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Review article

Sustainable bioelectrochemical systems for bioenergy generation via waste treatment from petroleum industries

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ABSTRACT

Petroleum industries are large water consumers and generate a lot of wastewater at various stages of industrial operations. Wastewater from the petroleum industries contain recalcitrant pollutants such as hydrocarbons that are present in high concentrations, dissolved solids and sulfur compounds that can pose potential environmental threat. Bioelectrochemical systems (BESs) are known to be sustainable processes to treat the various kinds of wastewaters such as petroleum wastewater, while simultaneously generating the bioelectricity and value-added chemicals. This review focuses on various applications of BESs such as microbial fuel cells (MFC), microbial electrolysis cells (MEC), and microbial desalination cells (MDC) using diverse types of wastewaters (petroleum sludge, produced water, formation water, and petroleum refinery wastewater) from the petroleum industries. Overall, a hybrid type BES with hydrocarbon wastewater achieved a 98% of columbic efficiency, 96.5% of chemical oxygen demand (COD), 99% of phenanthrene, 94% of pyrene and 80% of TDS removal which are superior to single and dual chamber BES performances. The review also compares the existing biological processes with BESs in terms of the treatment of hydrocarbons and process sustainability. Treatment efficiency of petroleum wastes via the BES can be further improved by integrating the biological and electrochemical processes to develop a sustainable approach to bio-refinery route.

1. Introduction

Bioelectrochemical systems (BES) are the sustainable processes that can generate value-added energy from the treatment of organic wastewaters. In BES, electroactive bacterial consortia or strains residing on the electrodes acts as the biocatalyst for the degradation of organic matters present in wastewater, which generates electron equivalents required for anodic oxidation and cathodic reduction reactions [1–3]. In some BES systems, enzymes acts as biocatalysts to produce electrochemical energy from the simple organic molecules which are distinctly called as enzymatic biofuel cells [4–6]. BES emerged into several applications such as microbial fuel cells (MFCs) for bioelectricity

Abbreviations: BES, Bioelectrochemical system; MFC, Microbial fuel cell; MES, Microbial electrolysis cell; MDC, Microbial desalination cell; BET, Bioelectrochemical treatment; MES, Microbial electrosynthesis; PW, Produced water; DRO, Diesel range organics; PRW, Petroleum refinery wastewater; FW, Formation water; TDS, Total dissolved solids; EOR, Enhanced oil recovery; TPH, Total petroleum hydrocarbons; VFAs, Volatile fatty acids; PAH, Polycyclic aromatic hydrocarbons; COD, Chemical oxygen demand; AOP, Advanced oxidation process; BOD, Biological oxygen demand; TSS, Total suspended solids; VSS, Volatile suspended solids; GG, Graphite granules; GAC, Granular activated carbon; CO_2, Carbon dioxide; H_2, Hydrogen; CH_4, Methane; OLR, Organic loading rates; PBS, Phosphate buffer solution; HRT, Hydraulic retention time; ABR, Anaerobic baffled bioreactor; AD, Anaerobic digestion; OsMFC, Osmotic microbial fuel cell; BC, Biochar; DS, Dissolved solids; SMFC, Soil/sediment MFC; LW, Labneh whey.

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(bioelectrogenesis), microbial electrolysis cells (MECs) for biohydrogen production, microbial desalination cells (MDCs) for water desalination and microbial electrosynthesis (MES) to produce the bio-chemicals [7–9]. Overall, research on BES can be a multi-disciplinary science, which includes biotechnology, bioelectrochemistry, microbiology, and environmental engineering [10–12]. BES, therefore, aims for a sustainable environmental management to provide solutions for waste management and greenhouse gases. In electrochemical reactions happening at the anode and cathode, minor electrochemical potential develops at the respective electrodes of the system that drives distinct functions, which define the application. Many types of wastewaters from industrial and domestic origins have been tested for the treatment and evaluated in types of BES [13–16]. The feasibility of utilizing the organics present in wastewater depends on the nature of the components present and their biodegradability. The complex reaction conditions prevailing in MFCs can be biological, electrochemical or a combination of which provoke higher treatment efficiency of the complex wastewater. Such combined biological and electrochemical treatment is termed bioelectrochemical treatment (BET). An author keyword search was made to visualize the spread of MFCs, MECs and MDCs using Vosviewer in Fig. 1.

Petroleum effluents contain hazardous compounds and the discharge of these compounds into the environment harmfully affects the ecology and environment. The production of petroleum based effluents is increasing with the increase in the global energy demand [17–20]. Wastewaters generated from petroleum and petrochemical industries are huge in quantities. Utilizing the organic content of such wastewaters to produce bioenergy and value-added products can unveil a new path for waste management in a sustainable approach [21–24]. BES is one such process that is known to generate value addition from the treatment. In this direction, the present review has focused on waste or wastewater treatment processes from the petroleum-based industries and their treatment via MFC, MEC, and MDC processes.

Wastewaters those generating from petroleum industries can be classified as produced water (PW), petroleum refinery wastewater (PRW), and formation water (FW). Whereas petroleum sludge is present in solid form. The major constituents of petroleum-based wastes/wastewaters are the organics (oil residues), high concentration of dissolved solids (total dissolved solids, TDS), and other compounds such as sulfur, nitrates, etc. This review discusses detailed aspects of choosing a suitable BES process along with the limitations and their advantages of the respective process. An attempt was also made to compare the BES process with the existing biological processes. The earlier reviews on treating petroleum effluent were centered around MFC and power generation [25–27] and overlooked the other BES such as MES and MDC, known for enhanced treatment efficiency. On the other hand, a comprehensive review of BES systems with various products (ex: bioelectricity and H_2) employing petroleum effluents as substrates is not available in the literature. Since the application of MES and MDC for treating petroleum effluents has dissimilar operating conditions over that of MFC, a full comprehension of the BES systems is essential. In this regard, the present review deals with the treatment of petroleum effluents in various BES systems.

2. Types of wastewaters and their characteristics

2.1. Petroleum sludge

Petroleum sludge is a residual deposit found at the bottom of the petroleum tanks and storage vessels. Typically, the composition of petroleum sludge contains organic, inorganic solids and water. Organics solids compose 90 % of the sludge material, which characterized as hydrocarbons, asphaltenes, and paraffin. Inorganic solids can be characterized as sand, iron sulfides and iron oxides [28]. Cooling the crude oil below the cloud point, mixing with incompatible materials, and

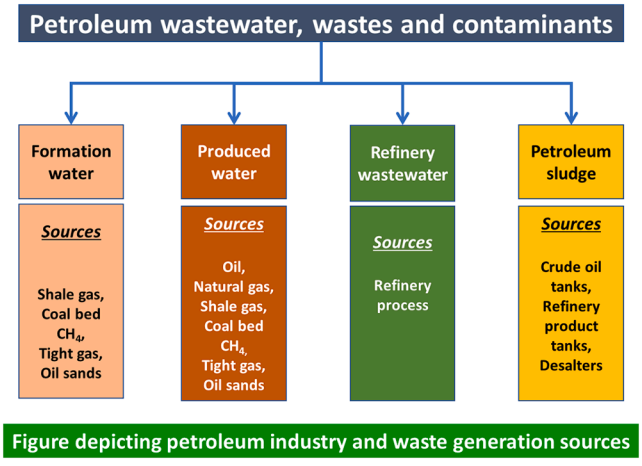

Fig. 2. Diverse types of wastewaters and waste generated from petroleum industries and associated activities.

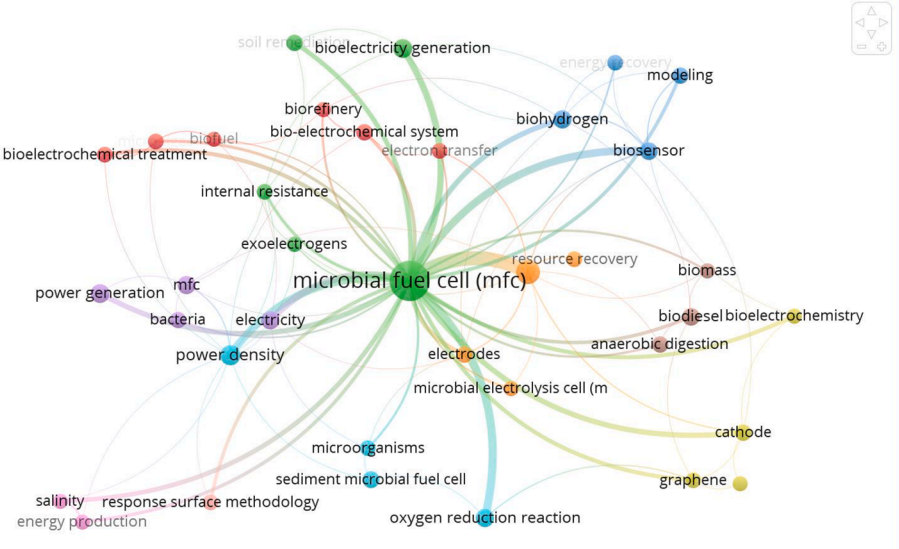

Fig. 1.V. OSviewer was used to perform the co-occurrence network analysis based on "author keywords" in publications related to microbial fuel cell, microbial electrolysis and microbial desalination cells with refinement of the keyword petroleum wastewater treatment. Keywords with>10 occurrences were selected for the mapping. Each circle represents a keyword, with the size of the circle indicating the frequency with which it appears. The intensity of the link between those terms is symbolized by the width of the lines connecting the circles.

mixing of water are the major factors for the sludge formation (Fig. 2) [29,30]. Detailed composition of petroleum sludge can be seen in Table 1.

Petroleum industries are responsible for generating massive quantities of sludge, which is the major source of environmental pollution. According to Environment Protection Act and Hazardous Wastes Handling Rules, oily sludge are categorized as hazardous wastes [31]. The sludge needs to be remediated completely before it is disposed in landfills. Low oil content sludge, which is defined as having<40 % recoverable oil, can be treated via bioremediation. Several methods are being used to separate the oil, water, and solids from the sludge material. The recovered oil is pumped back into the refinery process, while the solids and water requires treatment before the disposal [32]. To dispose the slop oil (crude oil which is emulsified with water and solids) thermal, mechanical, biological, and chemical methods are available. Each method of processing has specific advantages and disadvantages. It is common to utilize a combination of these four methods to maximize the output of usable oil from sludge of petroleum industry [33].

2.2. Produced water

Produced water (PW) is the water that lifts along with oil and gas from the subsurface layers of the earth to the surface. The ratio of produced water to the oil or gas that is generated is called as water-cut, which is found to be as high as 98 % with matured/old oilfields [34]. It was estimated that global production of PW is>77 billion bbl (oil barrel) per annum [35]. Water-cut was found to increase with an increase in the age of the well. Besides conventional crude oil and gas recovery processes, produced water is also associated with the latest fossil fuel forms such as shale gas, oil sands, and coal bed methane [26]. Based on the volume, PW is the most significant waste stream generated from the petroleum, oil, and gas industry. Also, PW is regarded as the most harmful pollutants as it contains heavy metals, organic compounds, inorganic pollutants such as sulfates [27]. In addition, PW is

Table 1
Characteristics of different types of petroleum-based wastewater and petroleum sludge.

Petroleum Refinery Wastewater (PRW)					
Ingredient	Units	Petroleum refinery wastewater 1 [62]	Petroleum refinery wastewater 2 [62]	Petroleum refinery wastewater 3 [63]	Petroleum refinery wastewater 4 [64]
COD	mg/L	596	4052	3970–4746	744–1673
BOD	mg/L	–	–	–	205–448
TDS	g/L	5.87	10.11	3.8–6.2	–
TSS	g/L	0.12	0.08	0.03–0.04	0.28–0.34
pH	–	6.5	9.5	8.3–8.7	7.5–9.41
Conductivity	ms/cm	–	–	5.2–6.8	–
Sulphates	mg/L	887	1222	14.5–16	40–50
Nitrates	mg/L	–	–	–	82–95
TPH	mg/L	–	–	–	–
Color		Slightly turbid	Dark green	Dark green	Light yellow
Grease	mg/L			–	48–97
Produced Water (PW)					
Ingredient	Units	Produced water [65]	Produced water [66]	Produced water [67]	Produced water [68]
Na^+	g/l	0.61	–	26.5	44.2 ± 2.5
K^+	g/l	0.06	–		0.968 ± 0.03
Ca_2^+	g/l	0.13	–	5.56	4.77 ± 0.1
Mg_2^+	g/l	0.12	–	0.46	0.763 ± 0.02
Cl^-	g/l	0.17	200	57.1	65.8 ± 1.6
HCO_3^-	g/l	–	3.99	–	
SO_4^-	g/l	0.05	0.01	<10	
TDS	g/l	2.33	2.67	9.82	129.3 ± 8.5
Formation Water (FW)					
Ingredient	Units	Formation water [69]	Formation water [70]	Formation water [65]	Formation water [71]
Na^+	g/l	54.4	57.3	0.56	38.1 – 45.09
K^+	g/l	0.05	5.87	0.05	0.38 – 0.90
Ca_2^+	g/l	10.6	13.6	0.12	1.46 – 2.76
Mg_2^+	g/l	1.61	2.67	0.10	0.22 – 0.384
Cl^-	g/l	107	127	0.18	63.05 – 75.37
HCO_3^-	g/l	0.176	–	–	–
SO_4^-	g/l	0.370	500	0.005	0.363 – 0.649
TDS	g/l	17.4	39.8	2.14	105.40 – 123.22
Petroleum Sludge					
Ingredient	Units	Petroleum sludge-1 [72]	Petroleum sludge-2 [73]	Petroleum sludge-3 [74]	Petroleum sludge-4 [75]
TCOD	g/kg	196	–	326.67 ± 4.62	1015–1440
SCOD	g/l	1.07	–	3.73 ± 0.92	–
TS	g/kg	2.80	–	–	1.090 – 2.439
VTS	g/kg	102	–	–	0.819 – 1.201
pH		7.40	7.14	6.73 ± 0.18	5.21 – 7.40
Density	Kg/l	1.20	–	–	1.03 – 1.12
Phosphate	mg/l	1.00	537	–	1.1 – 4.9
Nitrogen	mg/l	41.0	23.5	–	1.44 – 7.9
Sulphate	mg/l	–	–	–	12 – 19.6
TPH	%	26.2	–	–	–
TPH	g/kg	–	143.8	194.17 ± 3.80	–
TOC	g/l	0.20	–	184.89 ± 4.82	–

also a serious environmental problem due to the direct discharge of hazardous compounds with carcinogenic nature which remain soluble in water, during seepage into the sea at offshore areas [36,37]. It may cause irreparable damage to human health and the environment [38]. The oil and gas industries face two major issues with water management such as dealing with PW which is having significant amount of hydrocarbons. Mature oilfields increasingly require water-based enhanced oil recovery (EOR) methods and generate significantly more PW over the time. Petroleum effluents are hazardous and their discharge into the environment adversely affects the ecosystem. These pollutants are created mostly by rising global energy demands, which necessitates more exploration and exploitation of basic resources related to crude oil.

2.3. Formation water

Formation water exists naturally in the oil reservoir with the crude oil and it becomes produced water, including injected water and dissolved hydrocarbons, when brought to the surface (Fig. 3) [39]. The formation water characteristics mainly depends on the depositional environment, mineralogy of formation, influx/migration of fluids, and surrounding environmental temperature and pressure history [40]. Formation water contains soluble inorganic and organic compounds. Based on the salt composition, the formation water can be divided into four basic types viz., calcium chloride, magnesium chloride, sodium bicarbonate, and sodium sulfate and these can be further divided into subgroups by different combinations of dissolved salts in water. The properties of formation water mainly depends on pH value and total salinity (Table 1). The pH value between 4 and 9, is controlled by bicarbonate composition and its total salinity (~1000 to > 400,000 mg TDS/L) depends on the cationic and anionic concentration. When formation water brought to the surface it will lose the dissolved minerals and gases and alter the actual composition. Thus, it is essential to analyze under in-situ conditions [41].

Formation water composition plays a major role in souring by increasing the H_2S concentration risk in oil reserves [42]. Souring can be attributed to microbial activity, especially the sulfate-reducing bacteria, and the formation water serves as a source of nutrients in the form of low molecular weight organic acids (VFAs) to oil reserves. The souring process increases the risk to oilfields and decreases the sales value of produced hydrocarbons. It also results in higher corrosion rates in downhole equipment and surface facilities [43]. The analysis of formation water plays an essential role in predicting the source of water, possible water production rate, preparation of experimental fluid, degree of working fluid damage, dynamic modeling of reservoirs, estimation of oil and gas reserves using logging data of formation water resistivity, completion fluid for reservoir protection, compatibility with drilling and calculating completion cost [44].

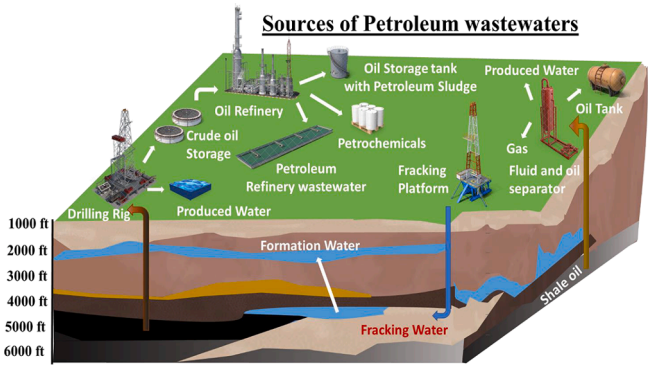

Fig. 3. Pictorial depiction of sources and petroleum-based waste and wastewaters.

2.4. Petroleum refinery wastewater

Petroleum refinery wastewater (PRW) is generated from oil refining processes that transform crude oils into many useful products such as liquid fuels, lubricants, and other petrochemical intermediates [42,45]. Typical PRW contains COD of 300–800 mg/L, BOD of 15–350 mg/L, TOC of 15–155 mg/L, total petroleum hydrocarbons of 300 mg/L, suspended solids of > 100 mg/L, and phenols of 20–200 mg/L (Table 1) [46]. The petroleum refinery units consume a large quantity of water in the process such as steam generation, distillation, cooling, desalting, cleaning, and hydro-treatment (hydro-cracking, hydrocracker flare, hydro-skimming, and hydro-skimmer flare) [47,48]. The quantity and compositions of wastewater being generated from different petroleum refineries depends on the crude oil characteristics, product generation, and plant configuration. Generally, the amount of wastewater produced from the refineries is almost 0.4–1.6 times higher than the refined petroleum products [49].

In the refinery technology, most of the water is recycled keeping in view to maintain water balance and optimizing the reuse, reduce and recycle process, thereby resulting in the generation of petrochemical wastewaters with less organic pollutants suitable for BET processes [50,51]. The generated petroleum wastewater can be categorized as (1) process PRW and (2) non-process PRW. The process PRW is produced during the refinery, while the non-process PRW is generated from the cooling towers and equipment flushing process [52]. Characteristics of non-process PRW is primarily influenced by organic and inorganic contaminants from crude oil (free, dissolved, and emulsified) and other pollutants of hydrocarbons (phenols, benzenes, toluene, benzo(a)pyrene, dibenzo(a, h) anthracene and xylenes), inorganic compounds (ammonia, nitrite, sulfide, cyanides, and heavy metals), suspended solids, and dissolved minerals [53,54]. High toxic level of contaminants need to be decreased to acceptable limits, by several water treatment techniques to minimize the destructive effects on flora, fauna, and water sources. PRW has to be sufficiently treated to maintain the quality of established regulations of the United States Environmental Protection Agency (USEPA) and World Health Organization (WHO) [55].

Different traditional techniques are employed for the treatment of PRW with physicochemical (membrane separation, adsorption, oxidation, and coagulation) and biological processes [56]. Most of the techniques are not suitable to remove high concentration of COD (4000 mg/L), complex chemical composition, poor biodegradability, and high toxicity. The conventional treatment methods such as photodegradation, photo-fenton process, biodegradation, ceramic membrane filtration, membrane bioreactors and electrochemical methods are effectively used in the treatment of PRW.

In addition to the traditional/conventional techniques, multistage treatment technologies are used to treat PRW and to generate biofuels. The first stage pre-treatment includes physicochemical and mechanical, followed by the second stage as advanced treatment of pretreated PRW. Physical treatment (sedimentation process) was used prior to biological treatment to remove the suspended particles, whereas coagulation process is used to remove turbidity in PRW. Physicochemical process of PRW resulted in reduction of total and aromatic naphthenic acid by 16 % and 24 %, respectively. In the case of biological process, higher reduction efficiency was achieved (65 % and 86 % respectively for total and aromatic naphthenic acid) [52]. The persistent pollutants in PRW generate secondary pollutants by traditional/conventional treatment process. The secondary pollutants can be treated effectively by heterogeneous photo catalysis which is an advanced oxidation process (AOP). The AOP has been studied widely to completely degrade persistent and non-biodegradable pollutants in PRW with the redox reactions initiated by light (UV/Vis/Solar) radiations. The novel nanomaterial such as ZnO, TiO_2, ZnS, and CdS generates electrons and holes with light irradiation and subsequently, redox reactions form highly reactive redox oxygen species (ROS) to degrade pollutants to CO_2 and water. Suitable media trapped the formed CO_2 to prevent pollution of environment [57].

Sonolysis (ultra-sound treatment) is another advanced AOP technique to treat rich organic pollutants in PRW by using nanomaterials [58]. In biological treatment, excessive sludge production is the main limitation that was defeated in secondary treatment by contact stabilization of activated sludge process with a maximum COD removal efficiency of 78 % and low sludge production (1.4 kg/day) [59]. The presence of low BOD/COD (<0.2) and high TDS in PRW make it unfavorable for biological treatment [60]. The hollow fiber membrane bioreactors were used to treat the organic and inorganic pollutants in PRW, and a high reduction of COD, BOD, TSS, VSS, and turbidity were achieved [61].

3. Value addition through BES treatment

3.1. Bioelectricity with MFC

Finding the optimized biological remediation process to degrade complex organics such as petroleum refinery wastes, which includes aliphatic, aromatic, asphaltenes, nitrogen, oxygen, and sulfur compounds, is one of the greatest tasks for researchers [76–78]. MFCs have gained importance for their efficient conversion of waste and wastewater to non-hazardous/low-hazardous compounds with simultaneous energy generation (Table 2). The electrogenic biocatalytic microbes of MFC use hydrocarbons as a substrate or co-substrate for the generation of electrons and protons (Fig. 4) [79–84]. Oxidation of hydrocarbons harvests electrons onto anode and the protons travels towards the cathode separator (typically Nafion), which leads to the development of potential. The potential difference developed between anode and cathode is called as voltage. External circuit connection combines electrons and protons to reduce oxygen to generate water. A wide range of wastes and wastewaters are tested in MFCs for the energy generation and potential degradation of toxics and different organic materials [15,85–88]. Due to the complexity of petroleum wastes or related products, it is presumed that MFCs can exhibit superiority in using such wastes as substrates compared to the conventional biological processes. The studies available on MFCs with treatment of petroleum industry-related wastewaters are focused on the limited aspects. However, results from the available studies are showing practical applicability with possible advancement in this direction.

In MFC, electron transfer rate through external circuit is determined by the potential difference developed between oxidative and reductive states of anode and cathode, respectively. The cathode potential of MFC governs the activity of electrons on the anode, which can be considered as one of the major constraints in MFC [104]. In this regard, uplifting the cathode performance of MFC by maintaining poised cathode potential of + 0.4 V (vs Ag/AgCl) with petroleum-produced water as an electrolyte was used as a technique. In a study showed a 4.2-fold (0.28 V control vs 1.2 V poised MFC) increase in cell potential by uplifting the cathode and noted a simultaneous increase in oxidation of petroleum compounds at the anode (Table 2). The COD, TDS and hydrocarbon removal were noted to be 91.25 %, 30 %, and 76 %, respectively [105]. The packing material used in the cathode chamber also found to influence bioelectrogenesis. Guo et al., (2016) have analyzed the influence of graphite granules (GG) and granular activated carbon (GAC) as packing materials in aerated cathode of MFC using petroleum refinery wastewater as a substrate. The superior performance of MFC in terms of power generation was noticed by GAC (330 mW/cm^3) as packing material, rather than GG (262 mW/cm^3) and control with no packing (241 mW/cm^3) [106].

Salinity or dissolved solids improve the electrolyte's conductivity, which further positively influences the bioelectricity generation potential of MFCs. Other operational factors such as redox mediators and temperature also stimulate the performance. Adelaja et al., (2015) have analyzed MFC performance using petroleum hydrocarbons in this direction. Salt concentration was varied from 0.5 to 2.5 % (w/v) to evaluate the MFC performance. An increase in cell voltage (0.2 to 1.1 mW/m^2) was noticed when the salt concentration increased from 0.5 to 1.5 %. Further increase in salinity had led to a decrease in voltage output and degradation of petroleum hydrocarbons [107]. Similarly, Minai-Tehrani et al. (2009) reported a sharp decrease in Polycyclic aromatic hydrocarbons (PAH) degradation in soil with an increase in salt concentration from 1 to 5 % [108]. This might be due to dehydrated conditions prevailing in the anodic biofilm/active electrogens at high salt concentrations [108]. The increase in temperature has also benefitted to elevate MFC performance (0.60 to 1.15 mW/m^2) in the range of 20 to 40 °C. Further increase in temperature to 50 °C, has decreased MFC performance (0.26 mW/m^2) and PAH degradation. The increase in performance of MFC with increase in temperature attributed to decrease in activation energy to drive the oxidation process, increase in kinetics as well as conductivity for the voltage amplification. The decrease in performance at 50 °C might be due to the unfavorable growth conditions or inhibition of bacteria suggesting that bacterial degradation of PAH in MFC can be limited at thermophilic conditions.

Al-Shehri et al., (2015) operated MFC for the biodegradation of benzene and naphthalene as substrates and noticed a maximum power density of 292 mW/m^2 at 40 °C. These values were higher in comparison to MFC operation at room temperature (30 °C, 156 mW/m^2) [109], suggesting that the decrease in activation energy at thermophilic temperatures enhancing power generation with increase in degradation rates. Adelaja et al.,(2014) tested the MFC performance by the variation of microbes to study the degradation of petroleum aromatic hydrocarbon i.e., phenanthrene [110]. The best performance was noticed using defined mixed culture with *Pseudomonas aeruginosa*. The maximum degradation rates, power density and COD removal using phenanthrene as a substrate were 27.30 μM/d, 1.25 mW/m^2 and 65 %, respectively [110]. In another study, Majumder et al., (2014) operated a single chamber air cathode MFC using the *Pseudomonas putida* as an electrogenic biocatalyst and refinery wastewater as a substrate [92]. This study demonstrated the treatment of refinery wastewater with simultaneous generation of electricity. The maximum current density and COD removal efficiency were 0.015 mA/cm^2 and 30 %, respectively.

Diluted petroleum sludge in water at different loading rates, was used as the substrate (anodic electrolyte) to operate MFCs for hydrocarbon degradation and bioelectricity production. Chandrasekhar and Venkata Mohan (2012) had operated a single chamber BES using petroleum sludge as a substrate by varying OLR from 3 to 30 g/L corresponding to 1.11 to 11.1 g TPH/L [60]. The increase in OLR from 3 to 30 g resulted in a decrease for the maximum power density from 20.62 to 0.12 mW/m^2 (Table 2). However, the operation of BES at high OLR resulted in achieving the higher substrate degradation. The bioaugmentation strategy (supplementation of additional bacteria) into the BES has helped to increase the overall performance.

Wang et al., (2012) operated a U-tube sediment MFC for the treatment of PAH contaminated site, where it was observed a decrease in water content from 33 to 23 % by evaporation, resulting in a decrease of charge output from 125 to 15 Coulombs. This might be due to the salt accumulation at the cathode that indirectly affects the current generation efficiency (Table 4) [111]. Likewise, Morries and Jin (2007) tested the feasibility of single-chamber MFC for the remediation of hydrocarbons in groundwater [89], with a maximum power density of 120 mW/m^2. In this study, the performance using complex substrates was compared to MFC that operated with acetate as a substrate. The decrease in cell performance with an increase in bridge distance (between anode and cathode) had led to an obvious increase in internal resistance and decrease in power generation. Similarly, Li et al., (2016) constructed a soil MFC to enhance the degradation of petroleum hydrocarbons in soil by using glucose (0.1 to 0.5 % W/V) as a co-substrate. MFC operation with a high concentration of glucose exhibited an enhanced performance (450 mV) in comparison to the operation with low concentration (400 mV) and no glucose additions (220 mV) [112]. MFC combined with membrane bioreactor (MFC-MBR) was evaluated for the treatment of PRW showed linear relationship between voltage generation and COD

Table 2
Single chamber bioelectrochemical systems (BESs) studied with hydrocarbon wastewater and wastes for the generation of energy and/or treatment.

Type of reactor	Working electrode potential	Energy generation	Type of waste treated	Type of electrode (Anode-A, Cathode-C)	Removal efficiency	Dominant bacteria	References
Single chamber MFC	N/A	120 mW/m^2	Groundwater	A: Stainless steel C: Carbon cloth	N/A	Mixed consortia	[89]
Single chamber MFC	Voltage varied from 9 − 20 V	0.05 mA/cm^2	Trichloroethene (TCE)	A,C: Stainless steel mesh	PCE removal: 23 μmol/d, Vinyl chloride removal: 72 μmol/d	*Dehalococcoides, Desulfitobacterium, Methanogenic &Homoacetogenic bacteria*	[90]
Single chamber MFC	N/A	111.76 mW/m^2	Vegetable waste	A,C: Graphite plates	69.4 % COD removal	Mixed consortia	[85]
Two single-chambered MFCs	N/A	343 mV and 53.11 mW/m^2	Real field petroleum-based oil sludge	A,C: Graphite	PHA degradation: dibenzo(A,H) anthracene and benzo (G,H,I)perylene (almost 98 %). THP (Aromatic) degradation: 75.54 %	Mixed consortia	[91]
Single chamber MFC	N/A	20.62 mW/m^2	Real-field petroleum sludge	A,C: Graphite	Higher petroleum hydrocarbons (TPH) removal: 35 %	Mixed consortia	[60]
Single chamber MFC	N/A	53.11 mW/m^2 and 343 mV	Real field petroleum sludge - oil refinery	A,C: Graphite plates	TPH removal: 35 %; Aromatic removal: 72 %; Aliphatic removal: 57 %.PHAs removal: dibenzo(A,H) anthracene (5-ring) and benzo(G,H,I)perylene (6-ring), 98 ± 1.2 %; 4-ring compounds: 70–80 % reduction	Mixed consortia	[60]
Single chamber MFC	N/A	0.005 mW/cm^2	Oil refinery wastewater	A: Carbon cloth C: Carbon cloth with PTFE coating	N/A	*Pseudomonas putida (BCRC 1059)*	[92]
Single chamber MFC	N/A	280 mW/ m^2	Refinery wastewater	A: Carbon brush C: Carbon cloth with PTFE coating	N/A	N/A	[93]
Single chamber MFC	N/A	631 mA/m^2	Synthetic wastewater (DSW)	A: Graphite C: Graphite	Color removal-63 %; Turbidity removal- 90 %; Phosphate removal-51 %; Nitrate removal-48 %; COD removal-90 %; Sulphate removal- 68 %	Mixed consortia	[94]
Single chamber MFC with enriched anode	N/A	N/A	Marine sediments	A.C: Graphite rods	TPH degradation in 1 snorkels- 12 ± 1 %; 3 snorkels-21 ± 1 %	*alphaproteobacteria, gammaproteobacteria, deltaproteobacteria*	[95]
Single chamber (EC and MFC)	N/A	90.8 mW/ m^2,115 mA/m^2 normalized to cathode surface	Diesel range hydrocarbons	A: Carbon brush anode ; C: Carbon cloth with PTFE	100 % with 1 year selected anode biofilm; 83.4 % with fresh biofilm.	*Geobacillus, Stenotrophomonas, Gordonia, Flavobacterium, Actinobacterium on anode*	[96]
Single-chamber MFC	N/A	304 mA/m^2,	Petroleum hydrocarbon contaminated soil	A: Carbon mesh C: Activated carbon	TPHs degradation- 60 ± 9 %	*δ-Proteobacteria, Flavobacteria, or Clostridia*	[97]
Single chamber (membrane-less BES)	N/A	280 mW/m^2	Synthetic wastewater (acetate)	A: Carbon brush C: Carbon cloth/ Pt	Coulombic efficiency, 60 ± 3 %	Mixed consortia	[98]
Single chamber (rectangular shaped reactor-MFC)	Applied potential: 0.4 to 1 V	278 mA/m^2, and 222 mW/ m^2	Petroleum refinery wastewater (PRW)	A: Cylindrical graphite C: Cup-shaped graphite	COD degradation rate- 0.364 kg COD/m^3-day; DROs degradation-> 90 %	Mixed consortia	[99]

(*continued on next page*)

Table 2 (*continued*)

Type of reactor	Working electrode potential	Energy generation	Type of waste treated	Type of electrode (Anode-A, Cathode-C)	Removal efficiency	Dominant bacteria	References
Single chamber (three layer soil MFC)	Applied potentials: 0.5, 1.0, 1.5 and 2.0 V	725 mW/m^2	Petroleum refinery wastewater (PRW)	A,C: Graphite	COD degradation- 69.2 %; TDS removal-53 %; TPH removal- 93 %; Oil and grease removal-76 %; Sulfate reduction-59 %	Mixed consortia	[100]
Single Chamber MEC	N/A	0.321C d^{-1} g^{-1}, 148.6 ± 0.8 mA/m^2, 5074C	Petroleum hydrocarbons (aged soil)	A: One layer carbon mesh C: Activated carbon – air	Hydrocarbon degradation: 328 %,	*Proteobacteria, Firmicutes, Bacteroidetes, Actinobacteria, Chloroflexi, Planctomycetes & Acidobacteria*	[101]
Single chamber (rectangular cuboid- soil MFC)	Applied potential: 0.6 to 1 V	0.74 A/m^2	Low-strength petrochemical wastewater	NA	COD removal-85.9 %; Coulombic efficiency-73.8 %; Methane production- 63 ± 1 mL	Mixed consortia	[102]
Single chamber (Soil-MFC	Applied potentials	286.7 mW/m^2	Petroleum refinery wastewater (PRW)	A,C: Carbon electrodes	Sulfates removal: 62.64 % and total dissolved solids removal: 12.08 %.	Mixed consortia	[103]
Single chamber AD-MEC system	Applied potentials	Single chamber: 789 mW/m^2	Petroleum refinery wastewater (PRW)	A: Graphite brush C: Platinum coated carbon cloth	Sulfates removal −79.6 %; TPH removal-47.6 %; COD removal-54.7 %	Mixed consortia	[8]

removal with R^2 of 0.9821, suggesting that MFC can be used as biosensor to control the combined system [113].

All the above-mentioned studies have proven to be efficient in degrading the petroleum wastes and their components such as alkanes, alkenes, and aromatic hydrocarbons (Table 1; Table 2). Also, several studies have focused on the degradation of complex hydrocarbons such as toluene, benzene, and other petroleum-related compounds by using poised anode potentials or by regular MFC operation with pure and mixed cultures [114,115]. Although these studies have analyzed the use of halogenated hydrocarbons in MFC, but the application of gasoline (product from crude oil) remains to be unsolved and needs to be accessed. One of the major constraints in bioelectrochemical remediation and anaerobic oxidation of hydrocarbons is the initiation of a metabolic pathway for the degradation. In terms of aerobic oxidation, it is well known for the addition of hydroxyl groups by oxygenase reaction. As for anaerobic conversion, the concrete metabolic pathways for aromatic carbon reductions remain unclear. Few studies have analyzed the individual treatment of PAH like anthracene and pyrene as a substrate at anode of MFC [116,117]. Based on gas chromatography-mass spectroscopy analysis, these studies have shown the breakage of benzene rings in anthracene and pyrene by microbes and forms phthalic acid and 2-hydroxy-phenol, respectively. These compounds (phthalic acid and 2-hydroxy-phenol) enters the tricarboxylic acid cycle and further used by microbes as energy source (Fig. 5).

3.2. Desalination with MDC

It is well known that the earth is abundant of water resources, in which 97 % is composed of seawater that cannot be used directly (Fig. 4). The increase in demand for freshwater is further increasing with an increase in global population and industrialization. In this regard, the desalination process was employed to meet freshwater demands, especially in areas limited to seawater and brackish water. Desalination with MDC is also crucial for treating industrial wastewater such as distillery wastewater, petroleum-based wastewater, and pharmaceutical wastewater with high TDS concentrations [85,118]. One of the major bottlenecks in desalination process is the high energy requirement for treatment and operation cost. Therefore, energy neutral and efficient MDC processes were developed. MDCs gained importance due to simultaneous treatment of wastewater and desalination without energy input. The MDC differs from MFC with a presence of a middle desalination chamber between anode and cathode and separated with anion and cation exchange membranes [119,120]. Due to the potential differences, the cations and anions present in the seawater present in the middle desalination chamber diffuse towards the anode and cathode chambers, resulting in low TDS. Several wastewaters were evaluated as substrate in the anode chamber of MDC to observe a power generation with simultaneous desalination. Sevda et al., (2017) studied the feasibility of MDCs for seawater treatment using petroleum refinery wastewater as a substrate, which exhibited a maximum power density of 243 mW/m^2 by using seawater with acidified catholyte, in comparison to MDC operation with phosphate buffer solution (PBS; 190 mW/m^2) as the electrolyte in the cathode chamber [121]. In another study, Ismail and Ibrahim (2015) operated the MDC with petroleum PW by coupling with forward osmosis membrane and ion exchange membrane and noticed a maximum COD removal of 92 %. The maximum power density and TDS removal were noted as 48.52 mW/m^2 and 80 % respectively [122]. These values were reported from 10 days of MDC operation in continuous mode. The COD removal and power generation were in a range of MDC operation by using simple substrates such as acetate [123]. Limited number of studies found with petroleum hydrocarbons and their related components as substrates in MDCs. Further research in this direction of MDC in utilizing petroleum hydrocarbons for sustainable desalination process needs to be encouraged. The low biodegradability of the petroleum hydrocarbons and high salt concentrations (TDS) of petroleum related wastewaters are not supportive of the required bioelectrochemical activity for efficient desalination process. The biodegradability of petroleum wastewater can be increased by adding highly biodegradable wastewaters as co-substrate. This helps in increase of overall biodegradable fractions of wastewater [2,81]. Further, chemical and electrochemical processes can be integrated to degrade the complex hydrocarbons into simple hydrocarbons. Simple hydrocarbons are more amenable to maintain bioelectrochemical activity than complex hydrocarbons. Integration of different process with distinct objectives promotes bio-refinery approach and circular bioeconomy.

3.3. Biohydrogen generation and hydrocarbon degradation using MEC

An increase in atmospheric temperature and rapid damage to ecosystems worldwide have led researchers to search for alternative

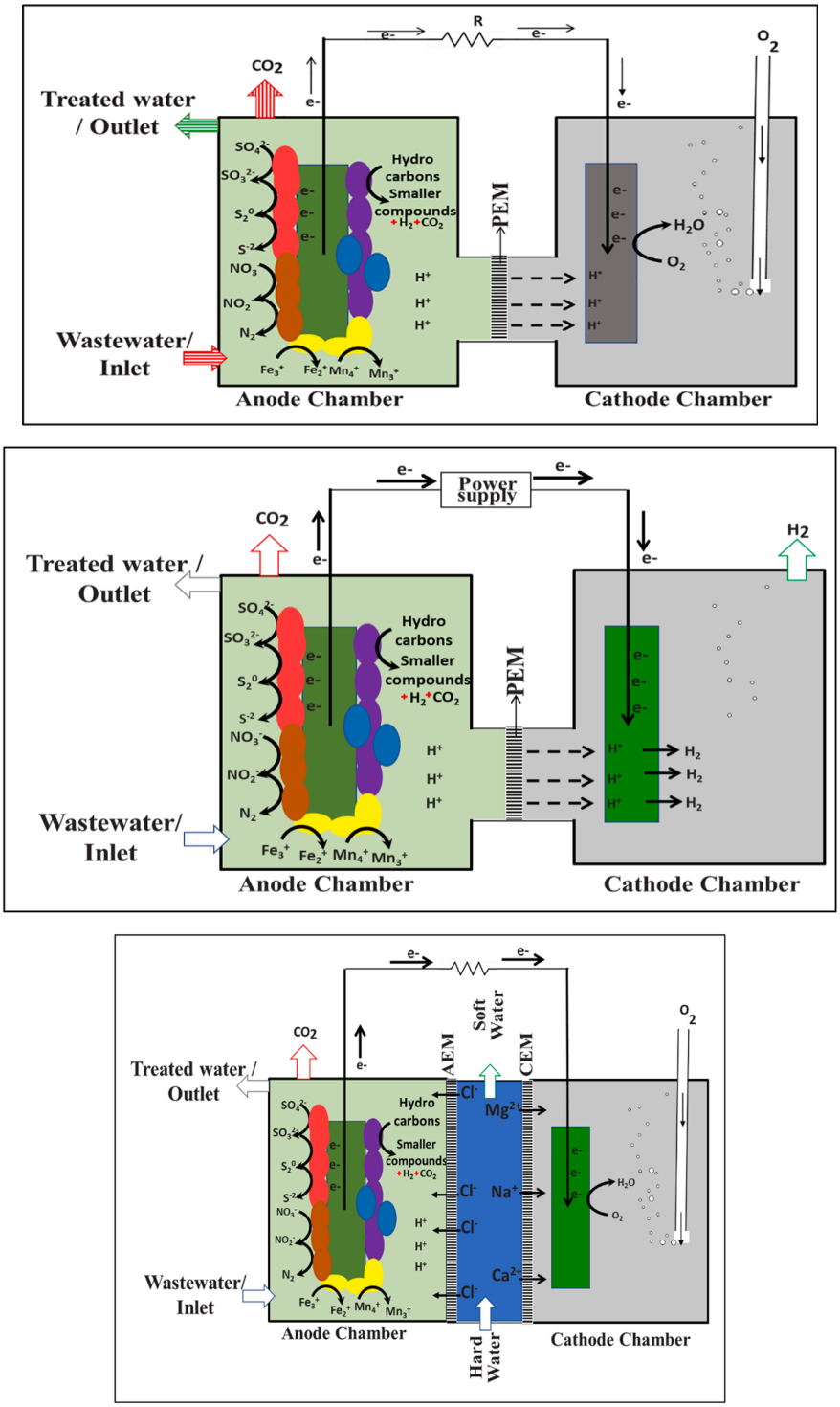

Fig. 4. Detailed mechanism of conventional (a) microbial fuel cell using organics as a substrate. Schematics of (b) microbial electrolysis cells, (c) microbial desalination cells and (d) microbial electrosynthesis that function towards the removal of pollutants present in petroleum effluents at the anode and concurrently generate bioelectricity, biohydrogen, water desalination, and pursue biochemicals production, respectively.

renewable energy sources to compensate the global energy demand. In this regard, the use of hydrogen (H_2) has attracted great attention as a potential alternative energy source since the burning of H_2 does not contribute to further production of any greenhouse gases (GHGs) [124,125]. Additionally, it is a well-known fact that among all the fuels (46.4 MJ/kg for gasoline, 55.6 MJ/kg for CH_4, 24.0 MJ/kg for ethanol), H_2 proves to be highly efficient (120 MJ/kg) in terms of energy densities [126]. In this regard, H_2 production through waste valorization has gained importance due to its clean, renewable, and sustainable approach [127–129].Yet currently, most of the H_2 (around 96 %) is generated from fossil fuel-based technologies such as steam reforming, thermochemical conversion (pyrolysis), and gasification (Table 2; Table 3) [130,131]. The generation of H_2 from carbon-neutral and biorefinery approaches to reduce environmental-related problems is highly

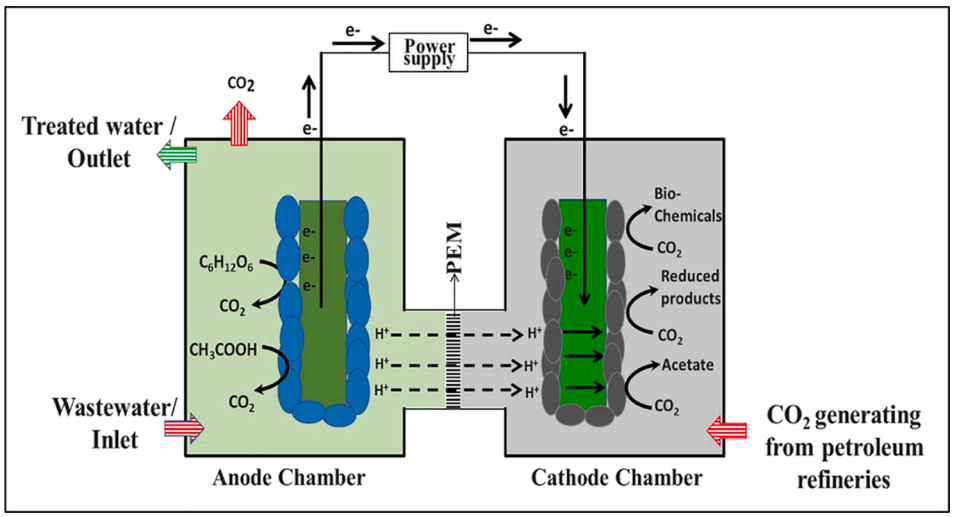

Fig. 4. (*continued*).

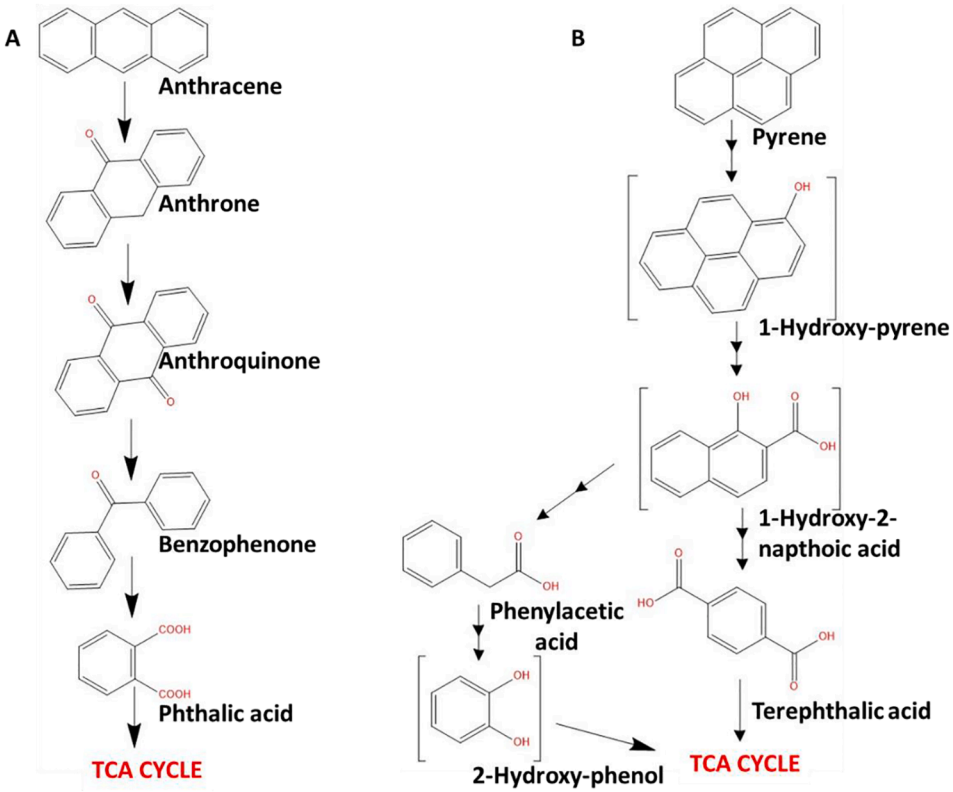

Fig. 5. Presumed degradation mechanism of anthracene (A) and pyrene (B) by active electrogens at the anode of MFC ().
Adapted from [116,117]

prioritized to decrease energy demand. Microbial electrolysis cells (MECs) have proven to be promising for H_2 generation by using organics in waste and wastewater.

Initially, MEC was discovered at Penn state university by Bruce Logan group in the year 2005 [142]. The MEC differs from MFC in terms of electron acceptor and supply of external electrical energy source (0.2 V vs SHE, approximately) (Fig. 4). MEC is generally analyzed in terms of energy generation (current), columbic efficiency, hydrogen recovery, and H_2 production rate [131,143]. The production rate of H_2 is dependent on the type of substrate and external voltage supplementation. Several substrates (ex: glucose, ethanol etc.,) and wastewater were evaluated in MEC to improve the H_2 production rate. Ren et al., (2013) were the first to evaluate the use of oil refinery wastewaters as substrate in MEC. In this study, six different refinery wastewaters collected at various points at several treatment facilities in the USA were evaluated [135]. Miniatured MECs were developed and operated with replicated experimental analysis to confirm the consistency. Initially, MECs were operated alone with refinery samples, and later domestic wastewater mixed to amplify the treatment process by providing additional biocatalyst and required nutrients. The performance of MECs was evaluated based on current density generation. The diverse types of wastewaters have noticeably diverse characteristics, resulting in variations of current

Table 3
Dual chamber bioelectrochemical systems (BESs) studied with hydrocarbon wastewater and wastes for the generation of energy and/or treatment.

Type of reactor	Working electrode potential	Energy generation (V/mA/P)	Type of waste treated	Type of electrode (Anode-A, Cathode-C)	Columbic efficiency	Removal Capability	Dominant bacteria	References
Dual chamber MFC	Electrode polarized at − 0.5 V (vs SHE)	2–3 μA	Trichloroethene (TCE)	A: platinum disk C: Glassy carbon electrode	60 %	Dechlorination product: ~ 0.6μeq	*Dehalococcoides* spp.	[132]
Dual chamber MFC	Graphite cathode polarized from − 0.800 to − 1.000 V vs Ag/AgCl	N/A	Trichloroethene (TCE).	A: Platinum wire C: Graphite	N/A	Dechlorination- 50 %; Methanogenesis: 18 %	*N/A*	[133]
Dual chamber MFC	Graphite electrode (+0.5 V or − 0.3 V vs SHE)	1 mA	Trichloroethene (TCE)	A,C: Graphite	98 %	Dechlorination rates (ca. 25 μmol/day)	*Geobacter lovleyi.*	[134]
Dual chamber MFC	N/A	31 mW/m^2	Diesel range organics	A: Stain less steel C: Pt	N/A	82 %	*Citrobacter* sp., *Pseudomonas* sp., *& Stenotrophomonas* sp., *were noted on anode electrode*	[89]
Dual chamber (UMDC)	N/A	30.8 W/m^3	Synthetic wastewater (Acetate as carbon source)	A: Graphite rods C: Carbon cloth/ Pt/C	17 %	TDS removal rate-7.50 g TDS L^{-1} d^{-1},	*N/A*	[123]
Dual chamber (Mini MEC)	0.7 V	2.1 A/m^2	Deoiled refinery wastewater + domestic wastewater	A: Graphite plate C: Stainless Steel mesh	N/A	COD removal − 89 %	*N/A*	[135]
Dual chamber MFC	N/A	500 ~ 700 μA	Sulfidic benzene-contaminated groundwater	A,C: Graphite fiber material	18 ~ 32 %	142–282 μmol	*Mixed culture dominated by δ-Proteobacteria (*i.e. *Desulfobacteraceae, Desulfobulbaceae and Geobacteraceae)*	[136]
Dual chamber MFC	−0.2 V vs Ag/AgCl	1.25 ± 0.11 mW/m^2	Phenanthrene with Inoculum	A: Carbon felt C: Carbon felt/ platinum (Pt)	N/A	COD removal-70.43 %; phenanthrene degradation rate- 27.30 μM/d	*Shewanella oneidensis, Pseudomonas aeruginosa, mixed cultures and combinations*	[110]
Dual chamber MFC	N/A	4.89 mA/m^2,	Minimal medium (Hydrocarbon-contaminated wastewater)	A: Carbon felt C: Carbon felt/ platinum (Pt)	N/A	benzene + phenanthrene removal-91.6 %; COD removal-79.1 %	*N/A*	[107]
Dual chamber BES	Applied voltages (1.0, 1.5 and 2.0 V)	−0.78 mA	Wastewater (Acetic acid and Butyric acid)	A: Carbon cloth/ Pt C: Carbon brush	4.60 %	N/A	Mixed consortia	[131]
Dual chamber MFC	N/A	225 ± 1.4 mW/m^2	Petroleum refinery wastewater	A: Carbon cloth C: Carbon cloth with platinum coating	Batch 10 % 8 hr(HRT) 2 %, 16 hr (HRT) 2 %	Phenol 85 ± 1.5 %; sulfide 79.5 ± 1.2 %	Mixed consortia	[137]
Dual chamber BES	+0.4 V vs Ag/AgCl (on cathode)	−20.47 mA	Petroleum produced water (PPW)	A,C: Activated carbon fabric	N/A	COD removal in Control: 23.2 %; Synthetic PPW: 76 %; real PPW: 69 %	*Gammaproteobacteria, Alphaproteobacteria, Clostridia, Betaproteobacteria & Deltaproteobacteria during real PPW. Bacteroidetes,* *Clostridia, during syn PPW*	[105]
Dual chamber MFC with cathode packing	N/A	GAC (330 mW/m^2) GG (262 mW/ m^2)	Petroleum refinery wastewater	A: Graphite rod C: Graphite flakes and granular activated carbon	N/A	Control: 66 %; GAC: 82 %; GG: 84 %	*Paenibacillus* sp. *and Deinococcus* sp.	[106]
	N/A				N/A	COD removal- 62.22–70.50 %	Mixed consortia	[18]

(*continued on next page*)

Table 3 (*continued*)

Type of reactor	Working electrode potential	Energy generation (V/mA/P)	Type of waste treated	Type of electrode (Anode-A, Cathode-C)	Columbic efficiency	Removal Capability	Dominant bacteria	References
Dual chamber MFC		3.35 mA, 1.12 mW,28.27 W/m3	Petroleum refinery wastewater (PRW) and Labanah whey wastewater (LW)	A,C: Cylindrical graphite				
Dual chamber (OsMFC)	N/A	7.467 W/kg COD, 136 mA/m^2 and 48.52 mW/m^2	Real wastewater and seawater	A: Carbon fiber brush C: Carbon cloth	N/A	COD removal-93 %; Salts removal: 48 %	Mixed consortia	[138]
Dual chamber MFC	Applied potentials	1089 mW/m^2	Petroleum refinery wastewater (PRW)	A: Graphite brush C: Platinum coated carbon cloth		Sulfates removal- 93.9 %; TPH removal- 53.1 %; COD removal 60.2 %	Mixed consortia	[8]
Dual chamber (EC and MFC)	Current densities- 26, 36, 48, 59 and 71 mA/cm^2.	Power − 2.74 mW	Produced water (PW)	A: Carbon brush C: Carbon cloth	17.30 %	TPH removal-89 %; COD removal-89.6 %; Sulfate removal- 42.6 %; TDS removal- 34.3 %.	Mixed consortia	[139]
Dual chamber MFC	771 mV	Power density − 14.04 W/m^3	Shale gas fracturing wastewater and leachate mixture	A: Carbon felt C: Carbon brush	15.9 % to 29.8 %	COD removal in anolyte- 61.9 %; COD removal in catholyte- 60.3 %	*Gammaproteobacteria, Alphaproteobacteria, Actinobacteria, Bacteroidia*	[140]
Dual chamber MFC		2636 ± 476 mW/m^3	Shale gas flowback water	A: 1071 HCB carbon felt C: Carbon cloth		COD removal 72 ± 7 %	*Bacteroidetes, Firmicutes, 50 Proteobacteria, and Chloroflexi*	[141]

generations. Among all the refinery wastewater evaluated, the de-oiled refinery sample exhibited better performance with 79 % COD removal and 2.1 A/m^2 of current density. These values were similar to the results observed by using domestic wastewater as a substrate. Early studies by Skadberg et al., (1999) reported that bioelectrochemical generation of H_2 as a suitable mechanism for degradation of 2,6-dichlorophenol by dehalogenation reaction by poising current in the range of −1 to −15 mA [144]. A similar mechanism was observed by Aulenta et al., (2008) for the reduction of tricholoroethene [133]. The feasibility of hydrocarbons degradation was less, due to the demand of higher energy input. The use of microbial electrochemical systems would be efficient in comparison to the electrochemical process due to a decrease in energy requirement. However, the side reactions which consume H_2 can lead to a decrease in performance. So, the direct electron transfer by cathode to reduce pollutants without H_2 generation can be an alternative strategy.

Furthermore, few studies have pointed the usage of electron shuttles (mediators) between the electrode and biocatalyst for enhancement in electron transfer rate. In one such study, electron shuttles were used to convert TCE (trichloroethene) to ethene via *cis*-DCE (dichloroethene) and vinyl chloride (VC) [132]. TCE removal in BES systems has also proven that a direct electron transfer mechanism from the cathode can be beneficial. The pure strain of a *Geobacter lovely* was able to convert the more oxidized form of TCE (i.e., tetrachloroethene, PCE) to *cis*-DCE by using the electrode as the sole electron donor [134]. In another study by Lohner and Tiehm (2009), the PCE converted to ethene by using H_2 from the water electrolysis. The generated O_2 from electrolysis stimulated microbial oxidation for dechlorination [90]. BES systems inoculated with mixed culture could dechlorinate 1,2-DCA by enriching *Dehalococcoides* species. Li et al., (2010), had operated a sediment electrokinetic reactor for the degradation of petroleum hydrocarbons with an applied voltage of 1 V/cm. Their study noted that an increase in external voltage had increased hydrocarbon degradation by amplifying the bacterial metabolic activity rate, which uses the petroleum carbons as a substrate (Table 4) [145]. Effective electrode materials with well-developed electrochemically active anodic biofilm dictates the overall process efficiency. High TDS concentrations pertaining to petroleum wastewaters is an added advantage for BES systems. Anolyte TDS concentrations positively influences the electron conductivity which further results in improved efficiency. The external power needed for the MECs is in the range of 0.6 V and 2.0 V which can be easily achieved from the photovoltaics. This kind of approach also initiates the possibility of participating BES system in biorefinery concepts.

3.4. Methane generation with MEC

Petrochemical wastewater with a high concentration of COD has tremendous potential for the generation of value-added products via MEC process with simultaneous treatment of pollutants. Biomethane recovery from petrochemical wastewater using anaerobic digestion has been reported [152,153]. The study considered a total of 8 different petrochemical wastewaters which coded as PTA (terephthalic acid), EO/EG (ethylene glycol), PVA (polyvinyl alcohol), PET (polyester), SCN − OSS (one-step-cyanide), SCN-TSS (two-step-cyanide), OSAA (one-step-acid-alkali), TSAA (two-step-acid-alkali) for the conversion of wastewaters to produce methane in anaerobic digestion. The process has recorded maximum methane yield of 305.9 ± 2.7 mL from gram of COD with purified PTA wastewater as substrate. EO/EG and PET found to exhibit lower methane yield than purified terephthalic acid. Comparatively, SCN-OSS wastewater produced the lowest methane yield of 4.7 mL from gram of COD. This drop in methane can be due to high toxicity and low biodegradability of SCN-OSS. The kinetic simulation studies showed a strong dependency of biodegradability, mineralization behaviors, and methane productivity on the characteristics of petrochemical wastewater. This demonstrates the technical feasibility of anaerobic biotechnology in treating most petrochemical wastewaters [154]. The co-digestion of petroleum wastewater with beef and cattle

Table 4
Hybrid types bioelectrochemical systems (BESs) studied with hydrocarbon wastewater and wastes for the generation of energy and/or treatment.

Type of reactor	Working electrode potential	Energy generation (V/mA/P)	Type of waste treated	Type of electrode (Anode-A, Cathode-C)	Columbic efficiency	Removal Capability	Dominant bacteria	Reference
Sediment electrode BES	+0.3 V vs Ag/AgCl (on Anode)	− 1 mA	TPH in soil	A,C: Graphite stick	N/A	Napthelene, benzene and toulene were majorly degraded	*Geobacter metallireducens*	[146]
Sediment MFC	N/A	2.1 mA	Water-logged soil with organic pollutants	A: Carbon felt C: Pt. coated cloth assembly	3.70 %	90 % in closed circuit; 27 % in open circuit using phenol as major component	N/A	[147]
U-tube MFC separated by membrane MFC	N/A	0.85 ± 0.05 mW/m^2	Petroleum hydrocarbons	A,C: Carbon mesh	N/A	TPH removal-15.2 % (120 % higher rate in closed circuit than in open circuit	N/A	[111]
Sediment MFC	N/A	17 mV	Contaminated lake sediments	A,C: Stainless Steel cylinders	N/A	Phenanthrene removal-99 %; pyrene-94 %	*Proteobacteria, Chlorofexi, Firmicutes,*	[148]
Fed-batch (up-flow) MFC	N/A	0.501 mW/m^2	Distillery wastewater	A,C: Plain graphite plates	N/A	COD removal- 72.84 %	Mixed consortia	[15]
Three chamber MFC	N/A	136.30 mA/m^2	Raw produced water	A,C: Graphite	16.69 %	COD removal- 92 %; TDS removal-80 %,	N/A	[122]
ABR-MEC with Four sequential compartment	Applied potential: 0.6 to 1 V	48.5 mA	Petrochemical wastewater	A: Carbon cloth C: Stainless Steel mesh	81.46 %	COD removal- 96.5 %; Maximum CH_4 production rate- 142 mL/day	N/A	[149]
Phyto-microbial-electrochemical system (PMES)	0.7 V	–	Hydrocarbon contaminated soil	Graphite tube as working electrode; Stainless steel counter electrode	–	TPH removal-18.0 ± 3 %	*Dietzia, Georgenia and Malbranchea*	[150]
Sediment microbial fuel cell (SMFC).	–	–	Crude oil contaminated sediment	Carbon fiber brushes with and without Ferric iron	–	Fe SMFCs reached 22.0 ± 5.5 %	*Gordonia*and *more*	[151]

manure could enhance methane production by 50 % and COD reduction by 98 ± 0.5 % at 10 days HRT. The methane yield increased by 50–60 % (mesophilic conditions) and 50–65 % (thermophilic conditions) with co-digestion in comparison with the digestion of PWW alone [155–157]. Further, the same research group studied the influence of microwave and ultrasonic pre-treatment on methane generation from anaerobic digestion of petrochemical wastewater and waste-activated sludge [158]. The results revealed that co-digestion of waste-activated sludge with petrochemical wastewater produced approximately 0.22 L CH_4/g VS_{added}. The anaerobic digestion performance with individual digestion of petrochemical wastewater and un-pretreated waste-activated sludge was low at 0.19 and 0.17 L CH_4/g VS_{added}, respectively. The maximum methane yields of 0.47 L/g VS_{added} and 0.33 L CH_4/g VS_{added} attained from the anaerobic co-digestion of 30-min microwave pre-treated waste-activated sludge and petrochemical wastewater, respectively. In this study, microwave pre-treatment's important role and influence in developing an energy-efficient strategy for waste management are emphasized [159].

Biological degradation of petrochemical wastewater which containing mono ethylene glycol (MEG) employing an anaerobic packed bed baffled reactor (AnPBBR) produced three different value-added byproducts such as hydrogen, ethanol, and methane. The maximum CH_4 production rate of 237.80 ± 21.67 mL/L/d achieved at organic loading rate (OLR) of 4 g COD/L/d at HRTs of 36 h revealing the potential use of AnPBBR in treatment and energy production simultaneously. However, there are limitations in the anaerobic digestion of petrochemical wastewater. These include treatment cost, large volumes of production tanks, odor nuisance, long startup period, slow growth rate of methane producing organisms, low nutrient removal, and low purity of products. Recently, MEC technology has suggested to overcome the limitations of anaerobic digestion using petrochemical wastewaters.

Arivin et al., (2019) conducted the first study on COD removal and methane production that integrated both anaerobic baffled reactor (ABR) and MEC using petrochemical wastewater [149]. The collected petrochemical wastewater from petrochemical industry was characterized by COD (1820–2200 mg/L), BOD (600–870 mg/L), pH (8.1–8.3), conductivity (1–1.2 ms/m), total phosphorous (1–2 mg/Ll), and total nitrogen (5–11 mg/L). The methane production performance of ABR-MEC reactor were investigated at various conditions of HRT and applied voltage. The maximum methane production rate was 142 ± 1 mL/day at HRT of 72 h at an applied potential of 1 V. The methane yield in ABR-MEC reactor was 1.4-fold than ABR reactor, and the COD removal efficiency in ABR-MEC (96.5 %) was also higher than in ABR reactor (66.7 %). The COD removal increased with the increase in the applied potential (0.6 to 1 V) due to the high degradation of VFAs by combining bio electrochemical system. The decrease in the COD removal observed with the decrease in the HRT due to the accumulation of VFAs, which led to bacterial inhibition under shorter HRT. The high methane production in ABR-MEC attributed to the high conversion of produced hydrogen by hydrogenotrophic bacteria capturing hydrogen and electrons for methane productions. The AD coupled MEC improved the generation and purity of biogas. Arivin et al., (2019) also reported similar MEC study using low strength petrochemical wastewater for the generation of methane in a single chamber MEC at different applied potentials and HRTs over a period of 110 days [102].

The petrochemical wastewater had a COD of 480–534 mg/L, BOD of 316–330 mg/L, pH 7.3–7.9, conductivity 1–1.1 ms/m, total

phosphorous of 0–1 mg/L and total nitrogen 6.50–8.15 mg/L. In startup phase, the MEC system operated in batch mode at an applied voltage of 0.6 V to stabilize biofilm formation. In the next phase, the applied voltage was increased from 0.6 to 1 V at HRT of 48 h and achieved a maximum COD removal (85.9 %) and methane production (63 ± 1 mL). This suggests that an increase in applied voltage positively affected COD removal, methane content, and methane production rate, using high-strength petrochemical wastewater. The COD removal was higher in batch mode than in continuous mode (75.3 %), but the methane production in continuous mode was 1.6-times higher than the value in batch mode. The availability of exogenesis bacteria in batch mode for substrate degradation was high than in continuous mode; hence the COD removal is more in batch mode. These results suggest that MEC is a promising technology for treating low-strength petrochemical wastewater and methane production. The MEC system observed the indirect electron transfer mechanism in all the phases due to the presence of hydrogen gas rather than direct electron transfer. Further studies are required to implement MEC for enhancing the methane production by exploring the effect of microbes and the organic pollutants transformation in methane recovery.

4. Bioelectrochemical treatment (BET) through BES systems

Application of BES such as MFC and MEC processes operating to treat organic and inorganic pollutants using electrogenic biocatalyst has emerged as an distinctive system capable of converting chemical energy to electrical/hydrogen and other value-added products by a simultaneous decrease in toxic products. Over the conventional systems, BES would differ in operation with electrodes and by supplementation of additional energy if required. In contrast to conventional fuel cells, BES operate comparatively under minimal operational conditions, such as room temperature, a requirement of expensive metal catalysts, etc. The current aspects in application of BES to treat complex waste and the development of alternative materials for electrodes, separator, and reactor configurations have made BES a viable technology. However, the use of polyaromatic hydrocarbons, alkanes, alkenes, and halogenated compounds was limited due to their structural complexity. Further studies on BES regarding the use of petroleum-related compounds need to be encouraged and pursued with at most importance.

4.1. Hydrocarbon removal from petroleum effluents

Petroleum effluents majorly encompass hydrocarbons, particularly polycyclic aromatic hydrocarbons (PAH) [160]. The minor concentrations of PAH are known to be detrimental to the environmental ecosystems as well as human nervous system [161]. Thus, for treating PAHs using advanced biological systems such as BES is essential. In this regard, a single chamber column type pilot-scale bioelectrochemical system was evaluated for the degradation of total petroleum hydrocarbons (TPH) from the soil. Biochar (BC) anode and granule anode (GA) were evaluated for TPH degradations. BC and GA electrodes exhibited a maximum power generation of 3.4 and 8.8 mW/m^2, respectively [162]. The TPH degradations in soil were measured at different distances (18 to 26 cm) from electrode for each BES system, and removals were monitored for up to 120 days. On day five, the TPH concentration at 1 cm near the electrode was reduced by 72 and 68 % for BES equipped with BC and GA electrodes, which was 241 % increase in the removal efficiency compared to a control that was limited to 21 %. The swift increase in TPH degradation efficiency can be attributed to adsorption by the anodes or by the activity of electrogenic active bacteria at the anode using TPH as a substrate for electricity generation. By the end of the operation at around 120 days, totally 89 % TPH was removed across both the reactors, but the fraction of TPH in the soil remained the same. The small amount of unresolved TPH in the soil was similar to recalcitrant hydrocarbon fractions that attached to soil particles with higher affinity. The *n*-alkanes with C8 to C12 at different distances degraded initially within five days of BES operation. The carbon compounds, especially with C10 ~ C12, were predominantly reduced from 282 mg/kg-DS to 25 mg/kg-DS. This might be due to their availability and relatively high-water solubility. By the end of the operation (120 days), the total *n*-alkanes removed were 77 ~ 86 % at various spatial distances from the electrodes. At the same time, the control reactors were limited to 61 % removal efficiencies.

In another study from the same group, the authors evaluated the effect of surfactants (CC-S) on TPH degradation in SMFC (soil MFC) equipped with carbon cloth (CC) as the anode electrode for which maximum current generation with BC, CC, and CC-S were 85, 73, and 21 mA/m^2, respectively. Likewise, TPH removal rates increased from day five till the end of the operation (64 days). BES operation with BC removed 78 % (compared to 41 % for control), CC removed 73 % (control, 40 %) and CC-S removed 35 % (control, 30 %). The decrease in current generation and TPH removal efficiency in SMFC with CC-S might be due to the dose of the surfactant (500 mg/l) used in this study. Such a high dose might have led to increased sorption of the surfactant onto the soil particles, resulting in increased TPH partitioning. This can lead to an increment in the phase transfer of hydrophobic petroleum hydrocarbons to the aqueous phase and decrease in soil phase. Therefore, the availability of substrates for the utilization by the electrogenic active bacteria on the anodes was reduced [163].

BES system was operated using petroleum hydrocarbons as a substrate in precultured/enriched anode (EAMFC) and new anode (NAMFC) systems at different substrates concentrations (800 and 8000 mg/l) [96]. BES operation with the enriched anode (EAMFC) exhibited a higher TPH removal rate of 93 % with an initial substrate concentration of 8000 mg/l. This was followed by NAMFC with a removal rate of 83 and 79 % at substrate concentrations of 800 and 8000 mg/l, respectively. This study suggests that bioaugmentation of the BES system for TPH degradation should be a pre-requisite step for the long-term selective enrichment of active electrogens for reducing contaminants and amplifying the current generation. In other study by Li et al., (2014) had operated BES with multiple anodes and single cathode for the degradation of petroleum hydrocarbons (PH) in soil [164]. In SMFC (soil MFC), anodes were placed horizontally (HA) at 1, 3 and 5 cm away from the cathode. In this study, a maximum power density of 37 mW/m^2 was noticed on day 5 and a gradual decrease in power generations was noticed until the end of the operation (180 days), which might be due to the degradation of simple substrates in the soil. The net degradation efficiency of TPH in the soil was found to be 12 % only, which might be due to usage of aged petroleum hydrocarbons contaminated soil with high salinity. The presiding PAHs in contaminated soil were phenanthrene (C14), fluoranthene (C16), pyrene (C16) and chrysene, which were in an accounting concentration of 72 %. The degradation rates of PAHs in each layer were as follows: SL4 (36 %) > SL1 (27 %) > SL2 (19 %) > SL3 (14 %). The denaturing gradient gel electrophoresis (DGGE) profiles of anodic bacterial community revealed that *Geobacteraceae sp*. and *Escherichia* sp. played a key role in TPH degradation and electricity generation. Concurrently, same groups have studied the vertical arrangement (VA) of anode in MSF and noted a variation in energy generations and TPH removals [165]. The MFC operation with VA and horizontal arrangement (HA) exhibited a maximum voltage generation of 285 and 280 mV, respectively. The maximum degradation percentage of *n*-alkanes of TPH in HA,VA and control were 22,9, and 7.8 %, respectively.

Although both the MFC (HA and VA anode arrangements) could degrade the hydrocarbons in soil; however, the variations in removal percentages with the dependency on anode electrode arrangement were clearly unknown. Wei et al. operated a double chamber MFC for 130 days by using groundwater containing 15 mg/l of benzene and 20 mg/l of ammonium [166]. In this study, the authors noted 80 % removal of benzene from the anode. During recirculation of anodic effluents to the cathode, the total benzene from ground water was removed and showed a 100 % removal efficiency. In summary, 80 % of benzene was reduced

from anode and ammonium was reduced from cathode. Bacterial community analysis at the anode electrode of MFC revealed bacterial phylotypes belonging to *Chlorobiales, Rhodocyclales, and Burkholderiales*.

Srikanth et al., (2016) operated the BES system in continuous mode, by using the petroleum refinery wastewater as a substrate. This study noted that BES operation with longer HRT (8to 16 h) could attribute higher performance in terms of power generation and TPH removal. BES operation in batch and continuous mode of operation at 8 and 16 h of the operation resulted in a maximum power generation of 55 and 225 mW/m^2, respectively. In terms of phenol (85 % batch vs 80 % continuous mode) and oil reduction (75 % batch vs 93 % continuous mode), BES operation in both batch and continuous mode of operation had resulted in more or less similar removal percentages [137]. In another study the cathode upliftment (+0.4 V vs/Ag/AgCl) for the treatment of produced water (PW) was performed. The gravimetric analysis for hydrocarbons pointed to a significant decrease during BES operation (Synthetic PW: 76 %; and real PW: 69 %) in comparison to control (23 %) [105].

Mohanakrishna et al., (2018) studied an enhancement in the treatment of petroleum refinery wastewater was observed with the application of a short-term applied voltage in MFC. In this study, the external voltage (0.1, 0.3, 0.5 V) was provided for an initial 24 h and further pursued as a typical MFC for harvesting bioelectricity. With the application of external voltage an improved bioelectrogenesis is noted with an increase in the maximum power density, COD, and diesel range organic reduction. A maximum power density of 132 mW/m^2 was noted at a short-term applied voltage for 24 h. This was three-times higher than control MFC (45 mW/m^2) [99].The power densities, DROs and COD removal were increased with variation in reactor configuration. At an applied voltage of 0.8 V, and by using graphitic type BES reactor systems, the maximum power densities, COD removal and DRO reduction were noted to be 278 mW/m^2, 75.8 % and 91 %, respectively [18]. This performance increase is contributed to the employment of membrane-less BESand having a high electrode surface with a decrease in electrode spacing.

In a follow-up study, the authors also tested the influence of single and double chamber reactor configurations while treating the produced water containing petroleum hydrocarbons and sulfates. The authors noted elevated performance with double chamber BES configuration in comparison with single-chambered system. This study observed maximum COD, TPH and sulfate removal of 56.92 %, 51.16 %, and 91.83 %, respectively [8]. In another study, influence of co-substrate on the treatment of petroleum refinery wastewater was evaluated, in which labneh whey (LW) wastewater was used as a co-substrate along with petroleum refinery wastewater in BES. The variation in the ratios of these two wastewaters resulted in difference in power generation and COD removal. The elevated performance in terms of COD removal of 63.1 % was noted by using 80:20 ratio. On the contrary the maximum power densities was noted at 80:20 ratio. Here, the large LW ratio led to anolyte's acidic pH, which conjointly hindered the power generation and substrate degradation [2]. On similar lines, a sediment BES was operated in continuous mode for the degradation of petroleum refinery wastewater. In this study a maximum COD removal rate of 265 mg/L-day was noted with a power density of 725 mW/m^2 at 2 V [100]. In a continuing study, sewage was used as an enhancer in BES for in situ treatment of hydrocarbons in petroleum-contaminated sediments. Using sewage and acetate as an enhancer, the maximum power generation was noted to be 176 mW/m^2 and 148 mW/m^2, respectively. Likewise, in sediment BES, the maximum TPH removal with sewage and acetate as an enhancer was noted to be 57 mg/l and 22 mg/l, respectively [167]. Likewise, Guo et al. controlled the cathode to enhance the degradations of petroleum refinery wastewater. In this study, BES operation with GAC and GG exhibited a similar removal percentage of oil around 82 ~ 84 %, respectively. In contrast, control reactors (no packing) were limited to 66 % of oil removal [106]. All these studies pointed out the possibilities in reduction/utilization of phenol, benzene, TPH, and PAH in BES. However, these studies are still limited with a longer retention time for an efficient reduction process. Further research in terms of increasing the removal rate with an accelerated decrease in TPH related compounds should be focused on and encouraged.

4.2. Removal of sulfates and nitrates from petroleum-based wastewaters

In general, petroleum effluent comprises the complex wastewater streams with a broad spectrum of constituents. Among these, sulphates, nitrates, and hydrocarbons are the most studied in BES and AD systems dealing with petroleum effluents. In AD, nitrates and sulfates acts as terminal e- acceptors to microbes, leading to lower and discerning removals of hydrocarbons. For instance, Davis et al., (1998) studied the CO_2 production rate in sand column treating crude oil to observe that aerobic treatment of hydrocarbons is severalfold higher than the anaerobic treatment [168]. Such slower removals can be overcome by choosing the BES, in which electrogenic environment at the anode can enhance the treatment process with concurrent product generation.

4.2.1. Sulfates

In BES, sulfate-reducing bacteria play a crucial role in treating sulfate. Rabaey et al., (2006) demonstrated the efficacy of BES in the treatment of sulfide from wastewater with a concurrent generation of bioelectricity [169]. Moreover, sulfides and their various forms were present in numerous wastewater streams, and several researchers have published many reports on their removal in BES and electrochemical systems [170]. In applied energy (voltage/current) systems (ex: MEC), the fate of sulfur removal depends on the energy provided. Table 5 summarizes the reactions of sulfur removal in aqueous solution. The conversion of sulfate to sulfide and further oxidation to elemental sulfur is the most frequent method employed in treating sulfate. Commonly, the treatment/conversion of sulfur requires a minimum voltage input, and there is less possibility of reconverting it to sulfide owing to bioelectrogenic environment. Also, sulfide is less toxic than other forms and is easily recoverable [171].

Typically sulfides are often detected in gaseous forms (H_2S) with a higher toxicity value of 100 mg/l [173]. Borole (2010) patented a BES system for treating various gaseous pollutants such as H_2S; Borole and Tsouris [174,175] patented an MFC system for treating fuel processing wastewaters with a concurrent treatment of oxidative hydrocarbons to generate electricity. Further, Zheng et al., (2014) evaluated the BES

Table 5
Theoretical potential required to remove various sulfur forms present in an aqueous solution. ().

Reaction	Theoretical potential (V vs SHE)
Acidic solutions	
$S_2O_8^{2-}$ (aq) + 2e$^-$→ 2SO_4^{2-} (aq)	1.96
2SO_4^{2-}(aq) + 4H^+ + 2e$^-$ →$S_2O_6^{2-}$(aq) + 2H_2O	−0.25
SO_4^{2-}(aq) + 4H^+ + 2e$^-$ →H_2SO_3(aq) + H_2O	0.16
2H_2SO_3(aq) + H^+ + 2e$^-$ →$HS_2O_4^-$(aq) + 2H_2O	−0.07
4H_2SO_3(aq) + 4H^+ + 6e$^-$ →$S_4O_6^{2-}$(aq) + 6H_2O	0.507
HS_2O_4(aq) + H^+ + 2e$^-$ →$S_2O_3^{2-}$(aq) + H_2O	0.87
$S_2O_3^{2-}$(aq) + 6H^+ + 4e$^-$ →S(s) + 3H_2O	0.6
S(s) + 2e$^-$ + 2H^+ →H_2S (aq)	0.144
S_2Cl_2(g) + 2e$^-$ →2S(s) + 2Cl^-(aq)	1.19
Basic solutions	
SO_4^{2-}(aq) + H_2O + 2e$^-$ →SO_3^{2-}(aq) + 2OH^-(aq)	−0.94
2SO_3^{2-}(aq) + 3H_2O + 4e$^-$ →2$S_2O_3^{2-}$(aq) + 6OH^-(aq)	−0.58
2$S_2O_3^{2-}$(aq) + 3H_2O + 4e$^-$ →2S(s) + 6OH^-(aq)	−0.74
S(s) + 2e$^-$ →S^{2-}(aq)	−0.45
2SO_3^{2-}(aq) + 2H_2O + 2e$^-$ →2$S_2O_4^{2-}$(aq) + 4OH^-(aq)	−1.13
$S_2O_4^{2-}$(aq) + 4H_2O + 6e$^-$ →2S(s) + 8OH^-(aq)	−0.50
SO_3^{2-}(aq) + 3H_2O + 4e$^-$ →2S(s) + 6OH^-(aq)	−0.66

Adapted from [172]

efficiency in treating high-strength sulfate and organic-rich wastewater (brine) and noted the change in microflora with varying pH. It was observed that *Desulfovibrio* were dominant at acidic pH conditions (4.5 and 6.5). By changing BES pH to basic, *Desulfomicrobium* was the highest [176].

In another study, Rabey et al., (2010) demonstrated the application of MEC in generating a caustic soda at a liter scale by employing local onsite brewery wastewater with highly alkaline (pH 12.5) cathodic conditions. It also has been known that electrode potentials can influence the removal of pollutants by enhancing the growth of electrogenic microbes. Pozo et al., (2016) hypothesized that autotrophic sulfate reduction depends on electron flux from cathode to sulfate via H_2 gas being an intermediate electron source [177]. Whereas, in the case of abiotic treatment of sulfate, the poised potential at the cathode dictates the type of reaction and end product.

4.2.2. Nitrates

Along with sulfates, nitrates are also remediated during the treatment of petroleum effluents in BES [27]. The nitrates are converted to nitrogen gas during the denitrification process by forming an intermittent nitrite and nitrous oxide (Table 6). It is well known that autotrophic denitrification of nitrates is beneficial over the heterotrophic denitrification, which depends on the organic carbon. Moreover, theoretical redox potential for the conversion of nitrate to nitrogen gas (+0.74 V vs SHE) is closer to oxygen (+0.82 vs SHE) [178], thereby making it an ideal electron acceptor in MFC systems after O_2. However, enhanced autotrophic denitrification rates are achieved by employing the applied voltages and poised potentials in BES. The increase in applied voltages and poised potentials has led to increased denitrification rates [179]. It was also noticed that variation of intermediatory product concentration with a change in electron flux. For instance, employing BES, the N_2O concentration was negligible, whereas without electron stimuli, N_2O concentration were around 300 ppm [180]. By the application of 0.7 V, the biotic cathode of BES exhibited similar nitrate removal as the expensive platinum cathodes [181]. These autotrophic denitrifying biocathodes were enriched with *clostridia* [78]. Also, biocathodes could denitrify nitrates and nitrites simultaneously. It was noted that 88 and 85 % during the individual removal of nitrates and nitrites, respectively. The variation in HRT could alter the denitrification rate; however, using longer HRT, a higher removal was achieved in comparison to shorter HRT. Also, increase in surface area by using multiple cathodes has increased the nitrate removal [182]. Likewise, it can be presumed that nitrates in petroleum effluents can be effectively remediated in BES.

5. Bio-remediation technologies for the treatment of petroleum-related compounds

Petroleum and its related products such as diesel, kerosene, and gasoline are significant sources for energy generation in daily life. Spills and leaks can occur regularly due to exploration, transportation, production, refining, and storage. Every year, it is estimated that around 600,000 metric tons of natural crude oil seepage with a range of uncertainty of around 200,000 metric tons is happening. The release of contaminants to the environment either by accident or due to human activity can lead to water and soil pollution. Soil contamination with petroleum-related compounds and other toxic-related pollutants can damage animals and plant tissue by death or mutations. Soil remediation technologies include mechanical treatment, evaporation, dispersion, washing, and burying. Nonetheless, these technologies are expensive and can lead to incomplete decomposition of pollutants. In this regard, the bioremediation process has gained importance due to its simplicity and cost effectiveness; for detoxifying contaminants using microorganisms.

There are two known approaches for the remediation of oil in the soil. (1) bioaugmentation of well-known oil-degrading microbes to existing soil microbial community and (2) biostimulation of oil degrading microbes by the supplementation of nutrients and co-substrates (Table 7). Adebusoye, et al., (2007) isolated nine microbial strains namely *Pseudomonas fluorescens, P. aeruginosa, Bacillus subtilis, Bacillus* sp., *Alcaligenes* sp., *Acinetobacter lwoffi, Flavobacterium* sp., *Micrococcus roseus, and Corynebacterium* sp., from polluted tropical streams in Nigeria that are capable in degradation of crude oil [184].

Jonas et al., (1983) have extensively studied the marine sediments to isolate the microbes capable of utilizing the alkyl aromatics as a substrate. In their study *Arthrobacter, Burkholderia, Mycobacterium, Pseudomonas, Sphingomonas, and Rhodococcus* were actively found to be involved in alkyl aromatic degradation [196]. Chaillan et al., (2004) isolated fungal genera namely *Amorphoteca, Neosartorya, Talaromyces, and Graphium and yeast genera,* namely, *Candida, Yarrowia, and Pichia,* from petroleum-contaminated soil, and these species of fungi are proven to be potential microbes for aromatic hydrocarbon reduction [185]. Similarly, Singh et al., (2006) pointed the terrestrial fungal groups,

Table 6
Summary of the denitrification reaction involved in the BES during autotrophic reduction of nitrates ().

Denitrification reaction	Theoretical potential (V vs SHE)
$NO_3^- + 2e^- + 2H^+ \rightarrow NO_2^- + H_2O$	0.430
$NO_2^- + e^- + 2H^+ \rightarrow NO + H_2O$	0.348
$NO + e^- + H^+ \rightarrow 0.5N_2O + 0{\cdot}.5H_2O$	1.172
$0.5N_2O + e^- + H^+ \rightarrow 0.5\ N_2 + 0{\cdot}.5H_2O$	1.352
$NO_3^- + 5e^- + 6H^+ \rightarrow 0.5\ N_2 + 3\ H_2O$ (Overall)	0.746

Adapted from [183]

Table 7
Bioremediation studies other than bioelectrochemical systems (BESs) for remediation of contaminants.

Type of waste treated	Dominant bacteria	References
Polluted tropical stream	*Acinetobacter lwoffi, Corynebacterium* sp., *Pseudomonas aeruginosa*	[184]
Petroleum-polluted soils and cyanobacterial mats	*Aspergillus, Penicillium, Fusarium, Amorphoteca, Neosartorya, Paecilomyces, Talaromyces and Graphium. Candida, Yarrowia and Pichia. a Gordonia, Brevibacterium, Aeromicrobium, Dietzia, Burkholderia* and *Mycobacterium*	[185]
Kerosine hydrocarbons	*Scolecobasidium*	[186]
Industrial and municipal sewage.	*Candida famata* and *Rhodothorula rubra*	[187]
Contaminated soil	*Scolecobasidium*	[188]
Active Oil Field	*Pseudomonas, Vibrio, Moraxella, Acinetobacter,* and *Aeromonas*	[186]
Hexadecene-1, pristane, hexadecane, dibenzothiophene, anthracene, and decalin	*Nocardia, Pseudomonas, Flavobacter, Vibrio,* and*Achromobacter species*	[189]
Petroleum hydrocarbons	*Alcanivorax, Cycloclasticus, Marinobacter, Neptunomonas, γ -Proteobacteria*, and genus *Planococcus* with in Gram-positive bacteria	[190]
Petroleum-contaminated soil	*Pseudomonas aeruginosa*	[191]
Polycyclic aromatic hydrocarbons (Anthracene, Pyrene)	*Pycnoporussanguineus H1*	[192]
Hydrocarbons in petroleum tank bottom oil sludge	*Shewanallachilikensis, Bacillus firmus,* and*Halomonashamiltonii*	[193]
Petroleum wastewater (PAHs petroleum hydrocarbons and phenolic compounds.	*Stenotrophomonas* sp. S1VKR-26	[194]
Flowback and produced water (Phthalate esters (PAEs), PAHs and petroleum hydrocarbons)	Halophilic and halotolerant microorganisms	[195]

namely *Aspergillus, Cephalosporium, and Penicillium,* that are capable of treatment of crude oil hydrocarbon contaminated soil [197]. Bogusławska-Was et al., (2001) studied yeast species and suggested that *Candida lipolytica, Rhodotorula mucilaginosa, Geotrichum sp, and Trichosporon mucoides* are capable of degradation of petroleum hydrocarbons in contaminated water [187].

Bacteria, yeast, and fungi mostly degrade petroleum contaminants in environment. The degradation efficiency varied from 6 % to 82 % for soil fungi [188,198], and 0.13 % to 50 % for the soil bacteria [188,198], and 0.003 % to 100 % for marine sediment bacteria [186,189]. Several studies have reported that mixed micro flora with different enzymatic reactions and capacities are required to degrade crude oil from soil, groundwater, and marine sediments. The removal percentages in these studies might be due to differences in operational parameters such as temperature (affects the solubility of hydrocarbons) and the use of nutrients (supplemental energy source for microbial metabolism). Hydrocarbons and oils are hydrophobic in nature, making their availability to bacteria limited, which can result in a slower reduction rate. The addition of surfactant to contaminated soil can help in desorbing of hydrocarbons and can lead to the enhancement of remediation process. Microbially synthesized surfactants such as rhamnolipids which contain rhamnose moieties and fatty acid tail, could help to increase the oil surface area, making it available for the bacterial degradation [190,191].

The operation of BES alone or the combination of electrochemical with biological process proves to be better in the remediation of petroleum hydrocarbons in terms of achieving a higher removal rate. For instance, the operation of BES has exhibited a removal percentage of 49.38 % during the treatment of 320 mg/l of TPH. Under similar conditions, the conventional bioremediation system has displayed only an 8.75 % removal of TPH. Suggesting that the electrogenic environment at the anode with active electrogens in MFC has facilitated the enhanced removal of TPH. However, these power generations can be varied with variation in the type of organic matter available in the wastes. Also, high-strength petroleum wastewaters rich in long chain hydrocarbons can limit power generation and CE in BES. Therefore, treating petroleum hydrocarbons with co-substrates like conventional simple organic wastes (ex: domestic wastewaters) can be beneficial. It should also be noted that the overall power generation in MFC, especially during the treatment of petroleum hydrocarbons, can be smaller due to long chain hydrocarbons compared to MFC operation with simple sugars like acetate. Therefore, BES treating petroleum hydrocarbons should be integrated with other processes like MES or AD [199] to generate value-added products (ex: acetate or ethanol) [200–204]. Through this integration (BES-MES) process, the regular abatement of GHGs can be achieved along with the reduction of CO_2 generated at the anode during the treatment of TPH. Overall, removing petroleum hydrocarbons benefits BES over the conventional bioremediation process.

6. Future perspectives

The production of petroleum wastes is increasing with an increase in the global energy demand. It is becoming increasingly essential to treat them in a sustainable way and in the process if we could produce products such hydrogen and methane, it is a win–win situation. Potentially MFCs can be used as a biosensor to measure COD removal based on the voltage generation. Further research to utilize petroleum hydrocarbons in MDC needs to be encouraged for maintaining energy neutral and biorefinery approach. By exploiting the organics in wastes and wastewater, MES can be promising for the H_2 generation. The limited biodegradability of petroleum waste, on the other hand, is a challenge, and more studies as to how to use this waste should be encouraged.

Most studies reveal that the product requires a longer retention time and has a lower removal efficiency, indicating that more research is needed. Until the individual processes are developed to their full potential of treating wastes independently, promising combined treatment such as microwave pre-treatment followed by MEC, anaerobic baffle reactor (ABR), etc., could be the immediate way forward for addressing petroleum wastewater in an energy neutral way.

It is necessary to undertake research on the effects of various bacteria on the production of hydrogen, methane, and other synthesized products. To find potential intermediates and techniques to accelerate them to end products, the degradation paths of the pollutants must be followed. In bio electrochemical systems, high-throughput experiments are critical for obtaining consistent results for scaleup investigations. This will necessitate the development of novel reactor designs with an elevated level of data gathering, and analysis using online computer systems to optimize and improve process conditions for yielding desirable products as output. To investigate the synergistic impacts of various process factors on the total yield, mathematical and statistical tools must be used in the conversion studies of petroleum wastes to valuable water and products.

The future of chemical synthesis production from petroleum wastes is influenced not only by technological advancements such as genetically altered microbes and complex bioreactor architecture, but also by cost economics and the separation of high purity products. Although the BES has been identified as a promising strategy for treatment of petroleum waste and production of hydrogen and methane, the rate and cost of production are still impeding the technology's adoption on a large scale. Demonstration of large capacity pilot size BES systems in one of the petrochemical industries is a huge challenge and a pressing requirement.

7. Conclusions

Petroleum-based wastewater and petroleum sludge have the huge potential for bioelectrogenesis, value-added products and biofuels generation through their stabilization and treatment. The petroleum waste/wastewater is not readily soluble in water, and so using mild surfactants is crucial to improve the biological availability at non-inhibitory conditions. Bioelectrochemical potential drives the versatile processes required for extended treatment efficiency in a sustainable approach. The BES process is a multidisciplinary field that demands an understanding of different processes together towards advancement and can be integrated with other biological and/or electrochemical treatment processes for better treatment. In BES processes, anodic oxidation and cathodic reduction processes were quite contrary to each other. Most of the degradation can be preceded at best with the combination of these processes. As the BES hosts both the processes and such electrochemical conditions can be generated from the biological catalyst since it requires no or low energy input. Therefore, valorization of such waste resources will permit sustainable development.

CRediT authorship contribution statement

Sanath Kondaveeti: Validation, Investigation, Writing – original draft. **Dhivakar Govindarajan:** Validation, Investigation, Writing – original draft. **Gunda Mohanakrishna:** Conceptualization, Validation, Investigation, Writing – original draft. **Dayakar Thatikayala:** Writing – original draft. **Ibrahim M. Abu-Reesh:** Validation, Supervision, Writing - review & editing. **Booki Min:** Validation, Investigation. **Indumati M. Nambi:** Validation. **Riyadh I. Al-Raoush:** Validation, Writing - review & editing. **Tejraj M. Aminabhavi:** Validation, Writing - review & editing.

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.

Data availability

This is a Review article. The data used in this review is available in scientific articles across leading publishers

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