Chapter 11
Bioelectroremediation of Sediments

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11.1 Sediment Bioremediation and Sediment Bioelectrochemical Systems (SBESs)

Contamination of the aquatic environment has become a worldwide problem, especially for the developing countries due to the fast urbanization process and unsustainable industry development. Water contamination causes many risks for human health, ecological balance, and society sustainability. Therefore, remediation of the contaminated aquatic environment has been paid unprecedented attention in the last decade.

Sediment was considered to be the most important and challenging component in aquatic environment remediation because plenty of contaminants from the water, land surface, and atmosphere eventually accumulate in aquatic sediments via various atmospheric or geochemical processes (e.g., surface runoff, adsorption, and precipitation) [1, 2]. Moreover, sediment accumulates most of the refractory contaminants such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), brominated flame retardants (BFRs), and heavy metals [3]. After being accumulated in sediment, those contaminants are then continually and longtermly released to the water body. Therefore, sediment is not only a sink but also a source of the contaminants in aquatic environment [1, 3].

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Apart from the contaminants, sediments usually contain high concentration of organics and biomass generated from hydrobiological or microbial metabolisms [4, 5]. It has been reported that marine sediments can accumulate 2.52–28.8 mg C m^{-2} of organic carbon every day, and that for lake sediments could be about 21.6 mg m^{-2} day^{-1} [6]. The typical energy density of such sediments is 6.1×10^4 J/L (based on a complete oxidation of 2.0% organic carbon content) [5]. Therefore, sediment is also considered as a huge energy reserve if the chemical energy stored in sediments could be extracted.

Many physicochemical methods such as dredging, capping, aeration, or electrochemical degradation have been practically used in sediment remediation [1, 6]. However, those methods are not suitable for wide and in situ applications because of their high energy consumption, cost inefficiency, and secondary contamination [1, 6]. Bioremediation refers to technologies that stimulate environmental cleanup by regulating or enhancing the contaminant degradation by microbes, plant, or protozoon [2]. In sediments, microbial metabolism is the key driving force in contaminant degradation. However, biodegradation efficiency in sediments is usually lower than that in aerobic and aquatic environments. One of the most important reasons is the low availability of electron acceptors in sediments [1, 3, 7]. Replenishing electron acceptors (e.g., nitrate, oxygen, Fe oxides) in sediment bioremediation has been demonstrated to be an effective method for the bioremediation of various contaminated sediments [8, 9]. In the past two decades, electrodes in bioelectrochemical systems (BESs) have been intensively used as artificial electron acceptors to stimulate biodegradation [2, 4, 10].

BESs deployed in sediments were termed as sediment BESs (SBESs). Sediment microbial fuel cells (SMFCs) are the mostly used SBESs that can simultaneously stimulate sediment remediation and harvest bioelectric energy from sediments. In 2001 and 2002, Tender and his research group reported the first SMFCs deployed in benthic sediments in situ and ex situ [4, 10]. To date, over 100 researches on SBESs have been published, and about 1/3 focused on sediment remediation. Figure 11.1 showed a brief profile of those publications. It can be seen that SBESs have been paid increasing interests in the past decade. Both the power output and system volume increased in recent years.

SBESs have been operated in sediments from various environments including rivers, lakes, marines, and salt marshes, and various contaminants have been tested in SBESs. Almost all studies showed enhanced contaminant degradation efficiency. In addition to sediment remediation, many reports have successfully managed to use the electricity generated by SBES to power electronics (e.g., ultrasonic receiver, cell phone, environmental sensors) in laboratory or practical environments [11–14]. Several reports have shown that enlarged or field-deployed SBESs could function as self-sustainable, long-term devices for simultaneous bioremediation and power supply, especially in remote or contaminated aquatic environments [5, 15]. Therefore, SBESs hold the possibility to be the first applicable BES in the near future. On the other hand, the evaluation and optimization of SBES are more challenging compared with other aquatic BESs due to the heterogeneity and low matter diffusion efficiency in sediments, slow bacterial metabolism, as well as benthos activities and

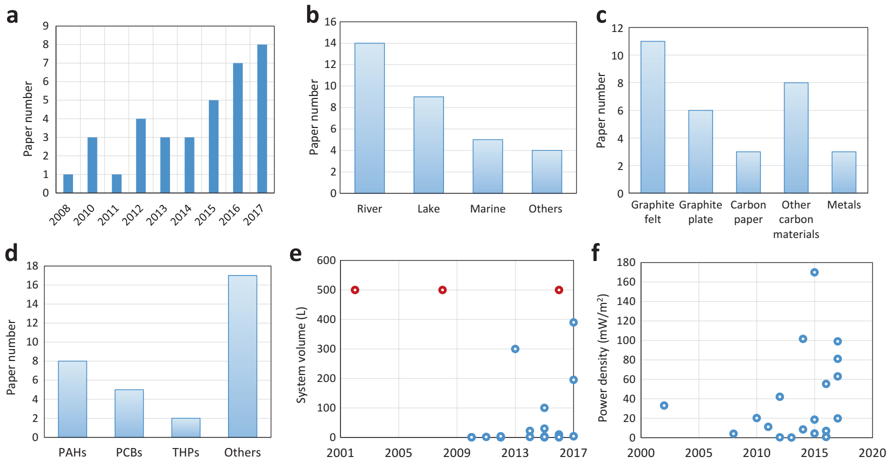

Fig. 11.1 The brief information of SBESs for sediment bioremediation. (**a**) Published papers in the last 10 years; (**b**) the sediments sources of the SBESs; (**c**) anode materials; (**d**) target contaminants of the SBES; (**e**) system volume (the red circles indicate in situ application); (**f**) power densities

some other unpredictable factors in field application. In this section, the structures, biogeochemical mechanisms, contaminant-degrading capacities, microbial ecological properties, and future challenges of SBESs will be introduced and discussed.

11.2 Structures and Principles of Different SBESs

In addition to SMFCs, several new types of SBESs including bioelectrochemical sediment caps (BSCs) [16, 17], plant-sediment microbial fuel cells (PSMFC), and bioelectro-snorkels (BSKs) have been developed recently for sediment remediation [18–21] (Fig. 11.2). Despite different structures and biogeochemical processes, microbial metabolisms and extracellular electron transfer (EET) in sediment are the core driven force in all of those SBESs.

11.2.1 Sediment Microbial Fuel Cells (SMFC)

Typically, the anode of SMFCs is embedded in anaerobic sediments, and the cathode is located in aerobic overlying water. Many microorganisms in the sediment can degrade contaminants and donate electrons to anode by their extracellular electron transfer (EET) pathway. The sediment-water interface can function as a natural layer to separate the anodic and cathodic environments. Driven by the natural

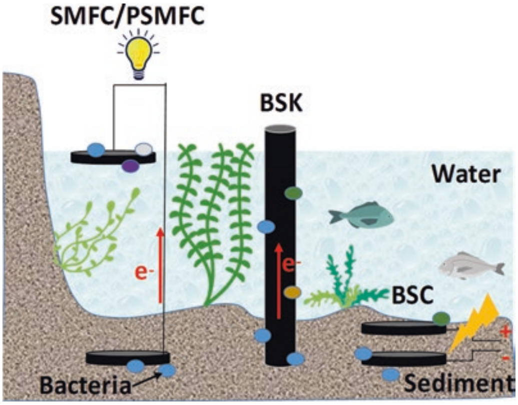

Fig. 11.2 Different types of SBESs

potential gradient between sediment and overlying water, the anodic electrons can flow via a conductive metal wire to form H_2O with protons and oxygen at the cathode. It can be seen that the outer frame and membrane, the most valuable components in a MES, are not needed in SMFCs. Therefore, SBES assembly is simpler and cheaper relative to most aquatic MESs [22].

SMFCs are expected to be long-term bioremediation or power supply devices in aquatic environments. Many factors such as microbial redox activities, sulfites, heavy metals, and high salinity would cause corrosion of the electrodes or metal wires. Therefore, SMFC materials should be corrosion resistant. To date, most studies used various carbon-based electrodes (e.g., carbon plate, felt, cloth, mesh, or brush) which have been proven corrosion resistant and suitable for long-term operation, and their power density are comparable to metal electrodes [1, 6]. Stainless steel has been used as SMFC electrodes in several reports but was still found to be corroded in long-term operation [23]. High-cost catalysts (e.g., Pt, copper, and iron) are usually unfeasible as the high concentration of sulfides or other toxic compounds accumulated on SMFC electrode surface in practical environments [1]. In addition to electrodes, the connection wires, especially the connecting knots of the wires and electrodes, should be also carefully protected. Holmes used watertight #20 AWG marine-grade wire screwed into holes in graphite electrodes, and the holes were then filled with silver epoxy and sealed with marine epoxy, by which SMFCs were operated over 7 months under both experimental and in situ marine sediments without corrosion [24]. Similar connection method was also adopted in a SMFC deployed in heavily contaminated freshwater sediments and sustained stable electricity generation for over 2 years [5]. Titanium wire was also frequently used. However, a coating layer (e.g., epoxy, polytetrafluorethylene (PTFE)) should be used to prevent corrosion of titanium wire and electron loss to the surrounding water [25, 26]. It should be noted that the concerns on corrosion of the electrode and wires are not only related to SMFCs but also to other SBESs.

11.2.2 Sediment Microbial Fuel Cell Stacks

For a single SMFC deployed in natural environments, the theoretically maximum voltage is ~1.0 V, while the practical values usually ranged from 0.3 to 0.8 V, which is much lower than the requirement of commercial monitors or electronics [27]. Like chemical fuel cells or batteries, MFCs can be operated as single unit or as parallel/serially stacked units for higher power output [28]. Stacking SMFC units in parallel can increase the current and in series can increase the voltage output. Currently, little is known on the SMFC stack. In fact, the parallel-stacked SMFC means an enlarged electrode area which will decrease the internal resistance and increase the voltage to some extent. Therefore, stacking provides a simple method to elevate the power level of SMFCs. A noteworthy drawback of serial SMFC stacks is that the electrode reversal and charge crossover will cause significant potential loss of the stack [28, 29]. On the other hand, the stacked SMFC means a larger effective area in terms of the electrode-dependent remediation [30]. The stack model can increase the electrode potential and electron transfer rate near the electrode; therefore, higher remediation efficiency can be expected. In support, a recent report showed higher substrate-consuming rate in parallel MFC stacks than that in serial MFC stacks [31]. However, before the field application of SMFCs stack, many questions such as the distance between anodes or cathodes, the area ratio of cathode to anode, and the optimal unit numbers remain to be investigated.

11.2.3 Plant-Sediment Microbial Fuel Cells (PSMFC)

In some cases, low availability and low diffusion efficiency of electron donor in sediments are important limits for the long-time performance of SMFCs. Aquatic plants were considered as a proper method to address those limits, as the plant roots located in sediments could directly generate rhizodeposits (including sugars, organic acids, polymeric carbohydrates, enzymes, and dead cell material) which subsequently serve as electron donors for the anodic bacteria [32]. Moreover, the plants could also be grown in cathodic part as the root-excreted oxygen is favorable for cathodic reaction. It has been reported that the growth of many plants such as *Glyceria maxima*, *Spartina anglica*, and *Arundinella anomala* in anodic sediments could significantly increase the power output and speed up heavy metal and organic contaminant removal in SMFCs [33–35]. The rhizodeposits account for approximately 20–40% of plant photosynthetic productivity. In terms of that, PSMFC is a technique converting solar energy into electricity. It has been estimated that net power generation of 21 GJ ha^{-1} $year^{-1}$ (67 mW/m^2) could be achieved by a PSMFC, which is comparable to the net energy yield by traditional biomass electricity production systems, such as digestion of energy crops (2.8–70 GJ ha^{-1} $year^{-1}$) and biomass combustion (27–91 GJ ha^{-1} $year^{-1}$) [6]. However, several field or laboratory experiments (rice paddy in Japan) of PSMFC showed no significant increase in power density compared to SMFC without plants, indicating the enhancement of plants on SMFC may be effected by a variety of factors such as the plant, solar radiation level, sediment composition, and temperature [32].

11.2.4 Electricity-Stimulating Systems (ESSs)

ESSs represent a group of technology that uses electric power to stimulate the bioremediation of sediments. The major difference between ESS and other SBES is that ESS consumes electricity, while the other SBESs are electricity generating or nonconsuming. One or two electrodes of ESSs were polarized at a certain potential by a potentiostat for a more specific or rapid degradation of contaminants. For example, when the electrode in the sediment was polarized at a negative potential (e.g., −0.4 V), it could serve as electron donors for microbial reduction of chlorinated organic compounds, azo dyes, Cr, and U, and when polarized at a positive potential, it could serve as electron acceptors to drive microbial oxidization of PAHs, benzene compounds, and antibiotics [16, 17]. Sun et al. recently assembled a novel ESS termed bioelectrochemical sediment caps (BSCs) by embedding two polarized electrodes (with an applied voltage of 4 V) into the cap layer [19]. Traditional sediment caps represent a thin layer of sand, activated carbon, or apatite that sequesters contaminants and further retards the movement of contamination from the sediments. In BSCs, the cathodic water electrolysis generated hydrogen which could serve as electron donor for microbial or chemical reduction of contaminants, while the anodic water electrolysis generated oxygen to serve as electron acceptor for the oxidization of contaminants [18, 19]. It can be seen that BSCs combine the advantages of both sediment caps and ESSs. Higher salinity or electron mediators could be used to further enhance the performance of BSCs. However, significant pH differences generated between the two electrode zones which might limit the long-term performance of BESCs. Despite the merits shown by lab-scale BESCs, the energy cost should be considered in long-term scaled-up application.

11.2.5 Bioelectro-snorkels (BESnK)

BESnk was firstly developed by Erable et al. as a wastewater treatment device modified from MFC [36]. Typically, a BESnk was a graphite rod (or other conductive rods) with the bottom part inserted in sediments (or activated sludge) and the top part exposed to overlying water [20, 36, 37]. BESnk could be considered as a conductive bridge linking the anaerobic sediment environment and aerobic overlying water environment, so that the electrons generated by the bottom bacteria can flow along BESnk to the overlying water. In comparison with SMFCs, BESnk cannot generate electricity. However, its simple structure, rapid electron transfer, and larger redox effects on local environment render a higher bioelectrodegradation efficiency in sediment remediation and wastewater treatment [38].

11.3 Common Biogeochemical Process in SBES

11.3.1 Anodic Biogeochemical Process

The biological and physicochemical properties of sediments from freshwater, marine, mash, or paddy vary largely, which means the electron donors, electricity-generating microbes, and electron acceptors are different among those sediments. Despite that, all kinds of sediments, even though the sterilized sediments, can generate electricity in SBES [39]. SBES anodes have been found to serves as a favorable electron acceptor for both microorganisms and reductive chemicals in the anaerobically heterogeneous sediments.

11.3.1.1 Electron Donors

Several field-deployed or scaled-up SBES have shown that SBES could generate electricity for several years [3, 5, 14]. A lifetime of 8.9 years was estimated of a 100 L SBES contained contaminated river sediments, indicating there are sufficient electron donors in sediments for SBES [5]. Generally, sulfides and organic matters are the main electron donors for SBES electricity generation [39].

Organic and inorganic sulfur compounds were not only the key factor causing odor and blackish of water body but also an important electron donors for SMES electricity generation. Sulfur-redox cycle is one of the most complex processes in sediments, especially on the anode surface (Fig. 11.3). Firstly, sulfate in the sediment and water body was reduced to sulfide by sulfate-reducing bacteria and then accumulated in sediments. When an anode was added, the sulfides can be either electrochemically (at redox potentials over −0.15 V) or microbiologically (e.g., *Thiobacillus* species) oxidized to elemental sulfur on the anode surface. Therefore, elemental sulfur accumulation was often observed on SBES anodes which may block further electron transfer from microbes or sulfides. Sulfur-oxidizing bacteria play a key role to succeed in further electron transfer. Those bacteria (e.g., *Desulfuromonas palmitatis*, *Desulfobulbus propionicus*) can oxidize sulfur to sulfate using anode as electron acceptor [39–41]. It has been estimated that sulfides oxidization could account for about 40% of the electrons in SBES electricity generation which will vary according to the sulfides amount and microbial composition in sediments [24, 39].

Organic matter oxidization contributes the most electrons of SBES electricity generation. Typically, electricity-generating bacteria can only use small molecular

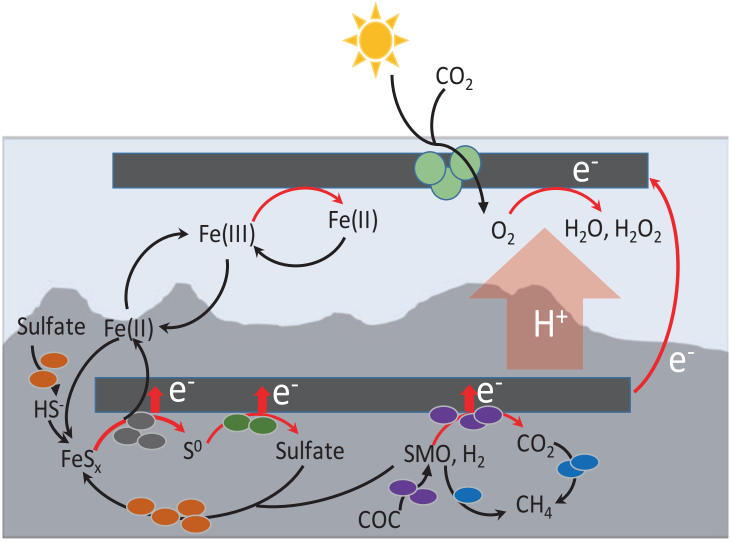

Fig. 11.3 Biogeochemical pathways of electrons at the anode and cathode in SBESs

organics (SMO) in electrode respiration. For example, *Geobacter* species use toluene, acetate, and H_2; *Shewanella* species use lactate, formate and H_2; and *Rhodoferax ferrireducens* uses glucose, sucrose fructose, and xylose [7, 42–44]. Despite those SMOs being ubiquitous in sediments, it was considered that the existing SMO (generally below 1 mM) will be rapidly depleted and most of the SMO come from the fermentation and hydrolysis of complex compounds by fermenters or other non-electricity-generating bacteria [45, 46]. The depletion of SMO by electricity-generating bacteria could alleviate the feedback inhibition of SMO to the fermentation or degradation of complex compounds. It can be seen that the main role of electricity-generating bacteria is to motivate the biodegradation of complex organic compounds (COC) rather than directly decompose them (Fig. 11.3). Organic matter are generally considered harmless for SBES, and higher concentration of organic matter can provide electrons for long-term electricity generation. Therefore, many studies added organics such as acetate, glucose, cellulose, or wheat straw to SBES [16, 47, 48]. However, Zhao et al. recently showed that higher organic contents (up to 16%) in sediments will cause unstable electricity generation, more methane emission, and higher worm activities [49].

Current studies using different kinds of sediment have demonstrated that electron donors are generally sufficient for long-term SBES operation. Moreover, additional organic electron donors may cause secondary contaminants or suppress the degradation of the local contaminants. Therefore there is no need to amending electron donors to SBES if bioremediation is the main object.

11.3.1.2 Competing Electron Acceptors

In addition to electrode, various inherent chemicals have been used as the electron acceptors by microbes in sediments. Dissolved oxygen in water column can be depleted within several micrometers below the water-sediment interface. In the deeper anaerobic environments, many other chemicals including sulfate, nitrate, humics, metal oxides, and CO_2 would compete with anode for electrons [50, 51]. The anode potential of spontaneously operated SMFC ranged from −0.2 to 0.2 V, which is relatively higher than the sulfate (SO_4^{2-}/H_2S, −0.21 V) and CO_2 (CO_2/CH_4, −0.24 V) reduction [6]. Consistently, several reports have shown that SMFC depressed the sulfate reduction and methane emission [6, 20, 25]. However, this is not always the case due to the large variation of sediment environments. For example, an in situ experiment in a specific riparian zone showed that the methane emission was depressed by SMFC deployed at upstream but slightly increased at downstream SMFC [52]. Moreover, in contrast to the assumed competing relationship between SMFC and other electron acceptors, several studies have shown that SBES performed better in the presence of some electron acceptors (e.g., Fe(III), humics) possibly because the redox intermediates of those compounds served as electron mediators or changed the local environment for electrode reduction. For example, Zhou et al. managed to improve the performance of SMFC through amending colloidal iron oxyhydroxide into freshwater sediments as the Fe(III)/Fe(II) redox species mediate electron transfer to electrode [46]. And the transformation of mineral oxides may accelerate the sediment conductivity and thus increase electricity generation [53, 54]. As another example, the redox cycle of sulfur species can release sulfate to the cathode, decreasing the cathodic pH and thus increasing the SBES performance.

11.3.2 Cathode Processes

Due to the low chemical diffusion efficiency in sediments, anode biogeochemical reactions were considered the main limit for SBES. Moreover, most recalcitrant contaminants (e.g., PAHs, PCBs, PBDEs) accumulate in sediments. Therefore, almost all SBES studies focused on the anode biogeochemical processes. However, it is possible that cathode suffers more charge transfer resistance in contaminated or nutrient-rich water bodies wherein the dissolved oxygen is low and microbe density is high. To maintain electricity generation, the electron acceptor redox potential should be higher than that of anode. Oxygen was the most favorable electron acceptor for SBES due to its high redox potential and inexhaustibility in natural water bodies. Improving the cathodic oxygen reduction could not only enhance SBES electricity generation but also the anodic biodegradation. Therefore, many synthetic cathodic catalysts and improved cathode configuration have been reported. The photosynthetic activity plays an important role in SBES cathode performance. Wang et al. improved the cathode performance by immobilizing oxygen-generating algae (*Chlorella vulgaris*) on cathode [55] (Fig. 11.3). He et al. increased the cathode oxygen concentration by developing a rotating cathode [56]. However, in addition to oxygen, PCBs, Cr(VI), Fe(III), sulfate, and nitrate could also function singly or multiply as electron acceptors at cathode [57, 58], which should be paid more attention in the future studies.

11.4 Bioelectroremediation of Sediments

About 30% of the reported SBESs researches dealt with the contaminant removal function of SMES, while the others focused on the power recovery, material or structure optimization, or microbial ecology effects of SMES. Table 11.1 summaries the brief information of the SBESs with aims to stimulate contaminant degradation. Among the diverse types of contaminants in sediments, POPs such as PAHs, PBDEs, and polychlorinated biphenyls (PCBs) were the mainly interested contaminants in the reported SMESs, followed by sulfur compounds, TOC, cellulose, and some other normal water quality indexes. POPs became the mainly targeted contaminants in SMES studies because of their wide existence, high toxicity, and low biodegradability by traditional bioremediation methods. Moreover, those contaminants generally have high hydrophobicity, and most of them are deposited and absorbed in sediments rather than water bodies.

11.4.1 SBES for PAHs Degradation

All the reported SMESs showed much higher removal efficiency on PAHs compared to the natural processes. Most of those reports used sediments from freshwater environments as inoculums. A 60-day experiment using a scaled-up SMFC showed 0.34-, 0.79-, and 0.4-fold higher removal efficiency on the benzo(a)pyrene (BaP), benzo(k)fluoranthene, and total PAHs in the river sediments. SMFCs operated for a longer term could generally further remove PAHs [59]. For example, Yan et al. reported that the BaP was decreased from 1.6 to 1.2 mg/Kg (wet sediment) after a 50-day treatment in SMFC and further to 0.8 mg/Kg at day 230, while no significant removal was observed in control [33]. It was also noted that the removal efficiency decreased over treatment time, as the removal efficiency at day 367 was comparable to that at day 230. A 970-day experiment also showed a BaP removal rate of 2 μg/Kg/day within the initial 180 days but only 0.19 μg/Kg/day in the following 800 days [3]. In addition to the decreased electricity generation, another proposed reason for the decreased PAHs degradation speed is the adsorption or transformation of PAHs or their byproducts into humic matters (humification) [3, 60]. Fertilized sediments generally showed no removal on PAHs, suggested that

Table 11.1 Contaminants degradation in different SBESs

SBES types	Sediment sources	Contaminants	Anode materials	Power densities	Scale (L)	Running time (days)	References
SMFC	River	Benzo(a)pyrene, benzo(k) fluoranthene, total PAHs	Carbon mesh	81 mW/m^2	195	60	[59]
SMES	Marine	Toluene	Graphite plate	431 mA/m^2	0.25	100	[25]
SMFC	River	TOC, ROOM, LOI	Graphite felt	18.6 mW/m^3	100	730	[5]
SMFC	River	68 organic compounds	Graphite felt	4.32 mW/m^2	30	30	[68]
SMFC	Lake	Pyrene, BaP	Graphite felt	1.1 mW/m^2	10	365	[33]
PSMFC	Lake	Pyrene, BaP	Graphite felt	1.02 mW/m^2	10	365	[33]
SMFC	Lake	BaP	Graphite felt	19.8 mW/m^2	4	970	[3]
SMFC	Lake	Phenanthrene, pyrene	Stainless steel	0.35 mW/m^2	4	240	[60]
ESS	River	PCB1, PCB61	Ti foil	49 mA/m^2	0.1	88	[17]
ESS	River	PCB61	Carbon paper	2.9 A/m^2	0.1	110	[16]
SMFC	Lake	ROOM	Graphite felt	4.08 mW/m^2	In situ	180	[15]
SMFC	Stream	TOC	Graphite felt	20.2 mA m^2	0.25	120	[75]
SMFC	Stream	CH_4, N_2O, SO_4^{2-}, Cl^-	Graphite plate	10 mA/m^2	In situ	42	[52]
SMFC	Marine	Sulfides	Graphite disks	33 mW/m^2	In situ	224	[4]
ESS	River	Naphthalene	Graphite felt	/	0.6	69	[18]
		Phenanthrene					
ESS	River	Tetrachlorobenzene	Carbon cloth	/	0.6	100	[19]
SMES	Harbor	Toluene, benzene, naphthalene	Graphite sticks	/	0.5	12	[7]
ESS	Fishing facility	CH_4	Graphite	34.9 mA/m^2	2	15	[76]
BSK	Marine	TPHs	Graphite rods	/	0.12	417	[20]
SMFC	River	TOC, PCB	Graphite brush	18.30 W/m^3	3.14	60	[63]
SMFC	Pond	COD	Graphite plates	0.1 mW/m^2	300	28	[77]

(continued)

Table 11.1 (continued)

SBES types	Sediment sources	Contaminants	Anode materials	Power densities	Scale (L)	Running time (days)	References
SMFC	Stream	LOI, DOM, cellulose	Graphite felt	0.68 mW/m^2	1.4	330	[66]
SMFC	River	TOC, DOC	Graphite fiber brush	99 mW/m^2	3.9	60	[62]
SMFC	Lake	NO_3^-, NO_2^-	Carbon paper	42 mW/m^2	0.5	38	[58]
SMFC	Lake	LOI, ROOM	Stainless steel	11.2 mW/m^2	1.4	160	[78]
SMFC	Lake	Volatile fatty acid	Graphite plates	55.2 mW/m^2	0.65	100	[49]
SMFC	Beach	TPHs	Carbon cloth	2162 mW/m^3	0.1	66	[67]
SMFC	Lake	LOI, ROOM	Graphite felt	101.5 mW/m^2	1	110	[46]
SMFC	River	Benzo(a)pyrene, benzo(k) fluoranthene, benzo(b) fluoranthene	Carbon mesh	63 mW/m^2	390	72	[69]
SMFC	Pond	COD, TN, NX^n	Graphite plate	8.47 mW/m^2	22	45	[48]
SMFC	River	BDE209	Carbon paper	280 mW/m^2	0.12	70	[64]

PAHs could only be removed by microbial degradation rather than chemical reaction in natural sediments [16]. However, the PAHs degradability of microbes in natural sediments usually decreases with the ring number in PAHs. Therefore, the concentration of PAHs with more ring number is generally higher in sediments. Recent results in our lab showed an interesting fact the SBES-enriched microbial consortia have equal or even higher degradability on PAHs with more rings. The study using a 3.5 V BSCs showed comparable efficiency with oxygen exposure for PAHs removal. Another advantage of BSCs is that the anode potential can be exchanged so that the PAHs could be degraded via either oxidative or reductive reaction [18].

Some other methods have been used to compare or integrate with SBES for a better PAHs degradation, for example, metal oxides. Fe is the most abundant metal element in subsurface environments. SMFC electrodes are generally thermodynamically more favorable than solid iron oxides in microbial respiration. Yan et al. have shown that SMFC performed higher phenanthrene and pyrene degradation efficiency than amorphous ferric hydroxide [60]. The degradation was further increased by using both SMFC and amorphous ferric hydroxide in treatment. The electron transfer rate at the microbe-electrode interface is a key factor determining the current generation and substrate degradation. Therefore, chemicals stimulating the microbe-electrode electron transfer would increase biodegradation. Zhou et al. used several kinds of iron compounds including colloidal iron oxyhydroxide, ferric oxyhydroxide, goethite, and magnetite to stimulate the electron transfer and organic degradation in the anodic sediment [46]. Among those compounds, colloidal iron oxyhydroxide showed the highest current density and substrate degradation. Zero-valent Fe (Fe^0) was also used to enhance the biodegradation and current generation in SMFCs mostly due to its highly oxidative activity [61]. In addition to role of iron species as electron donors (Fe^0) or acceptors (iron minerals), the redox cycle of Fe^{2+}/Fe^{3+} catalyzed by biological or chemical reactions is also believed to have a role in SMFC sediments [46, 62]. Rhizosphere oxygenation and root exudates have been demonstrated to play a key role in sediment phytoremediation. And some of the exudates may serve as co-substrates to stimulate PAHs degradation. It was recently shown that the removal efficiency of pyrene and BaP was enhanced by onefold by grown sweet flag (*Acorus calamus*) in a SMFC [33]. Two other reasons could also account for the enhanced degradation in PSMFCs: (1) the redox potential increased from −50 to over 100 mV when SMFC or sweet flag was added in the sediments; (2) the microbial community was significantly changed by growing the sweet flag.

11.4.2 SBES for Polyhalogenated Aromatic Compounds (PACs)

PACs are another group of recalcitrant contaminants with high toxicity and wide existence in sediments. Chun et al. applied different voltages on two electrodes vertically inserted in sediments to stimulate the PCB degradation [17]. The degradation efficiency increased with the applied voltage. Fertilized sediments showed no degradation. Therefore, the PCB was mainly degraded by microbes even though high voltages were used (4.0 V). However, H_2 or O_2 was generated at cathode or anode, respectively, when the applied voltage is bigger than 2.2 V. Therefore, both oxidative and reductive degradation was stimulated within the system. In contrast to the vertical electrodes, Sun et al. used two horizontal settled electrodes to form bioelectrochemical caps to stimulate the removal of 1,2,3,5-tetrachlorobenzene (TeCB) in sediments [19]. Considering that the microbial PCB degradation is generally initiated by reductive dechlorination, Yu et al. tried to use a negatively poised electrode (−0.3 V) as electron donor to reduce PCBs; however, no obvious PCB61 removal was observed within 1 year [16]. In contrast, a positive electrode (0.2 V) showed 58% removal efficiency within 120 days, 1.5-fold higher than that of natural degradation. They also showed that the microbial PCB dechlorination occurred primarily at para and meta positions but rarely at ortho position [16]. Those reports suggested that an anaerobic oxidative pathway possibly contributed to the PCB degradation in sediments, although it has not been evidenced. Surfactant has been used in desorption of the contaminants with high hydrophobicity in sediments. It was recently showed that the addition of surfactants (sodium dodecyl sulfate and Tween 80) could further increase the PCB degradation rate by 28.6% relative to a normal SMFC or 200% relative for natural degradation [63].

Similar to the PAHs and PCBs, PBDEs are a group of emerging contaminants with several members listed as POPs. High PBDE concentrations were commonly detected in the sediments contaminated by electronic wastes mostly in Guangdong and Zhejiang, China. Yang et al. have showed that the electrode respiration in SBES could enhance the debromination of BDE-209 by 1.5-fold [64, 65]. However the degradation products such as BDE-207 206 and BDE-183 could not be mineralized under anaerobic condition suggesting a subsequent aerobic treatment is needed.

11.4.3 Other Contaminants

Cellulose generated from the aquatic plants is an important component of the organic content in sediments. Due to the low degradable nature, cellulose is also an important reason for the contamination of water environments. Recent studies showed that adding cellulose to sediment enhanced the electricity generation of SMFC, indicating that some microbes in the SMFC could degrade cellulose to generate electricity [48]. A 330-day study showed that SMFC could enhance the sediment cellulose removal efficiency by 34.4%. A nanotube cathode could further increase the removal efficiency and electricity generation. Moreover, the cellulose activity in SMFC increased tenfold relative to that in natural sediments [66].

Toluene is also a common contaminant in sediments. Some bacteria (e.g., *Geobacter*, *Pseudomonas*) could use toluene as electron donor for electricity generation [7]. Daghio et al. found that adding toluene could significantly increase the electricity of SMFCs with a 16 mg/kg sediment/day degradation rate [25]. However, no electricity increase was observed after four batches. It was presumed that the toluene was inaccessible to the electrode-respiring bacteria when the electrode biofilm thickness increased. Sulfate was then used as electron acceptor by the thick biofilms for toluene degradation. The results also indicated that the electrode was more thermodynamic favorable than sulfate. By using [^{14}C]-toluene, Zhang et al. demonstrated that both toluene and benzene degradation could be enhanced within SMFC, and toluene could be completely oxidized to CO_2 under anaerobic sediments [7].

Two studies made efforts to stimulate the total petroleum hydrocarbons (TPHs) in sediments with SBESs. Morris and Jin reported an 11-fold (24% vs 2%) higher TPH degradation efficiency in SMFCs compared with natural sediments after 66 days [67]. Compared to SMFC, BSK was considered to be more efficient in biodegradation but no electricity generation. Viggi et al. reported a long-term (400 days) treatment of the TPH-contaminated sediments with BSKs [20]. After 200 days, 20% of the TPH was removed in BSK, while no significant removal was observed in natural or sterilized sediments. However, after 400 days, all reactors showed over 80% removal efficiency, indicating that the sediment itself had degradation capacity on TPH and BSK could stimulate the degradation process.

Despite that the degradation capacity of SBES on various organic contaminants has been demonstrated, almost all of those SBESs dealt with only one or a group of contaminants by using different reactors. There are no comparability of those reports. A key question for SBES application still remains unanswered: which kinds of contaminants are more suitable to be treated by SBES? Xia et al. analyzed 68 putative organic compounds belonged to 12 groups (alkanoates, aldehydes, ketones, alcohols, carboxylic acids and phthalate, alkenes and benzene homologs, alkanes, heterocyclic compounds, silanes, and others) in the SMFC-treated contaminated sediments [68]. The results showed a general trend that chemicals with higher polarity were more readily to be degraded in SMFC. A contrary trend (i.e., higher degradation efficiency of chemicals with lower polarity) was observed by using nitrate as artificial electron acceptor in the sediments. The results indicated that SMFCs are not proper for environments contaminated by low-polar chemicals such as petroleum pollution sites. A combination of SMFCs with soluble electron acceptors such as nitrate or sulfate would be more versatile for sediment bioremediation.

11.4.4 The Anodic Spatiotemporal Process in SBES

The spatiotemporal process is one central but less studied issue in the SBES-based bioremediation. Li et al. recently reported that the TOC decreased 17% at the anode surface, while no significant degradation occurred at 10 cm away from the anode within 18 days. At day 72, comparable TOC degradation was detected at the site 10 cm from anode [69]. Assuming that the degradation rate was linear with the distance, the effecting zone of the anode expanded at a speed of 0.25 cm/day. Several soil MFC reported the spatiotemporal property of the degradation processes. Wang et al. reported that the PAHs beyond 3 cm were not degraded after a 25-day treatment [70]. Biochar, graphite, and some conductive minerals have been used to stimulate electron transfer due to their possible role of bridging electron donors or acceptors in sediments [71–73]. A recent report showed a 70–300 cm effecting distance by using a graphite granule anode after 120 days, which largely elevated the practical bioremediation feasibility of SBES [74]. The spatiotemporal process of SBES depends on many factors including the electricity density, external resistor, and chemical and biological compositions of the sediments, but little has been known to date. Therefore, more efforts should be made on this issue.

11.4.5 Cathode-Stimulated Remediation

In contrast to the anodic processes, only several reports studied the pollutants removal by SMFC cathodes, including sulfate, nitrate, and TOC [58, 69]. Moreover, the anodic sediment environment is the main characteristic that distinguishes SMFCs from the other BESs, while the SMFC cathode processes are similar to those in the other types of MFCs, as introduced in other sections.

11.5 Microbial Mechanisms of the Bioelectroremediation in SBES

11.5.1 Microbial Communities

Electrodes are an exotic electron acceptor for the natural microbial communities in sediments. Therefore, the microbial communities will be shaped in response to SBES electrodes. Generally, the diversity decreases after SBES deployment [24, 79]. However, the specific composition enriched by different SBES varied largely from each other (Table 11.2), which could be attributed to many reasons: sediment composition, electrode potential, electrode material, or electron transfer rate. To date, most bacteria enriched by the anode belonged to *Proteobacteria* phylum; only one SMFC operated under high temperature (60 °C, marine sediment) showed the highest abundance of *Firmicutes* [80]. At the class level, *Deltaproteobacteria* showed the highest abundance in most reports, regardless of the sediment types (freshwater, marine, or lake). However, other classes belonging to *Proteobacteria* phylum such as *Alphaproteobacteria*, *Betaproteobacteria*, *Gammaproteobacteria*, and *Epsilonproteobacteria* were also enriched as the most abundant bacteria in several reports. Many operation factors such as substrate, electrode potential, or temperature could change the bacterial class composition. For example, adding Fe(III) to a SMFC changed the major microbial class from *Gamma*- to *Deltaproteobacteria* [62]. Growing plant *Acorus Calamus* in the SMFCs can change the most abundant anodic class from *Delta*- to *Betaproteobacteria*, and the most abundant family shifted from *Geobacteraceae* to *Anaerolineaceae* [33]. However, there was another report which showed that growing *Canna indica* did not change the SMFC microbial community at the class level [34], indicating the community shift could be attributed to many factors. Moreover, PCR-DGGE was a popular microbial community analyzing method before next-generation sequencing. However, two reports using PCR-DGGE method showed *Alphaproteobacteria* as the most abundant class which was rarely detected in the reports using next-generation sequencing method [21, 58]. It is possible that the two methods have different bias in microbial community analysis. The genus-level shifts in microbial community are much more susceptible to the geochemical and operational factors in SMFCs. *Geobacter* was one of the most frequently detected genus in SMFCs. Recent reports showed that

Table 11.2 Microbial communities in contaminant-degrading SBESs

SBES type	Sediment source	Phylum	Class	Genus	G/S[a]	References
SMFC	River	Proteobacteria	*Gammaproteobacteria*	*Longilinea*	+/−	[69]
SBES	Marine	*Proteobacteria*	*Deltaproteobacteria*	NA	−/−	[25]
SMFC	River	*Proteobacteria*	*Gammaproteobacteria*	*Geobacter*	+/+	[62]
			Deltaproteobacteria (Fe added)	*Pseudomonas* (Fe added)	+/+	
SMFC	Lake	*Proteobacteria*	*Alphaproteobacteria*	*Denitrifying* bacterium W73c	−/−	[58]
SMFC	Lake	*Proteobacteria*	*Deltaproteobacteria*	*Geobacter*	+/−	[33]
PSMFC			*Betaproteobacteria*	*Longilinea*	+/−	[33]
SMFC	Bog	*Proteobacteria*	*Deltaproteobacteria*	*Geobacter*	+/−	[79]
SMFC	Marine	*Firmicutes*	NA	*Thermincola*	+/−	[80]
SBES	River	*Proteobacteria*	*Epsilonproteobacteria*	*Arcobacter*	+/+	[16]
			Deltaproteobacteria	*Pseudomonas*	+/+	[16]
SMFC	Marine	*Proteobacteria*	*Deltaproteobacteria*	*Desulfuromonas*	+/−	[24]
				Desulfuromusa (AQDS added)	+/−	
SMFC	Salt marsh	*Proteobacteria*	*Deltaproteobacteria*	*Geobacter*	+/−	
SMFC	River	*Proteobacteria*	*Deltaproteobacteria*	*Geobacter*	+/−	
SMFC	Wetland	*Proteobacteria*	*Deltaproteobacteria*	*Geobacter*	+/−	[34]
PSMFC				*Desulfobulbus*	+/−	
SMFC	Marine	*Proteobacteria*	*Deltaproteobacteria*	*Geobacter*	+/−	[4]
SMFC	Lake	*Proteobacteria*	*Deltaproteobacteria*	*Desulfobulbus*	−/−	[49]
SMFC	Lake	*Proteobacteria*	*Deltaproteobacteria*	*Longilinea*	+/−	[59]
SMFC	Lake	*Proteobacteria*	*Betaproteobacteria*	*Thiobacillus*	−/−	[41]
SMFC	Fishing facility	*Proteobacteria*	NA	*Thiobacillus*	+/−	[76]
ESS			NA	*Geobacter* (−0.2 V)		
				Thiobacillus (+0.3 V)		
BSK	Marine	*Proteobacteria*	*Deltaproteobacteria*	NA	NA	[20]
BSK	Marine	*Proteobacteria*	*Alphaproteobacteria*	NA	−/−	[21]
SMFC	Lake	*Proteobacteria*	*Alphaproteobacteria*	NA	+/+	[81]
SMFC	Lake	*Proteobacteria*	*Deltaproteobacteria*	NA	+/+	[3]

NA indicates no available information

[a] Detection of *Geobacter* or *Shewanella* species: + indicates detected; − indicates not detected

Geobacter could be largely effected by the electrode redox potential and the electron donor [16]. *Geobacter* dominated on the anode polarized under −200 mV but was overcome by *Thiobacillus* on the anode of +500 mV (vs SHE) [76]. In the SBES by Yu et al., *Geobacter* increased from 1.8% in the seed sediment to 3.4% in non-acetate SBES and to 5.6% in acetate-added SBES, despite the electrode potential was polarized at +400 (vs SHE) [16]. Although *Geobacter* is a model metal-reducing organism, its abundance significantly decreased by adding Fe(III) to a SMFC, while the versatile respiring bacteria *Pseudomonas* increased to be the most abundant genus [62], possibly due to that more little organic acids were needed as electron donor to reducing the additional Fe(III). Some other microbial community shifts according to the operation factors can be seen in Table 11.2. *Geobacter* and *Shewanella* were two mostly used electrode-respiring model organisms, and they were widely observed in various subsurface environments. Therefore, their detection in SMFC was also noted in Table 11.2. It can be seen that *Geobacter* was detected in most (16/21) SMFC reports, while *Shewanella* was detected in only four reports. It is possible that *Shewanella* was more suitable to survive in redox-fluctuant environments rather than the stable and oligotrophic sediments.

In contrast to bacteria, the role of archaea in SBES was unclear to date. *Thermophilic* archaea such as *Thermoplasmatales*, *Desulfurococcales*, *Thermoproteales*, and *Thermococcales* were founded in SMFC and PSMFC sediments but were decreased compared with the original sediments, indicating those archaea did not participate in the electricity generation. Considering the competition between methane generation and electricity generation, the abundance of methanogens may have important effects on the performance of SBES. Lu et al. showed that the methanogen abundance was increased relative to the other archaea in SMFC, and most of the methanogens were hydrogenotrophic [34]. Similarly, hydrogenotrophic methanogens was the main methanogens in SMFCs with different levels of organic content [49]. In a SMFC operated for 970 days, the methanogens were significantly decreased compared with the control sediment, indicating an inhibition of electricity generation on methane generation [3].

Similar to the studies on biodegradation, most reported SBES microbial communities were grown on the anodes or sediments. Only one report studied the microbial communities on SMFCs operated in marine or salt mash sediments [24]. *Cycloclasticus* and *Methylotroph* I were dominated communities in the marine cathode, while *Rhodobacter* capable of photosynthesis dominated in the salt marsh cathode.

11.5.2 Functional Gene Communities

The functional gene or enzyme-based results are more reliable in understanding the enhancement of SMFC on the contamination biodegradation. However, only several reports showed available information on the functional gene or enzymes in SBESs [3, 64]. GeoChip is a powerful tool to test the almost all biogeochemical process-related genes in various environments. Yang et al. firstly used the GeoChip 4.0 to understand the anode-enhanced PBDE degradation. Over 9000 genes were detected, and 87.9% of them were detected in the BES but not detected in the normal anaerobic reactor. Almost all functional genes (including the genes in carbon, nitrogen, phosphorus, sulfur cycling, electron transfer, and aromatic hydrocarbon degradation) were upregulated under electricity-generating condition [64]. Yan et al. recently integrated 16S rRNA sequencing and Geochip 5.0 to analyze the bioelectrochemical BaP degradation in SMFC. A highly clustered gene network was observed in SMFC. The genes involved in electron transfer, carbon cycling, organic contamination degradation, and aromatic degradation were significantly enriched [3]. In addition to GeoChip, other methods (e.g., q-PCR and enzyme activity measurement) also evidenced that many functional genes or enzymes including dissimilatory sulfite reductase (*dsrA*), benzylsuccinate synthase (*bssA*), cellulose, and catalase upregulated on the anode of SMFCs or soil MFCs, compared to the natural sediments [25, 66, 74]. Those reports explained a confusing phenomenon in SBES researches that the degradation of almost all contaminants, regardless of oxidative or reductive, could be stimulated in SBESs, although the removal efficiencies were different. Lacking favorable electron acceptor is the main limit for the biodegradation in anaerobic sediments. The SBES provides an electron pathway from contaminants firstly to the anode and finally to the oxygen in the overlying water. The high redox potential of anodes driven by the oxygen reduction at cathode can provide much more energy for the sediment microbial community. As a result, the functional gene expression, microbial metabolism, and the cellular proliferation will be stimulated by the electrode respiration in SBESs, which can explain the stimulation of SBESs on various contaminants.

11.6 Future Development and Applications

Increasing reports have evidenced that SBESs is a promising technology to stimulate sediment bioremediation with simultaneous power recovery. How to operate SBESs in practical environments is the most urgent and challenging problem for a further development of SBESs. The first and most important step to address this problem is to deploy a SBES in a practical environment. Many unexpected problems will arise after the field deployment which may cause failure or cost much more money or labor force than that in laboratory experiment. Many of the problems can be avoided by careful considerations and designs before filed application. Firstly, the general environment of the operation sites must be evaluated before a field deployment, including the temperature range, sediment thickness and composition, water flow speed, tidal cycle, human activities, as well as the government management. Secondly, the structures and materials of the SBESs should be evaluated and optimized before application, including the SBES type, electrode material, electrode area, wire-electrode connection, system stabilization, and protection from biodisturbance. Thirdly is the operation mode. If high power output or large bioremediation zone is needed, parallel stack of multiple SMFCs will be a better choice than single or serially connected SMFCs. Moreover, a combination of different SBESs or SBESs and some other remediation methods should be considered based on their different degradation preferences.

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