]
Sandy soil	Artificial	Phenol	0.2	Polarity-reversal tank	Graphite	1.1 V cm ⁻¹	Polarity reversal	67% vs. 14%	10	2005	[75]
Sandy soil	Artificial	Perchloroethylene (PCE)	0.06	Recirculation tank	Graphite	1.0 V cm ⁻¹	0.01 M sodium carbonate	99% vs. 31%	10	2006	[83]
Sandy soil	Artificial	2,4-dichlorophenol	0.1	Polarity-reversal tank	Column-shaped graphite	1.0 V cm ⁻¹	Polarity reversal	73% vs. less than 20%	15	2007	[10]

(continued on next page)

Table 1 (continued)

Soil type	Contamination type (artificial/natural)	Targeted pollutants	Initial contamination level (g kg ⁻¹)	Types tank	Electrode materials	Applied electric field	pH control measure	Maximum removal (BEK vs. EK)	Duration (days)	Year	Reference
Sandy, silty, and clayed soil	Natural	TOC	10.8	Single electrode tank	Stainless steel	1.0 V cm ⁻¹	No	99% vs. No	60 h	2007	[84]
Clay	Artificial	Phenanthrene	0.20	Single electrode tank	Graphite plates	1.0 V cm ⁻¹	Polarity reversal	Over 80% vs. less than 20%	20	2010	[32]
Clay	Natural	Chlorinated ethene	0.142 and 0.464	Recirculation tank	Graphite plates	0.19 mA cm ⁻²	No	Not mentioned	20	2012	[34]
Sandy soil	Artificial	N-hexadecane	1.0% (v/w)	Single electrode tank	Graphite	1.3 V cm ⁻¹	No	54% vs. 30%	42	2013	[85]
Silty, clayey, and sandy soil	Artificial	Pyrene	0.15	Multi-electrode tank	Graphite	1.0 V cm ⁻¹	Polarity reversal	55% vs. 10%	42	2016	[33]

source. The research showed good effectiveness of bioelectrokinetics in rehabilitating silty clay contaminated with diesel [71]. The removal efficiencies of diesel ranged 20–67% (Table 1). Song et al. showed that diesel removal was positively correlated with the voltage gradient. After 15 days of treatment, the diesel removal efficiency increased from 44% with an applied voltage of 1 V cm⁻¹ to 67% with 2 V cm⁻¹ [72]. A homogenous diesel removal of approximately 39% was realized with an applied voltage of 1.0 V cm⁻¹ after 336 h. Continuous replacement of surfactants, inorganic nutrients, and a buffer solution was recommended for a long-time operation by the authors [35]. Compared with 10.5% of removal efficiency by bioremediation alone, diesel removal increased to 35.4% after 14 days by the periodic reversal of the electric field (1.5 V cm⁻¹) and continuous replacement of nutrients [73]. Another research showed up to 60% diesel removal in eight days was observed with an applied electric field of 0.83 mA cm⁻² under the condition of 0.05 M phosphate buffer electrolyte with a pH of 7 [74]. Besides bioaugmentation and biostimulation, electrokinetics can also be combined with permeable bio-barriers. Ramirez et al. [36] combined the electrokinetic soil flushing and permeable reactive biological barrier for the remediation of diesel-contaminated soils. After only 14 days of treatment, diesel removal of 29% was achieved with an initial concentration of 10 g kg⁻¹ at 1.0 V cm⁻¹.

Removal efficiencies of specific contaminants from contaminated soils by bioelectrokinetic remediation were also studied, such as phenol and chlorinated organics (Table 1). The removal efficiency of phenol reached 58% in ten days when the polarity of the electric field (1.0 V cm⁻¹) was reversed every 3 h in bioelectrokinetic remediation of phenol-contaminated soils with an initial concentration of 180.2 mg kg⁻¹ [26]. In another study, a maximum phenol removal efficiency of 67% was obtained after ten days in a bioelectrokinetic system with a periodic polarity reversal every 1.5 h [75]. A shorter interval of polarity-reversal showed better performance, but it would increase energy consumption [76]. Chlorinated organics can also be removed by bioelectrokinetic remediation. Biodegradation of 2,4-dichlorophenol (2,4-DCP) in soils was enhanced to 67% in the middle region by bidirectional operation of a 1 V cm⁻¹ electric field with a polarity reversal every 1.5 h [10]. Perchloroethylene decreased from 142 to 464 to 1.7–51 mg kg⁻¹ by the combination of electrokinetics and bioremediation with *Dehalococcoides* bacterial strains [34].

These results demonstrated that bioelectrokinetic remediation effectively removes different petroleum hydrocarbons and their derivatives, but the removal efficiencies varied in big ranges

depending on different operating conditions, which will be discussed in the following section. Moreover, it should be noted that most studies mentioned above were conducted with artificial soils, while only six studies examined the removals of TPHs using naturally contaminated soils. In practical application, the removal efficiencies in naturally contaminated soils might be lower than those in spiked soils.

3.2. Removal efficiencies of bioelectrochemical remediation systems

Bioelectrochemical remediation of petroleum-contaminated soils became popular only in the recent decade, but it has been tested to remove various contaminants, such as TPHs, PAHs, *n*-alkanes, and phenol, by using different configurations (Table 2).

Most studies used TPHs as the targeted contaminant in the bioelectrochemical remediation of soils contaminated by petroleum hydrocarbons. The removal efficiencies of TPHs ranged 15–90% (Table 2). Because of the negative effects of salinization around electrodes, the removal efficiency of TPHs only increased from 7% to 15% in U-type soil-MFCs with a weak power generation of 0.85 mW m⁻² in 25 days [45]. Horizontal multiple-anodes soil-MFCs also realized TPHs removal of 22% in 135 days compared to 6% in control [86]. TPH removal efficiencies and electricity generation were further enhanced to 30% and 17.3 mW m⁻² by using graphite rod anodes in column-type soil-MFCs for the remediation of natural petroleum-contaminated soils [56]. Wang et al. demonstrated that double-chamber soil-MFCs could be effective in crude oil degradation, and the removal efficiencies of TPHs increased from 22% to 37.5% within 137 days [58]. A column-type soil-MFCs with carbon cloth anodes showed a power generation of 132 ± 17 mW m⁻². The removal efficiencies of TPHs were enhanced by 52%, with 182 days [87]. Meanwhile, the dramatic enrichment in anodic denitrifying bacteria enhanced the denitrification process in this system. However, high resistance is a limiting factor in increasing the removal efficiencies of TPHs in soil-MFCs, which can be overcome by different amendments. The TPH removal in the petroleum-contaminated soil was correlated well with the variation of electrical conductivity; that is, a lower conductivity value reflected a lower TPH removal [53]. The maximum power density increased from 81 to 304 mW m⁻² in multi-anodes soil-MFCs with saline-alkali soils contaminated by petroleum hydrocarbons by adding carbon fiber (2%), in which the maximum removal of TPHs was enhanced to 60% after 65 days [55]. Recently, there was a nearly 67% increment in removal efficiencies of TPHs by adding chemical

Table 2
Removal efficiencies of bioelectrochemical remediation.

Soil type	Contamination type (artificial/natural)	Targeted pollutants	Initial contamination level (g kg ⁻¹)	BESs type	Electrode materials	Maximum power/current output	Maximum removal efficiency (%) vs. control	Duration (days)	Year	Reference
Sandy soil	Natural	TPHs	12.25	Column-type	Graphite granule or biochar anode, stainless steel mesh cathode	70.4 mA m ⁻²	82–90% vs. 68%	120	2014	[91]
Silty, clayey, and sandy soil	Natural	TPHs	11.46	Column-type	Carbon cloth or biochar anode-activated carbon cloth with catalyst layers cathode	73–86 mA m ⁻²	79% vs. 43%	64	2014	[92]
Sandy soil	Natural	TPHs	25.7	Multi-anodes type	Four layers of carbon mesh anodes, stainless steel mesh with a catalyst layer cathode	A total charge of 833 C	15% vs. 6%	135	2015	[57]
Sandy soil	Natural	TPHs	Not mentioned	Multi-anodes type	Carbon mesh anodes, activated carbon cathode	49 mW m ⁻²	22% vs. 6%	135	2015	[86]
Sandy and clayey soil	Natural	TPHs	6.50–6.96	Tank-type	Carbon felt anodes, carbon cloth with activated carbon catalyst cathode	26 mA m ⁻²	Sandy soil: 41% vs. 25%, Clayey soil: 23% vs. 15%	230	2016	[53]
Naturally clayey soil	Natural	TPHs	Not mentioned	Multi-anodes type	Carbon mesh anode-activated carbon air-cathodes	304 mA m ⁻²	60% vs. 22%	65	2016	[55]
Sediment soil	Natural	TPHs	Not mentioned	Dual-phase type	Graphite felt anode, active carbon felt cathode	29.8 mW m ⁻²	31% vs. 26%	45	2019	[93]
Naturally silty soil	Natural	TPHs	83 ± 3	Column-type	Carbon cloth-activated carbon air-cathodes	132 ± 17 mW m ⁻²	11.1 ± 1.4% vs. 7.3 ± 0.5%	182	2020	[87]
Sandy soil	Natural	TPHs	11 ± 3	Column-type	Carbon felt anode, activated carbon air-cathodes	178 mA m ⁻²	44% vs. 16%	110	2020	[94]
Sandy and clayey soil	Natural	TPHs	0.024	Double-chamber type	Carbon brush anode, titanium wire mesh	569 ± 2 mA m ⁻²	37.5% vs. 22%	137	2020	[58]
Silty, clayey, and sandy soil	Natural	TPHs	3.3	Tank-type	Both graphite pieces	63 mA m ⁻²	67% vs. 27.7%	30	2021	[88]
Saline –alkali soil	Natural	TPHs	0.15	Tank-type	Carbon cloth anode, an activated carbon air-cathode	115 ± 2 mA m ⁻²	66% vs. 8%	115	2021	[95]
Silty, clayey, and sandy soil	Natural	TPHs	4.5	Double-chamber type	Graphite anode, titanium cathode	9.5 A m ⁻²	72.5% vs. 2.7%	20	2022	[96]
Saline Soil	Natural	TPHs PAHs <i>n</i> -alkanes	28.3, Not mentioned, 0.263	U-type	Carbon mesh anode and cathode	0.85 mW m ⁻²	15% vs. 7%, 42% vs. 30%, 79% vs. 66%	25	2012	[45]
Aged sandy and clayey soil	Natural	TPHs PAHs <i>n</i> -alkanes (C ₈ –C ₃₇)	Not mentioned	Multi-anodes type	Carbon mesh anode-activated carbon cathode	43 mW m ⁻²	TPHs: 21% vs. 7%, PAHs: 44% vs. No, <i>n</i> -alkanes (C ₈ –C ₃₇): 53% vs. 37%	135	2016	[97]
Sandy and silty soil	Natural	TPHs PAHs, <i>n</i> -alkanes (C ₈ –C ₃₇)	Not mentioned, 0.01, 0.70	Column-type	Graphite rod anode-activated carbon cathode	17.3 mW m ⁻²	TPHs: 30% vs. 15%, PAHs: 42% vs. 5%, <i>n</i> -alkanes(C ₈ –C ₂₇): 55% vs. 42%	144	2016	[56]
Sediment soil	Natural	PAHs (Pyrene)	10 mg kg ⁻¹	Column-type	Both stainless steel	0.08 mW m ⁻²	92% vs. 74%	240	2012	[98]
Saline soil	Natural	PAHs	Not mentioned	Multi-anodes type	Carbon felt anode, air-cathode	37 mW m ⁻²	36% vs. 14%	180	2014	[99]
Aged sandy and clayey soil	Natural	PAHs	0.03	Multi-anode type	Carbon mesh anode-activated carbon air-cathodes	29.2 mW m ⁻²	75% vs. 16%	223	2019	[100]

(continued on next page)

Table 2 (continued)

Soil type	Contamination type (artificial/natural)	Targeted pollutants	Initial contamination level (g kg ⁻¹)	BESs type	Electrode materials	Maximum power/current output	Maximum removal efficiency (%) vs. control	Duration (days)	Year	Reference
Paddy soil	Natural	Phenol	80 mg L ⁻¹	Column type	Carbon felt anode, cloth cathode	29.5 mW m ⁻²	90% vs. 28%	10	2011	[90]
Sediment soil	Natural	Anthracene Phenanthrene Pyrene	0.10, 0.09, 0.08	Dual-phase type	Activated carbon fiber felt pieces	12.1 mW m ⁻²	Anthracene: 54.2% vs. 20.8%, Phenanthrene: 43% vs. 17% Pyrene: 27% vs. 12%	175	2017	[89]
Sediment soil	Artificial	Crude oil	5.0	Tank type	Both graphite felt	13.8 mW m ⁻²	82% vs. 20%	55	2019	[54]

oxidants (KMnO₄) into a tank soil-MFCs. It integrated bioelectrochemical remediation with chemical oxidation processes and was much more effective in removing TPHs from petroleum-contaminated soils [88].

Similar to TPHs, the removals of PAHs, phenols, *n*-alkanes, and diesel from contaminated soils were also evaluated by bioelectrochemical remediation (Table 2). Among the five reviewed studies, the removal efficiencies of PAHs were in the range of 36–92% in petroleum-contaminated soils. The removal efficiency was 44% for 16 priority PAHs and 53% for total *n*-alkanes (C₈–C₃₇) in column-type soil-MFCs with carbon fiber added into contaminated soils [56]. In U-type soil-MFCs with an initial water content of 33%, the removal efficiency was 42% for PAHs and 79% for *n*-alkanes compared to 30% (PAHs) and 66% (*n*-alkanes) in control tests [45]. Removals of PAHs from naturally contaminated soils were obtained with a maximum power density of 12.1 mW m⁻² in dual-phase type soil-MFCs, and the removal efficiencies of different PAHs were in the order of anthracene (54%) > phenanthrene (43%) > pyrene (27%) as opposed to 21% (anthracene), 17% (phenanthrene), and 12% (pyrene) in controls after 175 days [89]. A high phenol removal efficiency of 90% coupled with a simultaneous power generation of 29.45 mW m⁻² was obtained in column-type soil-MFCs over ten days compared to 28% under an open circuit [90].

Notably, a 50 L bench-scale study of bioelectrochemical remediation of soils contaminated by hydrocarbons in column-type soil-MFCs has been carried out by Lu et al. [91]. According to their research, TPH removal of 82–90% was observed in saturated soils with a simultaneous current generation (70.4 mA m⁻²). The maximum functioning radius of the soil-MFCs may reach 90–300 cm, indicating the feasibility of this technology to enhance *in situ* bioremediation.

4. Affecting factors and constraints of bioelectrokinetic and bioelectrochemical remediation

4.1. Affecting factors and constraints of bioelectrokinetic remediation

4.1.1. Configurations

The basic configuration of a lab- and bench-scale bioelectrokinetic system consists of contaminated soils in the central part and anodic and cathodic compartments on both sides. Anodes and cathodes are inserted inside electrolyte wells, and a direct current power supply is used to provide electricity to the system. The systems are usually run under an open-flow arrangement, making the injection of processing fluid (e.g., nutrients, surfactants, and buffers) into the contaminated soils possible. Typical lab-scale bioelectrokinetic systems are shown in Fig. 2. To ensure a homogeneous distribution of electric field intensity throughout the soils, the slice of graphite electrode was positioned to cover the same cross section as the fraction of the polluted soils (Fig. 2a) [32]. Due to the simple installation and easy maintenance, the single

electrode tank is widely used in petroleum-contaminated soil remediation. The symmetrical arrangement of cylindrical electrodes at four corners was also used in bioelectrokinetic remediation, called multi-electrode tank (Fig. 2b). The results showed the electric field intensity decreased from the corners (0.51 V cm⁻¹) to the center (0 V cm⁻¹) [33]. The cost of a multi-electrode tank was expected to be higher than a single one due to its multi amount of electrode materials. As shown in Fig. 2c, a peristaltic pump was used to circulate the electrolyte to maintain the normal operation of the electrode, called a recirculation tank [34]. Meanwhile, high-density polyethylene (HDPE) porous panels were used to separate electrode compartments and contaminated soils to maintain operational stability. In addition, a biological permeable reactive barriers tank (Bio-PRB tank) was designed that can work together with the electrokinetic cell by placing the PRB in the middle of contaminated soils (Fig. 2d) [35]. A bio-barrier is a fixed culture bioreactor that provides a suitable growth environment for diesel-degrading microbes, maintains neutral pH values, and improves removal efficiency. Biobarriers can be used to bioremediate several contaminants (e.g., diesel, gasoline, metals) [101]. Soil property, such as pH, is critical to the effect of bioelectrokinetic remediation. To avoid extreme pH, a polarity-reversal tank is developed by collector compartments that are added for both electrodes to collect and recirculate electrolytes, and the polarity of electric fields is reversed after a period (Fig. 2e) [36].

4.1.2. Electrode materials

The selection of electrode materials is the first step to carrying out bioelectrokinetic remediation of soils. In order to determine the appropriate electrode materials, three selection principles should be considered: (1) high efficiency, the selected electrode can ensure stable electric energy output to ensure the efficiency of remediation; (2) stability, the electrode has good corrosion resistance in the practical application environment; and (3) economy, the electrode has low cost and is suitable for large-scale applications. Currently, the commonly used electrode materials include inorganic and metal materials, such as graphite, stainless steel, aluminum, and titanium [73,77,78]. When aluminum and titanium were used as the electrode, an oxide film formed on its surface when the electrode was energized, which decreased current efficiency and affected the effect of remediation [102]. Stainless steel has good mechanical strength, making it easy to install in soils, but the relatively small specific surface area and liability to corrosion in an anoxic environment limit its application [103]. Graphite is an inert material that showed low cost, stable power generation, high-temperature resistance, and corrosion resistance; therefore, it was an ideal electrode material [104]. However, installing graphite plates into soils is difficult and costly, especially for *in situ* remediation. Compared with graphite plates, graphite rods can be more easily inserted but are relatively low surface areas. Stable, economical, and easy insertion of electrodes are desirable for bioelectrokinetic remediation. Additionally, it needs to mention that

the contaminants, including some chlorinated organics, can be reductively degraded on the cathode, such as 1-chlorobutane, polychlorinated biphenyls, and hexachlorobenzene. Meanwhile, the divalent heavy metal ions in contaminated soils will also deposit near the cathode to form hydroxide precipitates under alkaline conditions, resulting in fouling and heavy metal poisoning of microbes.

4.1.3. Electric field

The electric field (e.g., voltage gradient and distribution) is another important factor for bioelectrokinetic remediation. Under an electric field, microbes, contaminants, and nutrients can be moved in the soil. The voltage gradient has a direct effect on the removal of contaminants. One study showed the TOC removals of petroleum-contaminated soils increased with increased voltages from 66% under 0.33 V cm^{-1} to 70% under 0.67 V cm^{-1} and 73% under 1 V cm^{-1} [69]. Mena et al. demonstrated that the diesel degradation rate in contaminated soils varied from 16% to 36%, with the voltage gradient increasing from 0.5 to 1.5 V cm^{-1} after two weeks [105]. In contrast, Li et al. conducted experiments with different voltage gradients of 0, 1, 2, and 3 V cm^{-1} in PAHs-contaminated soils for 90 days, and the highest degradation efficiency of PAHs was 44% at 2 V cm^{-1} [106]. Another study on bioelectrokinetic remediation of organic contaminants from fine-grained soils also demonstrated that applied voltages above 1 V cm^{-1} had a limited influence on the removal efficiency of contaminants [107]. In addition, the energy consumption of bioelectrokinetic remediation is related to the voltage strength and remediation time. A larger voltage gradient can accelerate the migration of charged species, pollutants, and nutrients, but energy consumption will increase rapidly. Because a strong electric field has a limited effect, may kill microbes, and cause high energy consumption, the voltage gradient of 1 V cm^{-1} was considered optimal for bioelectrokinetic remediation of the soils contaminated by petroleum hydrocarbons and their derivatives. This also resulted in the remediation time of this method being longer than that of electrokinetic systems, which used a much higher voltage gradient ($20\text{--}200 \text{ V m}^{-1}$). However, the total energy consumption of bioelectrokinetic remediation ranged $4.8\text{--}13 \text{ kW h m}^{-3}$, which is far less than that of electrokinetic remediation ($29\text{--}60 \text{ kW h m}^{-3}$) [76].

The operation modes of electric fields are also important for bioelectrokinetic remediation. As shown in Fig. 5, uniform and non-uniform electric fields are the two main models to simulate the practical distribution of electric fields [108,109]. In a non-uniform electric field, a matrix of electrodes generates a two-dimensional electric field, which can have a radial or linear distribution. Previous studies have demonstrated that non-uniform electric fields have a better effect on bioelectrokinetic remediation of contaminated soils than uniform electric fields in operational stability. Moreover, the non-uniform electrokinetic remediation cost was lower than the common uniform electrokinetic treatment due to its

small amount of electrode materials [24]. The direction of two-dimensional electric fields can be fixed (unidirectional), periodically reversed (bidirectional), or changed in a rotational mode [110]. The effects of the bidirectional and rotational operation on the remediation of 2,4-DCP contaminated soils were investigated by Fan et al. [10]. The removal efficiencies of 2,4-DCP were 73% for the bidirectional mode and 35% for the rotational modes at 1 V cm^{-1} in 15 days. The reason was that the bidirectional operation could be used to overcome pH changes and water loss caused by soil heating and provide a suitable growth environment for microbes.

4.1.4. Microbial community

The microbial communities in soils play a vital role in bioelectrokinetic remediation. At present, petroleum-degrading microbes found in nature mainly include bacteria and fungi, with a total of more than 200 species. Based on research by Ambaye et al. the 16S rRNA gene amplicon and sequencing revealed that *Streptomyces*, *Nocardioideis*, *Arthrobacter*, *Pseudomonas*, and *Bacillus* were the main oxidative species as TPH degraders [111]. Some genera, such as *Acinetobacter* sp., *Bacillus* sp., etc., have been successfully used in bioelectrokinetic remediation for diesel-contaminated soils [112]. Because petroleum hydrocarbon component is complex, most pure cultures can only metabolize specific hydrocarbon molecules. Therefore, mixed microbial communities have a stronger potential to efficiently degrade petroleum contaminants. For example, Chen et al. used the microbial flora constructed by *Acinetobacter* sp., *Kocuria* sp., and *Kineococcus* sp. to remediate petroleum-contaminated soils, and the mixed bacteria have better remediation effects than a single bacterium [113].

In general, most studies indicated native microbes tended to perform better *in situ* remediation than exogenous microbes that often did not even survive in field conditions. However, some studies showed *Bacillus* and *Pseudomonas* could be selected as exogenous microbes to use petroleum hydrocarbons as carbon and energy sources in the remediation of petroleum-contaminated soils [114]. Rocha et al. assessed the efficacy of electrophoresis against the electro-osmotic flow to transport endospores of *Bacillus subtilis* LBBMA 155 and nitrogen-starved cells of *Pseudomonas* sp. LBBMA 81. Results showed that the higher transport efficiency of *B. subtilis* endospores was attributed to their higher negative charge on the cell surface [115]. Especially, *Bacillus* species could promote the growth of other bacterial species in soils and have a synergistic effect with the electric field [116].

In actual remediation, microbes tend to form a charged biocolloid wrapped with contaminants and tiny particles and will transport them across soil pores by electrophoresis [117]. The microbe's transport rate is usually in the range of $0.06\text{--}0.17 \text{ cm min}^{-1}$, depending on the voltage gradient, the charge, and the particle size of biocolloids [118]. The increase in voltage gradient will accelerate the transfer rate of microbes, promote the utilization of contaminants for their own growth and metabolism and improve the removal efficiency of contaminants, correspondingly [119]. However, an excessive voltage gradient (2.0 V cm^{-1}) obviously negatively affected petroleum-degrading microbes, resulting in irreversible penetration of microbial cell membranes [120]. Therefore, most studies chose $1.0\text{--}2.0 \text{ V cm}^{-1}$ as a suitable voltage gradient to maintain the stable diversity and structure of the microbial community [117].

4.1.5. Soil properties

Soil properties (e.g., pH, temperature, porosity, and water moisture) will also affect the removal efficiencies of bioelectrokinetic remediation. The microbial biomass, diversity, community structure, transportation, and the bioavailability of nutrients and contaminants were importantly affected by pH [121].

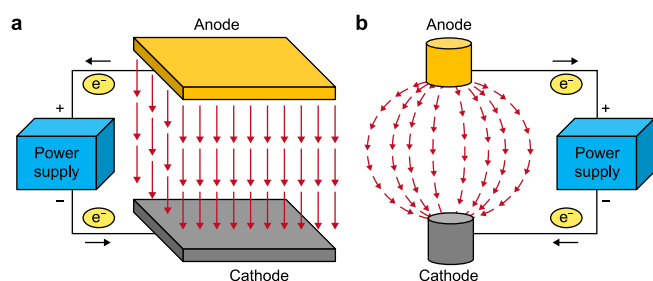


Fig. 5. Schematic of the electric intensity distribution in uniform (a) and non-uniform (b) electric fields [108,109].

A proper pH is necessary to maintain normal microbial metabolisms. Extreme pH conditions (<3 or >9) or sudden changes in pH showed some negative impacts on microbial respiration and carbon substrate utilization, then led to the inhibition of microbial growth [15,122]. Fan et al. showed that the soil pH decreased from 7.7 to 0.7 around the anode and increased to 9.5 near the cathode from an initial 7.7 after four days of operation [10]. Kim et al. demonstrated that the soil pH near the anode and cathode reached 3.5 and 10.8 after applying a voltage gradient of 1.0 V cm^{-1} for 25 days, respectively [21]. The movement of phenol and 2,4-DCP in soils can be controlled by regulating pH under the non-uniform electric field. When the soil pH increased from 7.7 to 9.3, the movement of phenol and 2,4-DCP greatly increased to two and five times faster, correspondingly [24]. There are several different methods to avoid or alleviate the influence of extreme pH value, including the addition of buffer solutions (e.g., bicarbonate and tris-acetate buffers), electrolytes circulation, and polarity reversal, but all of these methods will increase cost and energy consumption [16,68].

Electrokinetics can increase the soil temperature ($1\text{--}3 \text{ }^\circ\text{C}$) due to the electric heating caused by the ohmic drop in the soils, which have low conductivity [123]. A higher applied voltage will result in a larger increase in the soil temperature. Several studies also suggested that the treatment scales (up to 32 m^3) affect temperature changes differently. Under an applied voltage of 1 V cm^{-1} , the soil temperature at a pilot scale rose from 20 to $35 \text{ }^\circ\text{C}$, while it had no obvious change on the lab scale [124]. The change in soil temperatures may affect the metabolisms of microbes and change the soil moisture. It is controversial about the influences of soil heating on microbes. Some studies claimed that the increase in soil temperatures was negligible, while other studies revealed that soil heating decreased the removal efficiencies of contaminants due to negative effects on microbial metabolisms [35]. The increase in temperature is also known to enhance the evaporation of contaminants in bioelectrokinetic remediation. However, the evaporation of pollutants, especially for volatile organic compounds (VOCs) and semi-VOCs, may cause second contamination, which should concern bioelectrokinetic remediation [125].

Soil porosity affects the permeability, salinity, water content, temperature, and specific surface area of soils in bioelectrokinetic remediation. The soil porosity also affects the mass transportation process [126]. For example, in unsaturated soils, the oxygen mass transfer rate is higher within the soil with a larger total porosity [86]. Although sulfates and nitrates can be used as the electron acceptor under anaerobic conditions, the biodegradation rate of organic compounds is much slower than that under aerobic conditions. Generally, fine-grained soil (e.g., clayey soil) with low porosity shows lower permeability than coarse soil, such as gravel or sandy soil [127]. Most contaminants are difficult to effectively remove by conventional technologies in soils with low porosity and poor permeability, while bioelectrokinetic remediation has emerged as a potentially effective technology for low-permeable soils. The lower permeability of the soil, the better effect of electrokinetic remediation. The reason is the effect of electroosmosis obviously increases when the porosity increases. A study reported that clay soil and gravel particles were used as bio-barrier for diesel removal, and its removal efficiency by clay soil was higher than the latter at 1.0 V cm^{-1} after 14 days [128].

Water content in soil is an important parameter because biological degradation and electrokinetic operation need to work with soils near to moisture saturation point. The literature reported that the soil moisture decreased as the remediation time increased under the electric field. Meanwhile, spatial differences in moisture change during the process have been reported [69]. For example, the soil moisture was reduced by 24% around the anode while it increased by 18% around the cathode due to the influence of

electroosmosis [10]. Of course, water addition and periodic polarity reversal are two common methods to maintain the moisture content at a certain level in soils.

4.1.6. Others

The removal of petroleum contaminants from soils is influenced by the intrinsic stability of pollutants, which is relevant to their molecular weight, structural configuration, and chemical polarity, etc. [4]. The concentration of contaminants is also an affecting factor since high contaminants concentration in soils would inhibit microbial growth and lead to low removal efficiencies. In particular, the mixtures of contaminants in naturally petroleum-contaminated soils are more complex, and the removal efficiency of TPHs from natural soils on site was usually lower than that from spiked soils [129]. A previous study also demonstrated that the effects of electrokinetic remediation in natural soil are lower than with artificially petroleum-contaminated soil [118]. Conductive particle materials like carbon fiber act as electron transport promoters without inhibiting microbial metabolisms. Electron transfer via conductive particles and electrodes exhibits faster electron transfer mechanisms and could therefore enhance the bioelectrokinetic remediation of PAHs [128].

The surfactants have a hydrophobic and hydrophilic functional group that can reduce surface tension to solubilize hydrocarbon contaminants in micelle form. The addition of surfactants can also enhance the bioavailability of pollutants and improve the removal efficiency. In practical research, the adsorption capacity of PAHs depends on the type, polarity, and dose of the surfactants and the contact time between them [85]. Surfactants, such as ethylenediaminetetraacetic acid (EDTA), Tween-80, Brij-35, and Triton X-100, can be used to increase the solubility of contaminants but showed adverse effects on the soil microbial activity [130]. Biosurfactants would be a good alternative to chemical surfactants due to their non-toxicity and biodegradability in bioelectrokinetic remediation. Many hydrocarbon-utilizing microbes (e.g., *Corynebacterium*, *Rhodococcus*, *Pseudomonas*, *Serratia*, and *B. genus*) have emulsifying activities as they can synthesize different molecular mass emulsifiers [109]. Biosurfactant screening strains, such as *B. subtilis* AS2, *Bacillus licheniformis* AS3, and *Bacillus velezensis* AS4, were added into crude oil-contaminated soil was observed that the biodegradation rate of crude oil reached 92% in 48 h operation [82]. By increasing the surface area of hydrophobic water-insoluble substances and bioavailability, *Staphylococcus epidermidis* EVR4 enhanced the diesel removal efficiency from 67% to 84% [80]. Meanwhile, *Chryseobacterium* sp., *B. cereus*, *S. multivorum*, and *A. tumefaciens* produced efficient biosurfactants during the process of petroleum degradation. The removal of petroleum was positively correlated to biosurfactant production. The removal of aromatics, resins, and asphaltenes reached more than 70% within seven days [131]. Therefore, specific petroleum-degrading bacteria producing substantial biosurfactants should be domesticated in native microbial communities and applied in the bioremediation of petroleum-contaminated soils.

4.2. Affecting factors and constraints of bioelectrochemical remediation

4.2.1. Configurations

The efficiency of bioelectrochemical remediation is strongly dependent on the internal resistance of the system, which is affected by the configurations of reactors, electrode materials, and soil properties [132]. Previous research showed that a 57% decrement in internal resistance brought an increment of 100% in current density in soil-MFCs [91]. Therefore, minimizing the internal resistance is key to achieving a better effect of remediation in soil-

MFCs. By using proper reactor configurations, the internal resistance can be greatly reduced [103].

The double-chamber MFCs are a classic configuration that can simultaneously remove organic contaminants and heavy metals. Double-chamber soil-MFCs have the anode and cathode chambers separated by an ion exchange membrane (IEM) (Fig. 4e) [58]. Due to the addition of IEM, the internal resistance of the system increases. In order to reduce the internal resistance, Mao et al. removed the IEM and increased the maximum current density to 70.4 mA m^{-2} in the tank-type soil-MFCs (Fig. 4a) [53]. To simplify operation and maintenance, column-type soil-MFCs, which used an activated carbon air cathode instead of a graphite electrode, were developed (Fig. 4b) [56]. Due to the porosity of a PVC tube, the oxygen mass transfer rate was accelerated, and the efficiency of the oxygen reduction reaction at the cathode was further improved. Huang et al. [90] also constructed column-type soil-MFCs by coating a carbon cloth cathode with a mixture of graphite and MnO_2 . The removal efficiency of phenol reached 90%, approximately 23 times higher than that under open circuit control conditions. Compared to tank-type soil-MFCs, it is easily assembled and has a less adverse impact on soil structures and microbes. The U-type soil-MFCs have also been developed to improve the mass transfer and power output of the soil-MFCs (Fig. 4c) [45]. This configuration increased the compactness of the reactor by shortening the space between two electrodes and reducing intrinsic loss. U-type soil-MFCs with an internal resistance of 7.4Ω had better performance on contaminants degradation and power generation than tank-type soil-MFCs with an internal resistance of 42.6Ω [45]. However, soluble salts migrated towards the electrodes driven by the electric field, and the salt accumulation due to the evaporation of water increased with time, which led to unstable operations.

The areas of electrodes greatly affect the performance of BESs. When the anodic oxidation shows a lower rate than cathodic reduction, a fixed area of cathodes with a larger area of anodes can produce much more electricity in BESs. The current density with a larger anode area was ten times higher than that with a smaller area (800 mA m^{-2} vs. 80 mA m^{-2}) in soil-MFCs [133]. In order to maximize the utilization of the anode, multi-anode soil-MFCs were developed to remediate petroleum hydrocarbons contaminated soils (Fig. 4d) [57]. In these systems, multiple anodes were parallelly inserted in soils with different distances to the cathode, which divided the contaminated soil into several areas. It not only accelerated the degradation of hydrocarbons in sandwiched soils between two anode layers but also obviously enhanced the stability of remediation. However, from the perspective of practical application, the complex installation and the difficulty of maintenance added to the cost of this type soil-MFCs.

4.2.2. Electrode materials

Anodes on which EAB are attached are the core of BESs applied to the remediation of petroleum-contaminated soils. The choice of anodes affects not only the anodic microbial community but also the efficiency of electron transfer from microbial cells to the surface of the electrode [134]. Compared to bioelectrokinetic remediation, anodes used for bioelectrochemical remediation are more diverse. Various anodic materials (e.g., carbon, graphite, graphene) can improve the power output in MFCs. Carbon mesh, felt, and cloth has a relatively high surface area and resistance to corrosion, but installation and maintenance are relatively difficult [135]. Lu et al. [91] also used granular graphite as the anode in column-type soil-MFCs, which greatly increased the removal efficiency of TPHs, with 82–90% of initial TPHs degraded in 120 days. Biochar performed well in TPH degradation and demonstrated its suitability for bioelectrochemical remediation [136]. However, the current density of biochar anodes was 35.2 mA m^{-2} , which was lower than that

registered with graphite anode (70.4 mA m^{-2}) in the tank-type soil-MFCs for hydrocarbon degradation [100].

Ni, Cu, stainless steel, Al-alloy, and titanium mesh were also applied in some BESs. In recent years, nanocomposites have been successfully used in MFCs, based on optimal electrode properties such as excellent conductivity, thermal stability, and mechanical strength [137]. Little et al. reported a novel electrode material comprised of a conductive polymer hydrogel layer dispersed onto carbon nanotubes (CNTs), which made the electron transfer from the electrode surface more efficient [138]. Furthermore, a highly conductive matrix loaded with sufficient enzymes by immobilizing multi-copper oxidases (MCOs) onto CNTs was made with a porous 3D structure which greatly increased the power generation of BESs [139]. Additionally, the arrangement of anodes also impacts the performance of BESs. Li et al. [99] designed multi-anodes type soil-MFCs with three layers of anodes parallelly inserted in the soil. The accumulated charge reached 918 C, seven times higher than that with one layer of anodes (125 C) during the 180 days test period. In addition, two different anode arrangements, the horizontal arrangement (HA) and the vertical arrangement (VA), were discussed. HA was a better choice as the removal efficiencies of TPHs in HA reached 50% compared to 8% in VA [58].

The catalytic activity and stability of cathodes are also very important for BESs. Developing high-activity and long-lifespan cathode materials is a vital issue that needs to be resolved. Because of infinite quantity and easy acquisition, oxygen (O_2) is commonly selected as the electron acceptor at the cathode to reduce the remediation cost. Therefore, the air cathode was the most widely studied for bioelectrochemical remediation [140]. The oxygen reduction reaction (ORR) needs catalysts (e.g., platinum) to reduce the overpotential barrier of this reaction. The activated carbon exhibited similar characteristics and excellent stability compared to Pt catalysts, which have been used as the air-cathode in soil-MFCs [141]. Various non-noble materials (e.g., MnO_2 , TiO_2 , FeOOH , and Cu) were also investigated to replace Pt since they are inexpensive [142]. $\text{MnO}_2/\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{GAC}$ as the cathode achieved a maximum power density of 1176 mW m^{-3} in MFCs [143]. A study demonstrated that the surface biofouling on copper cathodes was lower than on stainless cathodes. The maximum power densities with copper cathodes ($440 \pm 38 \text{ mW m}^{-2}$) were higher than that with stainless cathodes ($370 \pm 21 \text{ mW m}^{-2}$). The reduction in biofouling was shown by less biofilm formation on the copper cathode surface compared to stainless steel cathodes due to the antimicrobial properties of copper. The surface biofouling on copper cathodes was lower than on stainless steel cathodes due to the antimicrobial properties of copper [144]. Moreover, copper electrodes are easy to install in fields. However, the dissolved copper may also be toxic to the soil bacteria. Recently, electrode modifications with conducting binding materials and immobilization of enzymes on electrode surfaces provide a new perspective for electrodes [133]. The cathode coating with poly (3-methylthiophene) significantly improved the stability and power generation capacity of BESs [145].

Cathode aging due to biofouling and salinity accumulation is a critical issue for bioelectrochemical remediation, which will reduce the performance of soil-MFCs. Many methods have been investigated to decrease cathodic biofouling, such as chemical and electromagnetic cleaning [146]. The maximum power density increased from 965 ± 45 to $1040 \pm 35 \text{ mW m}^{-2}$ after removing soluble microbial products from the cathode by chemical cleaning. At the same time, the ohmic, diffusion, and charge transfer resistances of this system decreased by 5%, 27%, and 33%, respectively [147]. Electromagnetic cleaning technique to inhibit biofouling and sediments on cathodes. After removing the foulants, the power density of column-type soil-MFCs increased by 30%–

$802 \pm 14 \text{ mW m}^{-2}$ [148]. In the future, research on a prolonged soil-MFCs performance evaluation for over one year is necessary to examine the influence of cathode aging on power output. In order to reduce the interference of these factors on cathode performance, new cathode material with resistances to biofouling and salinity needs to be developed in soil-MFCs.

4.2.3. Microbial community

Microbes in soils significantly contribute to the power generation and contaminant degradation in BESs. The typical EAB species include *Geobacter sulfurreducens*, *Shewanella putrefaciens*, *Rhodospirillum rubrum*, *Clostridium* spp., and *Bradyrhizobium* spp., etc. [149,150]. *Actinomycetes*, *Firmicutes*, and *Bacteroidetes* are primary polycyclic hydrocarbons degrading bacteria and play an important role in petroleum contaminants' biodegradation [151,152]. Proper selection of microbe consortiums is important to realize effective bioelectrochemical remediation of soils contaminated by petroleum hydrocarbons. Unlike bioelectrokinetic remediation, bioelectrochemical remediation has no negative effects on the metabolic activity of microbes, although the microbial community structure may be changed after bioelectrochemical remediation. Meanwhile, EAB (e.g., *Comamonas testosteroni*, *Pseudomonas putida*, and *Ochrobactrum anthropicum*) were obviously identified on the anode during the hydrocarbon degradation by bioelectrochemical remediation [92]. Meanwhile, the number of PAHs degrading bacteria around the anode increased about two orders of magnitude compared to the control test [45]. Recent studies showed that enriched biofilms have excellent performance for hydrocarbon contaminants degradation. For example, the diesel removal efficiency reached 80% using enriched biofilm anodes, and the electricity generation increased from 15 to 91 mW m^{-2} after 30 days [153].

Adding nutrients can increase the number, diversity, and activity of microbes in soils and, thus, the degradation rate of pollutants. Devi et al. treated oil-contaminated sludges from the site by adding degrading bacteria and nutrients. Maximum removal of TPHs was observed by integrating bio-stimulation with bioaugmentation (44%), followed by bioaugmentation alone (34%), co-substrate supplemented operations (23%), and the control (4%) [154]. With the addition of carbon sources, the performance of soil-MFCs can also be promoted. Li et al. found that the electricity generation of soil-MFCs and the removal of petroleum hydrocarbon increased by 262% and 200% after adding glucose as the substrates, respectively [97]. Carbon sources can be replaced by agricultural waste (e.g., corn stalks), animal manure, or even by sources of plant rhizosphere exudates. Meanwhile, Dunaj et al. indicated better performance of petroleum-contaminated soils over forest soils because it has more biodegradable carbon sources than forest soils [155]. Although removing efficiency can be further improved by adding nutrients and carbon sources, there has been a little report on how nitrogen conversion is accompanied by microbial degradation in electric fields. It is important to clarify the synergistic mechanism between the TPHs-degrading and nitrogen-converting bacteria under the electric field. It will help to reveal the relationships between the two concurrent degradation activities (the carbon and nitrogen conversion processes) and consequently provide a new perspective for bioelectrochemical remediation of petroleum-contaminated soils.

4.2.4. Soil properties

Soil properties (e.g., pH, temperature, conductivity, moisture contents, and salt contents) will also affect the removal efficiencies of bioelectrochemical remediation. Unlike bioelectrokinetic remediation systems, the pH and temperature changes of soil-MFCs are not so obvious [91]. Zhang et al. used a multi-anodes soil-MFC to

remove PAHs in naturally contaminated soils, and the soil pH only increased slightly after 135 days [57]. However, when electrodes serve as the electron acceptor in bioelectrochemical remediation, the pH value around the cathode would change greatly with the extension of treatment time due to the accumulation of hydroxyl (OH^-). When the contaminated soil contains divalent heavy metal ions, they will deposit near the cathode under alkaline conditions, resulting in heavy metal poisoning of microbes. Each microbe has its own suitable temperature range. If the temperature is too high or too low, it will negatively affect the performance of soil-MFCs. The study on the relationship between incubation temperatures and power outputs of column-type soil-MFCs revealed that the power output and microbial activity were proportional to the incubation temperatures. When the incubation temperature increased from 5 to 35 °C, the power output increased from 7.5 to 24 mW m^{-2} [156].

The soil conductivity determines the internal resistance of the system, and large internal resistances limit the application of remediation effectiveness for BESs. Different soil types have different conductivities. For example, sandy soils usually have high conductivity due to large porosity and strong substrate permeability, while clayey soils have the opposite characteristics due to the weak retention of water under saturated conditions [157]. Li et al. mixed the sand with contaminated soils to reduce the inherent resistance of soils and accelerate the mass transfer rate, the power output of soil-MFCs was enhanced, and the degradation efficiency of PAHs increased by up to 268% in 135 days [86]. In order to further increase soil conductivity, biocompatible conductive particles, such as graphene oxide, bio-carbon, carbon fiber, and silica colloids, have been added to contaminated soils [158]. The conductive particles were directly dispersed into the soil porosity and stretched a conductive network with electroactive microbes gradually increasing in soils. They can promote electron transfer and enhance current intensity, achieving a significant optimization in the performance of BESs. For example, the maximum current density and power density of soil-MFCs mixed with carbon fiber were 10 and 22 times as high as the control test [56]. Silica colloids were added into soils enhanced the power generation of cylindrical soil-MFCs by ten times because the stereoscopic reticular characteristics of silica enhanced ion mobility and reduced soil resistivity [159]. Meanwhile, soils were also supplemented with electrically conductive particles of biochar as a strategy to construct a conductive network with microbes in the soil matrix, thus extending the radius of influence of the hydrocarbon biodegradation of petroleum-contaminated soils [160].

Compared with wastewater, sludge, and sediment, the petroleum-contaminated soils showed salinization. The high salinity of soils has negative effects on the activity of microbes. Due to high osmotic pressures, water will be lost from cells, weakening the metabolization activity of EAB. Qin et al. showed that the removal efficiency of TPHs increased by 30% in soils when the soluble salt concentration decreased from 2.9% to 0.1% [14]. Therefore, some measures should be taken to decrease salinity before the soil-MFC treatment, such as irrigation and leaching. Most bioelectrochemical remediation of contaminated soils was conducted under water-saturated conditions ensuring, above all, the electrical conductivity of the system [92]. When soil moisture content decreases, microbial cells can't maintain the equilibration of normal osmotic pressure, which may lead to the death of microbes. Increasing soil moisture contents will also decrease the internal resistance and benefit the power generation of soil-MFCs. For example, when the water content in the soil increased from 23% to 33%, the internal resistance decreased from 10.8 to 7.4 Ω , and the TPHs degradation rates increased from 3.7% to 6.9% [45]. Additionally, low-cost and environmentally friendly hydrogels can

be used in soil-MFCs to enhance the electricity generation and the removal efficiency of TPHs. Moisture-retaining layers of polyacrylamide hydrogels were added around the anodes of soil-MFCs, maintaining the biological activities and significantly enhancing the hydrocarbon degradation in soils [94]. This provides a new method to maintain the humidity of soils and improve the performance of soil-MFCs.

4.2.5. Others

The efficiency of bioelectrochemical remediation also depends on the radius of influence (ROI) in the field. In theory, the closer the distance of contaminants to the electrode is, the better removal efficiency is. The research reported that the degradation of TPHs rose with the increase of electrode radius in column soil-MFCs. The TPHs removal was always higher in the soils near the cathode relative to the anode in closed-circuit treatments. Because the electrodes of column soil-MFCs were set in a ring structure, a high TPH degradation rate was achieved [44]. When the ROI of column-type soil-MFCs was 34 cm, the TPHs removal efficiency was 82–90% [91]. However, several factors, such as soil conductivity, water content, and electrode types, can lead to a non-linear relationship between the removal of TPHs and the influence of ROI. Therefore, more research is needed to optimize these parameters under specific conditions to achieve a high removal efficiency of TPHs and a more suitable ROI parameter. In addition, the size of the soil-MFCs needs to be considered. The efficiency of contaminants removal in the pilot-scale reactor as well as the energy output, were higher than that in the lab scale. For example, compared to a lab scale of 1.5 L, the 50 L pilot-scale soil-MFC produced ten times more power than the former (8.8 mW m^{-2} vs. 0.85 mW m^{-2}). The removal efficiency of TPHs was 66–79% in the 50 L reactor, which is four times higher than the 1.5 L reactor (15%) after 25 days [91]. Given the low power output of soil-MFCs, more attention should be paid to removing petroleum contaminants rather than power generation. Previous research indicated that the low efficiency of soil-MFCs did not affect its promotion of biodegradation. It suggested that the power efficiency loss may be limited to electron transfer to the anode rather than reduced electron release from the organic compounds [50]. However, determining the accurate conversion rate of carbon sources and electron fluxes is currently experimentally challenging, which is the key to theoretical calculations of soil-MFCs.

5. Future perspectives

Although there are several constraints of bioelectrokinetic and bioelectrochemical remediation, all literature showed optimistic attitudes toward their application of them. In view of these constraints, suitable voltage gradient (1 V cm^{-1}) and polarity reversal are usually used to maintain the activity of petroleum-degrading bacteria in bioelectrokinetic remediation. New nano-electrode materials and high-conductivity mediums can be used to reduce the internal resistance of the system and improve the effect of remediation in the bioelectrochemical remediation. Bioelectrokinetic remediation holds a high potential for treating soils with low permeability and low porosity, which are difficult to treat by conventional technologies. It also has an outstanding advantage that can be used for direct *in situ* remediation without the need to move soils, which can significantly save the cost of treatment, especially for remediation projects with large contaminated areas. Bioelectrochemical remediation is suitable for sandy soils, especially for petroleum-contaminated soils with high conductivities and permeability. Because of its configuration flexibility, bioelectrochemical remediation systems are easily integrated into existing small-scale contaminated areas for contaminated soil

treatment. Compared to bioelectrokinetic remediation, it can sustainably convert biochemical energy into electrical energy and reduce energy input. In addition, the less impact on soil properties makes it more attractive for practical applications in the future. The advantages and disadvantages of bioelectrokinetic and bioelectrochemical remediation of petroleum-contaminated soil are summarized in Table 3.

Although there is a great potential for bioelectrokinetic and bioelectrochemical remediation to treat contaminated soils, there is still a long way to realize their practical applications. From our perspectives, more attention should be given to the following four aspects to better understand these two technologies and make them applicable in practice: (1) *in situ* remediation, (2) degradation mechanisms of contaminants, (3) microbial activity, and (4) coupled with other processes.

- (1) *In situ* remediation. Although many studies have demonstrated the effectiveness of bioelectrokinetic and bioelectrochemical remediation for petroleum-contaminated soil treatment, most were conducted on a lab scale with spiked contaminants. The soil properties and the pollutants in real petroleum-contaminated soils were much more complex than in experimental studies, concluding the distribution of contaminants is uneven, and the controllability is poor. It is also accompanied by a great quantity of toxic heavy metals in microbes. According to previous studies, there are many factors to be considered in scaling up the process from lab-scale to pilot-scale: configuration of the system, properties of contaminated soil, area and volume of the site, arrangement of electrodes, and electrolyte characteristics [117]. It remains unknown how to determine these practical engineering parameters to be efficient and cost-effective. For example, soil-MFCs need to be scaled up at least in cubic meter scale for *in situ* application. The method that achieves this goal is to enlarge the size of soil-MFCs, such as increasing the total reactor volume or stacking multiple reactors, but there is no doubt that they will change the internal resistance and interactions in the system. Additionally, compared to chemical remediation technologies of petroleum-contaminated soils, such as solution leaching and extraction, photocatalytic oxidation, and chemical oxidation, the bioelectrokinetic and bioelectrochemical remediation rates are relatively slow [161]. These factors have limited the scale-up of the two technologies. Therefore, maintaining high efficiency and obtaining cost-effective evaluations is a big challenge when bioelectrokinetic and bioelectrochemical systems are scaled up for *in situ* remediation of petroleum-contaminated soils with a long-term operation. In the future, collaborating interdisciplinary areas, such as electrochemistry, soil biochemistry, organic chemistry, and material engineering, coupled with innovative engineering designs, will help to better solve the problems of the two technologies from laboratory to large scales and bring hope for practical *in situ* applications.
- (2) Degradation mechanisms of contaminants. In most previous studies, researchers evaluated the efficacy based on the removal efficiency of TPHs or specific contaminants, without considering the intermediates and end products. For those studies using naturally hydrocarbon-contaminated soils, the characterization of contaminants was also limited to measuring the TPHs and PAHs by gas chromatography with mass spectroscopy (GC-MS), lacking detailed illustration of degradation mechanisms and intermediates/products identification [92]. In fact, some intermediate compounds and accessory products of refractory and persistent organic

Table 3
Comparison of bioelectrokinetic and bioelectrochemical remediation of petroleum-contaminated soil.

Contrastive contents	Bioelectrokinetic remediation	Bioelectrochemical remediation
Suitable soils	Low permeability, low porosity, and high concentration of contaminants	High conductivities and permeability
Working mode	Applied electric field	Bioelectric field
Removal efficiencies	20–99%	11–92%
Remediation time	48 h–98 d	30–180 d
Energy consumption	4.8–13 kW h m ⁻³	Negligible
Effects to soils	Most temperature ↑, pH (anode: ↓, cathode: ↑), and moisture (anode: ↓, cathode: ↑)	Conductivity ↓
Scale of remediation	A large-scale <i>in situ</i> remediation	Laboratory (small) scale
Constraints	Voltage gradient and microbial activity	Cathode aging and internal resistance of the system

Note: In the table, the removal efficiencies, remediation time, and energy consumption ranges reported here mostly refer to lab scale with artificially contaminated soil and could not reflect outcomes of full-scale *in situ* treatments that nowadays are still lacking.

pollutants from incomplete degradation may be harmful. For example, PAHs may be transformed into oxygenated polycyclic aromatic compounds (PACs), potentially toxic, mutagenic, and carcinogenic. If the process of halogenated hydrocarbon degradation is not complete, toxic substances may appear, such as dichlorotoluene and benzyl chloride, etc. [134]. Most PAHs derivatives are known to have polar functional groups, enhancing their aqueous solubility and making them more difficultly available for degradation. Therefore, it is very important to understand the composition changes during contaminant degradation. This may require us to use more advanced test technologies, such as coupled Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS) with electrospray ionization (ESI) and two-dimensional gas chromatography (GC × GC) [58]. In addition, the application of isotope labeling may also help to study the intermediates and final products of biodegradation, which would deepen our understanding of the involved mechanisms in bioelectrokinetic and bioelectrochemical remediation.

- (3) Microbial activity. The removal efficiencies of bioelectrokinetic and bioelectrochemical remediation largely depend on the activities of microbes in petroleum-contaminated soils. Due to producing different alkane hydroxylases and substrate preferences, bacteria have distinct degradation efficiencies of alkanes with different chain lengths [162]. More highly efficient petroleum-degrading bacteria will help to increase the rate and the extent of biodegradation in both bioelectrokinetic and bioelectrochemical remediation. Therefore, it is necessary to culture efficient composite bacteria exhibiting broad environmental tolerance in petroleum-contaminated soils. Researchers mixed crude oil degrading bacteria *Dietzia* sp. CN-3 and biosurfactant-producing bacteria *Acinetobacter* sp. HC8–3S to enhance TPH degradation [163]. The highly efficient EAB needs to be screened to further improve the performance in soil-MFCs. The synergistic effect of electricity-producing and petroleum-degrading bacteria can greatly promote bioelectrochemical remediation of petroleum-contaminated soils. The hyper thermophilic exoelectrogens have the potential to broaden the application of bioelectrochemical technologies under extreme environmental conditions [164]. Strains for de-chlorination have been isolated from the petroleum-contaminated matrix, which can use solid electrodes serving as the electron donor for the de-chlorination reaction, making it possible to remove halogenated hydrocarbons by bioelectrochemical remediation [165]. Additionally, how soil types and properties affect the development of microbial communities with different functions is still largely unexplored. A better understanding of the interactions between microbes, soils, and contaminants is

extremely important to establish stably functional microbial communities.

- (4) Couple with other processes. Coupling bioelectrokinetic and bioelectrochemical remediation with other processes (e.g., solar radiation and plants) could further improve the performance of these two systems. Solar is a renewable energy source without adversely impacting the environment and microbes. Using a solar system as the power supply can significantly reduce energy consumption compared with traditional direct current power supply. It provides a novel and economical power option for electrokinetics *in-situ* remediation, especially in remote areas. For example, solar photovoltaic panels generate sustainable electricity for the process of bioelectrokinetic remediation [166]. Recently, an innovative solar energy and plant-microbial electrochemical system were developed called solar-plant MFCs [167]. Plants in this system can utilize CO₂ via photosynthesis under sunlight and release O₂ that the cathode can use. The released organic secretions can also be used as substrates by anodic microbes to maintain the metabolism and generate electricity. The hydrophobic contaminants can be adsorbed in the rhizosphere and further removed by biodegradation in the rhizosphere. In fact, rhizoremediation was also a green technology for the remediation of petroleum-contaminated soils [168]. Besides application in removing contaminants from petroleum-contaminated soils, there are some new potential uses of solar-plant MFCs, including removing greenhouse gas (CO₂), micropollutants, heavy metals, and the recovery of resources.

6. Summary and concluding remarks

Both bioelectrokinetic and bioelectrochemical remediation are promising technologies for petroleum-contaminated soils. Bioelectrokinetic remediation has great application potential in soils with low permeability, where conventional bio-remediation technologies are difficult to be implemented, although it has several side effects, such as extreme pH and increasing temperatures. Contrarily, bioelectrochemical remediation is suitable for sandy soils with high conductivities and permeability showing fewer side effects on microbes and soil properties. However, the treating time is usually longer than that of bioelectrokinetic remediation limited by the microbial electron transfer process. In order to further optimizing the bioelectrokinetic and bioelectrochemical remediation technologies requires interdisciplinary research on biology, electrochemistry, and environmental engineering. To scale up these two technologies, comprehensive studies should be taken on specific petroleum-contaminated soils to further optimize the process parameters and apparatus. Site-specific design based on the characteristics of the petroleum-contaminated soils is needed before

remediation, more highly efficient petroleum degrading bacteria and EAB need to be screened out, and more than one technology can be incorporated to meet multi-needs of practical remediation. Meanwhile, overall contaminant removal efficiencies, mass balances, and end products should be considered in remediation. In a word, the important selection criteria for the remediation of petroleum-contaminated soils should be more adaptive, sustainable, eco-friendly, and cost-effective.

CRedit authorship contribution statement

Jun Lan: Investigation, Methodology, Writing - Original draft preparation. **Fang Wen:** Investigation, Writing - Original draft preparation. **Yongxiang Ren:** Conceptualization, Writing - Reviewing. **Guangli Liu:** Conceptualization, Writing- Reviewing. **Yi Jiang:** Writing - Reviewing and Editing. **Zimeng Wang:** Writing - Reviewing and Editing. **Xiuping Zhu:** Supervision, Writing - Original draft preparation, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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