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APPLICATION OF MICROBIAL FUEL CELLS FOR BIOREMEDIATION OF ENVIRONMENTAL POLLUTANTS: AN OVERVIEW

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ABSTRACT

Microbial fuel cells (MFCs) have been demonstrated as a challenging and promising technology towards management of various environmental problems. Bioremediation has been accepted widely as viable option utilizing the naturally inhabited microorganisms, but sometimes the severe low efficiency of the microorganisms and poor controllability limits its application. In this context, MFC technology can be used as a potential means to stimulate bioremediation for effective removal of various pollutants. The special features including energy saving, less sludge and energy production make MFCs as outstanding technology compared to conventional technologies. This article is mainly focused on application of MFCs towards removal of various environmental pollutants viz. antibiotics, synthetic dyes, phenolic compounds, nitrogen based compounds, ethyl acetate, toluene, polycyclic aromatic hydrocarbons, perchlorate, pesticide, sulphur, emerging contaminants, trace organic compounds etc. from aqueous environment. Although the current applications of MFC technology is still at laboratory level, it will definitely prove a great potential towards commercial applications in near future.

Keywords: Bioremediation, microbial fuel cell (MFC), pollutants, stimulation, wastewater

INTRODUCTION

Microbial fuel cells (MFCs) have gained enormous global importance over the last few decades. It is a device which uses bacteria for the oxidation of organic and inorganic substances in the anode for the bioremediation of pollutants along with the generation of bioelectricity **(He *et al.*, 2015)**. The major advantages of MFC technology towards remediation of pollutants include (i) less production of activated sludge (ii) gas treatment not necessary (iii) energy input not needed for aeration (iv) conversion of substrate energy to electricity directly and (v) can be operated at any temperature.

Wastewater containing various pollutants is the major causes for environmental pollution in surface and ground water bodies **(Gude, 2016)**. Researchers across the globe have investigated the application of MFCs for the treatment of different types of wastewaters viz. municipal or domestic wastewater **(Khan *et al.*, 2017)**, food processing wastewater **(Boghani *et al.*, 2017)**, protein food industry wastewater **(Ieropoulos *et al.*, 2017)**, acidogenic food waste leachate **(Li *et al.*, 2013a)**, beverages industry wastewater **(Çetinkaya *et al.*, 2015)**, beer brewery wastewater **(Köroglu *et al.*, 2014)**, winery wastewater **(Penteado *et al.*, 2015)**, confectionary industry wastewater **(Patil *et al.*, 2009)**, dairy industry wastewater **(Mansoorian *et al.*, 2016)**, yoghurt wastewater **(Cercado-Quezada *et al.*, 2010)**, agro-processing industry wastewater **(Gurung and Oh, 2015)**, cassava mill wastewater **(Sangeetha and Muthukumar, 2013)**, palm oil mill effluent **(Nor *et al.*, 2015)**, mustard tuber wastewater **(Guo *et al.*, 2015)**, livestock industry wastewater **(Katuri *et al.*, 2012)**, animal carcass wastewater **(Li *et al.*, 2013b)**, swine wastewater **(Cheng *et al.*, 2014)**, mining and allied industry wastewater **(Choi and Hu, 2013)**, recalcitrant pharmaceutical industrial effluent **(Zhang *et al.*, 2015a)**, steroidal drug production wastewater **(Liu *et al.*, 2012)**, paper recycling industry wastewater **(Kassongo and Togo, 2011)** and petrochemical industry wastewater **(Morris and Jin, 2012)**. The MFC technology has been accepted because of its eco-friendly approach compared to conventional technologies towards the control of environmental pollution.

Moreover, several reviewers have focused on various issues related to MFC technology including designs and configurations of MFCs, surface modifications of electrodes and electrode surface, power generation, analysis of microbial communities, operational conditions for performance and biofilm formation, various challenges and possibilities, fundamental electron transfer mechanisms, organic removal capacities, environmental impacts and economic answers towards industrial commercialization of MFCs etc. **(He *et al.*, 2015; Gude, 2016; Tharali *et al.*, 2016)**. However, a comprehensive review on the application of MFCs towards remediation of pollutants is still lacking. Therefore, the present review aimed at summarizing the recent reports on application of MFCs towards removal of pollutants from aqueous environment.

Microbial fuel cell

Microbial fuel cell (MFC) unit is composed of an anode, cathode, cation sensitive membrane and an external wire. Aerobic and anaerobic conditions are maintained at the cathode and anode respectively. MFC can be operated with or without a mediator. Addition of external bacteria is needed for the oxidation of the substrate in case of mediated fuel cells **(Saeed *et al.*, 2015)**. The microorganisms in the anodic chamber are provided with a favourable substrate which are anaerobically degraded and release electrons. These electrons are transported from anode to the cathode through external circuit and the generated protons are passed selectively through membrane. In MFCs, chemical energy is being converted to electrical energy by oxidation of various carbon sources or organic wastes carried out by electrochemically active bacteria. There are different designs of MFCs depending upon the chambers viz. single chambered MFC, two-chamber MFC, stacked MFC etc. **(Tharali *et al.*, 2016)**. Different types of MFCs viz. mediator-less MFC **(Moon *et al.*, 2006)**, membrane-less MFC, **(Jadhav *et al.* 2013)**, catalyst coated electrode MFCs **(Rahimnejad *et al.*, 2015)**, sediment-type MFCs **(Thomas *et al.*, 2013)** have also been reported. Proton exchange membranes (PEM) are important component of MFC which assist in separation of the anode and cathode chambers facilitating transfer of protons to cathode for sustaining the electric current **(Chae *et al.*, 2007).**

Environmental pollutants

The application of MFCs for the remediation of various environmental pollutants viz. antibiotics, synthetic dyes, phenolic compounds, nitrogen based compounds, ethyl acetate, toluene, polycyclic aromatic hydrocarbons, perchlorate, pesticide, sulphur, emerging contaminants, trace organic compounds have been discussed below.

Antibiotics

Antibiotics present in the aqueous environment have been considered as pollutants over the last few years **(Popa *et al*., 2014)**. There is an urgent need for the removal of antibiotic compounds from wastewater releasing into the environment **(Ahmed *et al*., 2015).**

Chloramphenicol (CAP), a broad-spectrum nitroaromatic antibiotic has been extensively used in the animal breeding industry due to its cost effectiveness. But CAP has been banned for use in many developed countries due to its serious toxicity causing bone-marrow depression and aplastic anaemia in humans and animals **(Martelli *et al*., 1991)**. Recently, **Zhang *et al*. (2017a)** studied the performance of a microbial fuel cell (MFC) for the degradation of chloramphenicol (CAP) which showed 84 % degradation of 50 mg l^{-1} CAP within 12 h in the MFC.

The rapid degradation of sulphamethoxazole (SMX), a broad-spectrum antibiotic has been reported using microbial fuel cell (MFC) (Wang *et al*., 2016). SMX and its degradation product, 3-amino-5-methylisoxazole (3A5MI) were found to be effectively degraded in MFC reactors. Degradation of SMX (20 ppm) was approximately 85 % within 12 h. The biodegradation rate was more rapid that has been reported so far in literature. The degradation product, (3A5MI), a toxic chemical which was formed in the SMX degradation process, could be further mineralized. Antibacterial activity test showed that the biotoxicity of SMX towards *Shewanella oneidensis* MR-1 and *Escherichia coli* DH5a was greatly reduced after MFC treatment. ATP level of the MFC microbe was almost three times higher than that in open-circuit controls which might be the reason for rapid degradation of SMX in MFCs.

Paracetamol (PAM) is a common analgesic and antipyretic drug which is generally safe for use at recommended dose. But small overdoses may lead to fatal liver damage **(Yang *et al*., 2009)**. PAM has emerged as an important wastewater contaminant due to its wide usage in pharmaceutical industries and daily life. The clinical trade name of PAM is acetaminophen (ACTP) or acetyl para-aminophenol (APAP). **Zhang *et al*. (2015a)** reported the bio-electrochemical degradation of paracetamol in a microbial fuel cell-Fenton system. Dual-chamber MFC reactors were used. For bio-electrochemical degradation of PAM, Fenton reactions were introduced to microbial fuel cells (MFCs) without external power supply. No continuous addition of Fenton reagents was required in MFC-Fenton system compared to conventional Fenton reactions system. Maximum PAM degradation efficiency of 70 % was found within 9 hr at initial pH 2.0, external resistance 20 X and total iron concentration of 5 mg l^{-1}. PAM (25 %) could be completely mineralized and the majority was mainly converted to intermediate metabolites of p-nitrophenol via p-aminophenol and dicarboxylic/carboxylic acids which is less toxic. These results suggested that microbial fuel cell-Fenton system can be applied as an energy-saving and efficient tool for the remediation of PAM-containing wastewater.

Phenolic compounds

Phenols and phenolic compounds are being used in the petrochemical, oil refining, plastic and pharmaceutical industries. Exposure to phenol and its derivatives may lead to respiratory disorders, and long-term phenol inhalation may even lead to cancer **(Calabrese and Kenyon, 1991)**. Disposal of these compounds to water bodies has become a great concern due to their toxicity and harmful effects to human and aquatic lives **(Chen *et al*., 2013)**. The World Health Organization (WHO) indicated substituted phenols as most toxic substances for human health.

2,4-dichlorophenol (2,4-DCP) is the most recalcitrant and hazardous contaminant found in industrial wastewater. The maximum permissible limit for 2,4-dichlorophenol (2,4-DCP) is 40 µg l^{-1} **(Davi and Gnadi 1999)**. **Hassan *et al*. (2016a)** used pure culture *B. subtilis* for degradation of recalcitrant toxic compounds through MFC systems. **Hassan *et al*. (2016b)** explored the degradation of 2,4-DCP in a double chamber microbial fuel cell (MFC) using the bacteria *Bacillus subtilis.* MFC system enabled the removal of contaminants by the bacteria with concurrent electricity generation through electron transfer mechanisms. The exoelectrogenic bacterium *B. subtilis* was found to show good capability for generating maximal power density of 9.5 mW m^{-2} with a peak current 1.11 mA over a potential 0.45 V, while degrading over 60 % 2,4-dichlorophenol. The change of pH and chemical properties of potassium persulfate as catholytes showed significant impact on bio-electrochemical activities. Based on the results, the *B. subtilis*-catalysed MFC was reported as feasible technology for remediation of toxic phenol pollutants from industrial wastewater.

Hedbavna *et al*. (2016) reported the biodegradation of phenolic compounds and their metabolites in contaminated groundwater using microbial fuel cells. This was the first report on application of bioelectrochemical systems (BESs) on biodegradation of phenolic compounds and their organic metabolites in contaminated groundwater. The anaerobic biodegradation of phenol occurred via 4-hydroxybenzoic acid and 4-hydroxy-3-methylbenzoic acid in the anode chamber. The electrode along with sulphate, nitrate, oxygen, iron (III) were electron acceptors for biodegradation. The electricity (~1.8 mW m^{-2}) was produced by electro-active bacteria attached to the anode, while acetate was utilized as an electron donor. Removal of acetate was enhanced by 40 % in the presence of the anode. However, enhanced removal of phenols occurred only for a short time.

A microbial fuel cell (MFC), with graphite electrodes as both the anode and cathode, was reported by **Zhang *et al*. (2017b)** for phenol degradation. It was operated with a soil-free anaerobic consortium. High efficiency with a current density of 120 mA m^{-2} and a coulombic efficiency of 22.7 % were shown by this phenol-degrading MFC. The phenol-degrading MFC was maintained by the microorganisms in the anode biofilm, not by the planktonic bacteria. The structure of microbial community of the anode biofilm and the planktonic bacteria was analysed based on 16S rRNA gene sequences. *Geobacter* sp. was suggested the phenol degrader in the anode biofilm which was responsible for current generation.

Application of dual chamber microbial fuel cell (MFC) operated at fed batch mode was reported for the treatment of coconut husk retting wastewater containing phenol at a concentration of 320 ± 60 g m^{-3} (**Jayashree *et al*., 2014**). Hydraulic retention time (HRT) of the reactor was varied from 40 days to 10 days. Highest phenol removal of 93 % at 40 days HRT of the microbial fuel cell was noted. The bacterial strains viz. *Ochrobactrum* sp. RA1 (KJ408266), *Ochrobactrum* sp. RA2 (KJ408267) and *Pseudomonas aeruginosa* RA3 (KJ408268 in anode region were reported to be responsible for potential phenol removal.

Efficient strategies for faster start up and quick pentachlorophenol (PCP) degradation were explored using of MFC **(Huang *et al*., 2013a)**. There were reports on degradation of recalcitrant (PCP) in both the bioanodes and the biocathodes of MFCs **(Huang *et al*., 2012)** but the start up time was very long and PCP degradation was very slow. In bioelectrochemical cells (BECs), development of biocathodes at −300 mV vs. SHE (versus standard hydrogen electrode) and bioanodes formed at an optimal potential of 200 mV vs. SHE enhanced the performances of MFCs compared to the un-treated controls. PCP degradation rates were improved by 21.5 % in biocathodes and 28.5 % in bioanodes. The start-up times were reduced to 320 h in bioanodes and 420–440 h in biocathodes. The bacteria viz. *Desulfovibrio marrakechensis*, *Comamonas testosteroni* and *Comamonas* sp. on the biocathodes at −300 mV and *Desulfovibrio carbinoliphilus* and *Dechlorospirillum* sp. on the bioanodes at 200 mV were included as recalcitrant organic degraders and/or exoelectrogens/electrotrophs. This was found to be a feasible approach to develop both bioanodes and biocathodes for efficient PCP degradation.

Mineralization of 4-chlorophenol (4-CP) in microbial fuel cells was reported by **Huang *et al*. (2013b)**. The 4-CP is found in industrial wastewaters, soils, surface waters and ground water **(Field and Sierra-Alvarez, 2008)**. It was co-metabolically degraded and mineralized in presence of glucose in microbial fuel cells (MFCs). The degradation rate was 0.58 ± 0.036 mg l^{-1} h^{-1} (7.2 ± 0.5 mg g^{-1} VSS-h).

p-Nitrophenol (PNP) is an important nitro-aromatic compound which is used in manufacturing of dyes, explosives, fungicides, industrial solvent and organic phosphorus pesticide **(Kuosa and Kallas, 2009).** PNP is recognized as list I toxic pollutant by the European Economic Community and a priority pollutant by the US Environmental Protection Agency. The permissible limit of PNP in natural water is restricted to less than 10 mg l^{-1} for environmental safety **(Zhang *et al*., 2009).** Degradation of p-nitrophenol (PNP) was monitored in a two-chambered microbial fuel cell (MFC) system (Liu *et al*., 2013). The results showed that maximum PNP degradation was 64.69 % when PNP was used as a sole substrate.

Electrosorption driven microbial fuel cells was used for the removal of phenol without external power supply **(Yang *et al*., 2013)**. During MFC sorption, the operating parameters viz. pH, electrolyte concentration, initial phenol concentration, MFCs connection numbers and mode were studied along with adsorption isotherms and kinetics of a novel electrosorption driven by microbial fuel cells. Maximum adsorption capacity by MFC-sorption and electrosorption was observed 48 % and 65 % higher than that by conventional adsorption. The initial adsorption rate of phenol was increased 26.99-fold with an increase of initial phenol concentration from 20 mg l^{-1} to 300 mg l^{-1}.

Synthetic dyes

Synthetic dyes released from printing, textile, leather and other industries are considered as recalcitrant pollutants containing complex structure which cannot be degraded easily **(Devi *et al*., 2009).** Azo dye is very widely used in dye industry but it has been found to be highly toxic and mutagenic **(Bafana *et al*., 2011)**. MFC has shown the ability to decolorize azo dye, a widespread environmental pollutant associated with dye manufacturing industry, while simultaneously generating bioelectricity **(Sun *et al*., 2011).**

Recently, application of microbial fuel cell (MFC) with redox mediator modified anode was reported for enhancing simultaneous decolorization of azo dye and electricity generation **(Huang *et al*., 2017)**. Redox mediator (RM) modified anodes were made by electrodepositing riboflavin (RF) and humic acid (HA) on the surface of graphite felt. Air-cathode single-chamber MFCs with different modified anodes were built for decolorization of Congo red and generation of bioelectricity. Compared to the bare anode MFC, MFC with 0.5C RF, 0.5C HA, 1.25C RF and 2.5C HA modified anode exhibited excellent electrocatalysis

activity. MFCs with 2.5C HA and 1.25C RF modified anode showed high decolorization efficiency of Congo red to 86 % and 75 % in 16 h. Significant RM crystals on the modified anodes, and bacterial colonies on the anode surface were noted after MFC running. The RM crystals on the modified anodes could accelerate electron transfer for decolorization and bioelectricity generation both.

Novel bioelectro-Fenton technology was applied for azo dye wastewater treatment using microbial reverse-electrodialysis electrolysis cell **(Hou *et al.*, 2017)**. The single-chamber microbial electrolysis cell was constructed with a TiO_2-coated photocathode termed as photocatalytic microbial electrolysis cell (PMEC). The PMEC was developed to accelerate methyl orange (MO) degradation and concurrent hydrogen (H_2) recovery under UV irradiation. Faster MO decolorization rates were achieved from the PMEC compared with those without UV irradiation or with open circuit. With increase of MO concentrations ranging from 50 to 300 mg l^{-1} at an applied voltage of 0.8 V, decolorization efficiencies was found to decrease from 98 % to 76 % within 12 h, and cyclic H_2 production was reduced from 113 to 68 ml. Bioelectrochemical reduction, co-metabolism reduction and photocatalysis were involved as the possible mechanism of MO degradation. The intermediates of MO mainly sulfanilic acid and N, N-dimethylaniline were further degraded by ·OH generated from photocatalysis. This could make MO mineralization possible in the single-chamber PMEC.

An innovative concept of using microbial reverse-electrodialysis electrolysis cell (MREC) based Fenton process was proposed for the treatment of azo dye Orange G bearing wastewater **(Li *et al.*, 2017).** In this integrated process, the production of H_2O_2 was key reactant of Fenton-reaction driven by the electrons harvested from the exoelectrogens and salinity-gradient between sea water and fresh water in MREC. Complete decolorization and mineralization of Orange G at concentration of 400 mg l^{-1} was achieved.

Oon *et al*. (2017) reported the application of microbial fuel cell (MFC) using monoazo and diazo dyes as terminal electron acceptor for simultaneous decolourisation and bioelectricity generation. Monoazo and diazo dyes [N Acid orange 7 (AO7), New coccine (NC), Reactive green 19 (RG19) and Reactive red 120 (RR120)] were chosen as electron acceptors in the abiotic cathode of MFC. When NC was employed as the electron acceptor, the chemical oxygen demand (COD) removal and dye decolourisation efficiencies obtained at the anodic and cathodic chamber were 73 ± 3 % and 95.1 ±1.1 %, respectively. The results showed that the rates of decolourisation of monoazo dyes were ∼50 % higher than diazodyes.

A stacked microbial fuel cell-biofilm electrode reactor (BER) coupled system was applied for the enhanced degradation of azo dye, Reactive Brilliant Red X-3B **(Cao *et al*., 2017).** Electrical energy generated by the MFC degraded the azo dye in the BER in this system without the need for an external power supply. The effluent from the BER was used as the inflow for the MFC with further degradation. The results showed that removal of X-3B was 29.87 % higher using this coupled system than in a control group.

Sun *et al*. (2016) reported the performance of biocathode microbial fuel cell with respect to azo dye degradation using two anodic inoculums, textile dyeing sludge (MFC-I) and municipal sludge (MFC-M). The results showed that the MFC-I exhibited excellent performance in Congo red decolorization, whereas the MFC-M performed well in electricity generation. The MFC-I achieved fast Congo red decolorization. The MFC-M exhibited 3.22 times higher power output (29 mW m^{-2} vs. 9 mW m^{-2}) and 38.0 % lower anode impedance (749 U vs. 1208 U) compared to MFC-I. The stains isolated from anodic biofilm of MFC-I were identified as *Pseudomonas* sp. and *Aquamicrobium* sp. whereas the isolates from MFC-M belonged to *Pseudomonas* sp. and *Bacillus* sp. These four selected isolates were proved to be exoelectrogens and simultaneous decolorizer which exhibited different dye decolorizing potentialities and bioelectrocatalytic activities.

Microbial fuel cell coupled constructed wetland (CW-MFC) was used for azo dye Methyl Orange (MO) decolorization and further degradation (**Fang *et al*., 2016**). The N, N-Dimethyl-p-phenylenediamine (DMPD), 1,4-Benzenediamine, N-methyl- and p-Phenylenediamine (PPD) were intermediate products formed during MO decolorization. The decolorization rate of MO in closed and open-circuit CW-MFC were and 87.60 % and 75.59 % respectively. The degradation rate of DMPD in the anode layer of closed and open-circuit CW-MFC were 96.33 % and 84.96 % respectively. In the anode layer, further degradation of azo dye degradation products was possible using suitable anode materials. But in the cathode layer, close current promoted further degradation of the decolorization products.

Nordin *et al*. (2017) developed a novel hybrid system composed of a photocatalytic fuel cell (PFC) and Fenton reactor for degradation of azo dye Reactive Black 5(RB5) and generate electricity. Compared to previously reported bioelectro-Fenton system, microbial fuel cell (MFC) system has significant challenge towards the development and operation system. Instead of MFC, PFC was used to generate electrons for the Fenton system. In the PFC reactor hybrid system, higher degradation efficiency of RB5 was observed.

The first-attempt was made by **Hong *et al.* (2016)** on optimal operation mode(s) of microbial fuel cells (MFCs)-assisted decontamination of azo dye, Reactive Blue 160. Internal resistances could be significantly reduced for effective electron transfer (ET) to dye decolorization (DD) and bioelectricity generation (BG) with supplementation of energy substrate(s) and textile dye(s). The results showed that increase in nutrient substrates and azo dye- reactive blue 160 favoured dye decolourization (DD) in MFC systems.

A highly efficient single chambered up-flow membrane-less microbial fuel cell was used for the treatment of azo dye Acid Orange 7-containing wastewater **(Thung *et al*., 2015)**. Single chambered up-flow membrane-less microbial fuel cell (UFML MFC) was developed to study the decolorization of Acid Orange 7(AO7) and electricity generation simultaneously. The performance of UFML MFC was evaluated in terms of voltage output, chemical oxygen demand (COD) and color removal efficiency by varying the concentration of AO7 in synthetic wastewater. The results showed the voltage generation and COD removal efficiency decreased along with the increase of initial AO7 concentration which indicated that there was electron competition between anode and azo dye. The breakdown of the azo bond and naphthalene compound in AO7 were confirmed based on UV–visible spectra analysis.

A single-chamber microbial fuel cell (MFC) with an aeration electrochemical system was proposed for decolorization of a typical azo dye, Methyl orange (MO) which is a well-known recalcitrant pollutant in dye wastewater **(Zhang *et al*., 2015b)**. The color removal observed was 90.4 % within 360 min with voltage across the aeration electrolytic reactor fixed at 700 mV. The GC–MS analysis indicated the degradation of MO, with generation of benzene derivatives, low molecular weight compounds. The indirect electrochemical oxidation of MO by generated H_2O_2 was mainly responsible for MO decolorization.

Hsueh *et al*. (2014) reported the reductive biodegradation of reactive green 19 (RG19) in *Enterobacter cancerogenus* BYm30 in single chamber membrane-less microbial fuel cell (SCML-MFC) and non-MFC cultures. The results indicated that menaquinone from BYm30 might have played a role as an electron-shuttling carrier or a redox mediator for stimulation of dye decolorization and bioelectricity generation in MFC. Color removal in MFC was significantly faster than non-MFC cultures due to the formation of reduced organic sulfides from RG19 in MFC. Moreover, tertiary alcohols and phenols which were present in LB broth might have degraded by BYm30 under anaerobic condition irrespective of MFC or non-MFC condition.

Kalleary *et al*. (2014) highlighted the application potential of microbial desalination Cell (MDC) for effective biodecolourisation of industrial dyes. This was the first report where dye house effluent was used as an organic substrate in MDC and biodecolourisation of effluent along with considerable desalination and power production was achieved. Two novel bacterial strains, viz. *Bacillus subtilis* moh3 and a MTCC strain *Aeromonas hydrophila* subsp. *hydrophila* 8049 were used for decolourisation of two model dyes-Malachite Green (C.I.42,000) and Sunset Yellow (C.I.15,985). Complete biodecolourisation along with considerable desalination (62.2 ± 0.4 % and 57.6 ± 0.2 %) and power production were seen using these novel cultures.

Bacterial–fungal interactions can enhance the power generation in microbial fuel cells and drive dye decolourisation of dyes **(de Dios *et al*., 2013)**. The potential for sustainable energy production from wastes was tested using a combination fungus–bacterium in microbial fuel cell (MFC) and electro-Fenton technology. The fungus *Trametes versicolor* was grown with *Shewanella oneidensis* so that the bacterium could use the networks of the fungus for the transport of electrons to the anode. Stable electricity was generated with stable voltage of 1000 mV approximately. Due to the dual benefits of the in situ-designed MFC electro-Fenton, the simultaneous dye decolourisation and the electricity generation were demonstrated. This newly developed MFC fungus–bacterium with an in situ electro-Fenton system could ensure a high-power output and a continuous degradation of organic pollutants.

A microbial fuel cell coupled constructed wetland (CW) (planted with *Ipomoea aquatica*) system was used for azo dye decolorization **(Fang *et al*., 2013)**. Electricity was simultaneously produced during the co-metabolism process of glucose and azo dye. The results showed that plants grown in cathode could enhance the cathode potential and promoted dye decolorization. The electrodes promoted the dye decolorization efficiency in the anode. The planted CW–MFC system achieved highest decolorization rate of 91.24 % and a voltage output of 610 mV.

Enhanced simultaneous decolorization of Congo red and bioelectricity generation with anthraquinone-2,6-disulphonic disodium salt (AQDS), riboflavin (RF) and humic acid (HA) as mediators in air-cathode microbial fuel cell (MFC) was demonstrated **(Sun *et al*., 2013).** The MFC with added 0.005 mmol l^{-1} AQDS, 0.005 m mol l^{-1} RF or 1 g l^{-1} HA showed 36 %, 26 % and 15 % increase in maximum power density along with 394 %, 450 %, and 258 % increases in decolorization rates of Congo red, respectively compared with mediator-free MFC. At higher concentration, addition of mediators further increased power and Congo red decolorization. Microbial analysis showed that addition of mediators changed the composition of anodic microbial community and stimulated the growth of species belonging to *Chlorobi*, *Endomicrobia* and *Firmicutes*.

Xu *et al*. (2013) investigated the functions and interactions of predominant microorganisms in microbial fuel cells (MFCs) for simultaneous electricity generation and Congo red decolorization. Four strains isolated from the anodic biofilm, and identified as *Pseudomonas* (M-P and I-P), *Bacillus* (M-B) and *Aquamicrobium* (I-A). Using pure cultures, I-P and M-P as inoculums, lower Congo red decolorization rate (by 3.2 % and 5.9 %) and higher maximum power

density (by 158.2 % and 58.1 %) were achieved in MFCs than those using I-A and M-B, respectively. Using co-cultures (MP and B versus M-B and M-P, I-P&A versus I-A and I-P), the maximum power density of MFCs were increased 82.0 %, 15.1 %, 94.6 % and 24.6 % but decolorization rate decreased 33.3 %, 29.4 %, 7.9 % and 5.0 % respectively. The results indicated that specific interaction could enhance the performance of MFCs and which may be beneficial for the development of bio-process controlling.

Nitrogen based compounds

Among various global environmental pollutions, nitrogen-containing organic compounds have drawn extensive attention as recalcitrant pollutants in the last few decades. They usually contain compounds like aromatic rings or azo bond having complex structure that may cause harmful and irreversible environmental problems even at quite low concentrations **(Devi *et al*., 2009).**

Wang *et al*. (2015a) proposed a promising single-chamber microbial fuel cells (MFCs) as renewable power sources, an aerated electrochemical system for removal of nitrogen-containing organic compounds (pyridine and methyl orange). Lower initial contaminant concentration and lower initial pH value could improve the performance of MFC. A degradation efficiency of 82.9 % for pyridine was achieved after 360 min electrolysis at initial pH 3.0, initial concentration 200 mg l^{-1}, and applied voltage 700 mV. Indirect electrochemical oxidation by generated hydrogen peroxide was responsible for their degradation.

Nitrogen removal in continuous flow microbial fuel cell (MFC) and microbial electrolysis cell (MEC) was reported under laboratory condition **(Hussain *et al*., 2016)**. An ammonium removal efficiency of 30-55 % with near-zero nitrite and nitrate concentrations was observed in the MFC operated at an optimal external resistance. An open-circuit MFC operation resulted reduced ammonium removal of 21 %. In the MEC, removal of ammonium was limited to 7-12 % under anaerobic conditions. Removal efficiency was increased to 31 % under micro-aerobic conditions. Ammonium removal efficiency was reduced to 4 % at zero applied voltage.

During last few decades, nitrate pollution in groundwater has become a serious issue in most parts of the world due to discharge of domestic, industrial wastewater and increased usage of nitrogenous fertilizers **(Park *et al*., 2005)**. Excessive nitrate can do harm to humans and animals. causing methemoglobinemia or blue baby syndrome in infants and gastrointestinal cancer in adults **(Mousavi *et al*., 2012)**. Thus, the maximum contaminant levels (MCL) are stipulated to be 10 mg l^{-1} nitrate nitrogen (NO_3^-–N) and 1 mg l^{-1} nitrite nitrogen (NO_2^-–N) respectively by both USEPA and China.

Zhang *et al*. (2014) demonstrated the enhancement of bacterial denitrification for nitrate removal in groundwater with electrical stimulation from microbial fuel cells. Electricity generated from the microbial fuel cell (MFC) was applied to the bioelectrical reactor (BER) directly as electrical stimulation means for the enhancement of bacterial denitrification to remove nitrate effectively from groundwater. With maximum power density of 502.5 mW m^{-2} and voltage outputs ranging from 500 mV to 700 mV, the nitrate removal was accelerated with less intermediates accumulation, compared with control sets without electrical stimulation. Denitrification bacteria proliferations and activities were promoted as its number and Adenosine-5'-triphosphate (ATP) concentration increased 1.5 folds in one order of magnitude (3.5×10^7 in per millilitre biofilm solution). Effects of electricity from MFCs on enhancement of bacterial behaviours were reported for the first time.

Ethyl Acetate (EA)

Ethyl acetate (EA), a VOC, is an organic solvent which is highly volatile and damaging to the respiratory system **(Coopman *et al*., 2005)**. It is commonly used as a solvent in varnish, adhesives, paint, and organic syntheses, especially food and pharmaceutical industries **(Kleinbeck *et al*., 2008)**. Exhaust emissions of VOCs from various industrial sources cause environmental damage and human health hazard **(Mohseni and Allen, 2000)**. Prolonged exposure to EA irritates human eyes. High concentration of EA steam may cause health hazards such as headache and fatigue **(Wu and Lin, 2016).**

Wu *et al*. (2016a) developed an innovative biotrickling filter-microbial fuel cell (BF-MFC) for the use in electricity production and removal of ethyl acetate (EA) emitted from a gaseous stream. A new membrane design was developed which provided effective delivery of protons released from microorganisms biodegrading EA from the anode to the cathode using a polyvinyl alcohol-membrane electrode assembly. The elimination capacity (EC) reached almost the 100 % as the EA concentration was increased from 0.18 g m^{-3} to 1.44 g m^{-3}, along with the increase of the voltage increased from 49.4 mV to 658 mV, and EA organic loading rate ranged from 14.41 to 29.58 g m^{-3} h^{-1}. Microbial community analysis revealed that two distinct groups of exoelectrogenic microbes and EA-degraders were mainly involved in the conductive coke surface and in the inner tube wall of the BF-MFC, respectively.

The modification of membrane anode was done in the microbial fuel cell to improve the removal of gaseous ethyl acetate without reducing generation of electricity **(Wu *et al*., 2016b)**. An S-type flow-field pipe-microbial fuel cell (SFP-MFC) was developed to remove ethyl acetate (EA) from an air stream. The SFP-MFC included the use of polyvinyl alcohol and a membrane electrode assembly (PVA-MEA) as the gas diffusion membrane and proton exchange membrane (PEM) which could separate anode from the cathode. The performance of the SFP-MFC system was evaluated with an empty bed residence time (EBRT) of 14.35 s and an organic loading rate of 63−3700 g m^{-3} h^{-1}, with or without the modification of the PVA-MEA electrode using conductive carbon black (CCB). The maximum elimination capacity (EC) and voltage were 2288 g m^{-3} $h^{-1}$and 330 mV, respectively, which were obtained when the PVA-MEA was modified using CCB. The PVA-MEA that was modified with CCB which exhibited a 90 % higher EA elimination capacity than the PVA-MEA without CCB modification. The maximum EC and EBRT of the modified SFP-MFC were 3−10 times higher and 2−12 times shorter, respectively, than those of biotrickling filter.

Toluene

Toluene is a common solvent and used as a raw material for the production of xylene and benzene. Global consumption of toluene has now reached 2×10^7 tons annually **(Morata *et al*., 1995).** Toluene is a toxic, refractory and mutagenic pollutant **(Asenjo *et al*., 2011).**

Wu *et al*. (2014) tested the toluene degradation and electricity generation under varying toluene doses. Pyocyanin was added to evaluate the effectiveness in improving electricity generation via toluene degradation. Addition of pyocyanin significantly improved electricity generation by reducing system impedance, increasing electron density and substantially lowering the internal resistance of microbial fuel cell (from 500 to 100 V). The results of this study demonstrated the possibility of adding pyocyanin for enhanced power generation in a toluene-fed MFC which can be used for the treatment of xenobiotics-contaminated wastewater like toluene.

A new anode design allowing direct delivery of the gaseous substrate to the biofilm was developed by spraying an additional hydrophobic gas layer and PTFE layers onto the gas side of the anode **(Li *et al*., 2013c)**. The MFC with this design exhibited a two-fold increase in the toluene removal efficiency (from 43.1 ± 2.7 to 91.2 ± 2.4 %) and an approximately eight-fold increase in the maximum power density (from 0.72 ± 0.02 to 6.19 ± 0.45 mW m^{-2}) compared to the MFC equipped with a sparger for toluene delivery. MFC also showed a maximum toluene elimination capacity of 274.5 ± 14.4 g m^{-3} h^{-1}, which was higher than the values usually reported for biofiltration systems and biotrickling filters.

Polycyclic aromatic hydrocarbons (PAHs)

Pollution of groundwater by polycyclic aromatic hydrocarbons **(**PAHs) is a serious threat to human health as the hydrocarbons are toxic, mutagenic and carcinogenic. MFCs could be employed in the treatment of these recalcitrant pollutants with concomitant bioelectricity generation **(Morris and Jin, 2012).**

The treatment of a mixture of phenanthrene and benzene using two different tubular microbial fuel cells (MFCs) were designed for either in situ and ex situ applications in aqueous systems over long periods (up to 155 days) **(Adelaja *et al*., 2017).** Simultaneous removal of the petroleum hydrocarbons (>90 % in term of degradation efficiency) and bromate, used as catholyte (up to 79 %) with concomitant biogenic electricity generation (peak power density up to 6.75 mW m^{-2}) were obtained at a hydraulic retention time (HRT) of 10 days for in situ studies. In the MFC designed for ex situ studies, optimum MFC performance was obtained at HRT of 30 h showing COD removal and maximum power output of approximately 77 % and 6.75 mW m^{-2} respectively. The results of this study suggested the potential use of MFC technology for in situ/ex situ petroleum hydrocarbon-contaminated groundwater treatment even at extreme toxic level conditions.

A sediment microbial fuel cell (SMFC) was explored to bioremediate polycyclic aromatic hydrocarbons (PAHs) in water originated from soil. **Sherafatmand and Ng (2015)** reported the ability of SMFCs in stimulating microorganisms for bioremediation of complex and recalcitrant PAHs. The results showed consistent power generations of 6.02 ± 0.34 and 3.63 ± 0.37 mW m^{-2} under an external resistance of 1500 Ω by the aerobic and anaerobic SMFC respectively. Although the power generations were low, the significant benefit of this system was its bioremediation capabilities, achieving removal of naphthalene (41.7 %), acenaphthene (31.4 %) and phenanthrene (36.2 %) respectively in the aerobic environment and respectively, In the anaerobic environment, the removal was 76.9 % for naphthalene, 52.5 % for acenaphthene and 36.8 % for phenanthrene was noted.

The use of MFCs in biodegradation of phenanthrene was investigated with respect to its biodegradation rate, biodegradation efficiency and power production using different inocula (*Shewanella oneidensis* MR1 14063, *Pseudomonas aeruginosa* NCTC 10662, mixed cultures, and combinations) **(Adelaja *et al*., 2014)**. All the inocula showed high capacity for phenanthrene degradation with a minimum degradation efficiency of 97 %. The best overall performing inoculum was anaerobically digested sludge supplemented with *P. aeruginosa* NCTC 10662, having a degradation rate, maximum power density

and COD removal efficiency of 27.30 μmol l^{-1} d^{-1}, 1.25 mW m^{-2} and 65.6 %, respectively.

Pesticide

Hexachlorobenzene (HCB), as refractory organic pesticide, is toxic to human beings and environment. A range of remedial techniques were developed to remove toxic refractory organics in soil. The soil microbial fuel cells (MFCs) were constructed in the topsoil contaminated with toxic refractory organic pesticide, hexachlorobenzene (HCB). The performance of electricity generation and HCB degradation in the soil-MFCs were investigated **(Cao *et al*., 2015).** The HCB removal efficiencies in the three groups viz. (i) soil MFCs group (ii) open circuit control group and (iii) no adding anaerobic sludge blank group were 71.15 %, 52.49 % and 38.92 %, respectively. The highest detected power density was 77.5 mW m^{-2} at the external resistance of 1000 Ω. HCB was degraded via the reductive dechlorination pathway in the soil MFC under anaerobic condition. The existence of the anode promoted electrogenic bacteria provided more electrons which could promote the removal efficiencies of HCB in soil MFC.

Perchlorate

Perchlorate ($ClO4^{-}$) is a major inorganic pollutant which is widely used in the manufacture of automobile airbags, missiles, road flares, rocket propellants, fireworks etc. and ultimately enters water resources **(Xu *et al*., 2015)**. Presence of perchlorate has been observed in bottled water, groundwater, vegetables, rice, milk. It shows direct effect on the uptake of iodine by the thyroid gland **(Wang *et al*., 2014)**. So, the removal of perchlorate from wastewater has become an important environmental issue now days. There are few studies on the use of MFCs to treat perchlorate.

Lian *et al*. (2016) reported that resazurin as mediator could improve the performance of microbial fuel cells (MFCs) for the removal of perchlorate. MFCs with 3 µmol l^{-1} resazurin, 6 µmol l^{-1} resazurin, and 9 µmol l^{-1} resazurin showed increases in perchlorate reduction ratio of 50.8 %, 72.6 % and 101.6 %, respectively, compared with a mediator-free MFC. Increases in the output voltage of 24.5 %, 33.3 % and 41.7 %, respectively was also noted. Addition of resazurin decreased the anode resistance and enhanced the biocatalytic activities of the anode. Addition of resazurin changed the composition of the anodic microbial community which stimulated the growth of certain dominant microbes for the removal of perchlorate.

The effects of different electron donors and electron acceptors on the performance of microbial fuel cell (MFC) for perchlorate removal were investigated **(Lian *et al*., 2017)**. The results of microbial analysis showed that the electron acceptors did not change the composition of the anodic microbial community. Dominant species were responsible for the enhancement of the higher perchlorate reduction rate and the higher output voltage.

Sulfur

Sulfur species at high concentration are harmful to living organisms. These sulfur pollutants are present in many processes such as pulp and leather, animal husbandry, detergent manufacture, mining, dye and food processing **(Lens and Pol, 2000)**. Most of dissolved sulfur species flow into sea with the surface runoff. Sulfur pollutants negatively affect the natural ecosystems. Microbial fuel cells (MFCs) have been used as a promising technique for the removal of sulfur pollutants in wastewater. **Fang-Yuan *et al*. (2013)** described the roles of electrode chemical reaction and involvement of microorganisms in sulfur pollutants treatment using MFCs. The effecting factors and mechanism of treatment were demonstrated. The separator types, electrode materials and catalysts, sulfur recovery and electrode regeneration were also demonstrated. In addition, the feasibility of sulfur pollutants removal in MFCs was assessed by capital cost assessment.

Emerging contaminants (ECs)

The discharge of emerging contaminants (ECs) into the aquatic environment has become a great concern now-a-days due to their negative effects on the ecosystem. A number of endocrine disruptors and antibiotic resistant microorganisms were observed due to long-term contact of EC **(Sun *et al*., 2012)**. Wastewater treatment plants (WWTPs) are the primary sources for the release of ECs into the water environment **(Yu and Chu, 2009)**.

Wang *et al*. (2017) investigated the removal of four ECs viz. Bisphenol A (BPA), Estrone (E1), Sulfamethazine (SM2), Triclocarban (TCC) using the microbial fuel cell (MFC)-Fenton system. Glucose and graphite rod with stacked graphite granules were used as the substrate and electrode, respectively. Both batch and continuous flow modes of MFCs were compared. Successful application of the MFC-Fenton technology for the effective removal of ECs viz. E1, BPA, TCC and SM2 was demonstrated which may provide a cost-effective and promising alternative strategy for ECs removal in future.

Trace organic compounds (TOrCs)

Trace organic compounds (TOrCs) are reported as common contaminants in domestic wastewater resulting from human activities like disinfection by products, personal care products, pesticides and pharmaceuticals **(Ratola *et al*., 2012)**. Removal of TOrCs from wastewater has become a great concern because of their significant impacts on public health **(Coday *et al*., 2014)**.

The removal of 26 trace organic compounds (TOrCs) using single-chamber air-cathode MFC (SMFC) and double-chamber air-cathode MFC (DMFC) microbial fuel cells (MFC) was reported **(Wang *et al*., 2015b).** The results of the study showed that both sorption and biodegradation processes were involved for the removal of TOrCs. The removal efficiency was affected by the hydrophobicity and biodegradability probability of the compounds in case of neutral TOrCs. Electrostatic interactions played an additional role in the MFCs. The removal of positively charged TOrCs was generally higher than negatively charged TOrCs. Negligible impact on MFC power generation was noted in presence of TOrCs.

CONCLUSION

This review mainly summarizes the recent reports on application of MFCs for the remediation of various pollutants. MFC technology can also be targeted towards future application in space technology on account of its power generation ability, sustainable energy generation and biosensor application. Although MFCs have been applied intensively over the past decades, limited success has been noted in practical application due to some limitations and challenges. Therefore, MFC technology has yet to find recognition and commercial success in environmental applications.

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