Biocatalysis and Agricultural Biotechnology 22 (2019) 101370

Contents lists available at ScienceDirect

Biocatalysis and Agricultural Biotechnology

journal homepage: http://www.elsevier.com/locate/bab

A mini review on biofouling on air cathode of single chamber microbial fuel cell; prevention and mitigation strategies

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ARTICLE INFO

Keywords:
Microbial fuel cell
Biofouling
Air cathode
Single chamber
Mitigation
Cleaning

ABSTRACT:

Microbial fuel cells (MFCs) have gained considerable interests due to their potential for bioenergy generation from the biocatalytic conversion of a broad diversity of organic wastes. However, low power generation and high material cost in MFCs posed challenges to modify the system in terms of materials and configurations. Membrane-less design of single chamber microbial fuel cell (SCMFC) with air breathing cathode is one of the most practical configuration for large-scale implementation due to simplicity, low capital cost and needless of aeration energy while producing the high power output. However, the formation of aerobic bacterial biofilm (biofouling) on the surface of catalysts coated air cathode during long-term operation reduced the system performance and increased the internal resistance. Therefore, suitable prevention and mitigation strategies were required to be addressed and applied. This study summarized the strategies that were applied recently and potential to be applied in future to prevent biofouling formation on the air cathode in SCMFC. The article also reviewed the cleaning methods and catalysts regeneration techniques on the air cathode to maintain long-term stability and performance.

1. Introduction

Energy is extremely necessary for world development, which 80% of it, is provided by fossil fuel (Kumar et al., 2019). However, extensive use of fossil fuel negatively affected the environment by the emission of carbon dioxide, causing air pollution, global warming and health problems (Zhao et al., 2018; Zheng et al., 2018). Furthermore, this unsustainable energy source is being depleting (Martins et al., 2018). Water scarcity and high rate of wastewater generation are two other global challenge as a result of population growth, rapid industrialization, urbanization and climate change (El Moussaoui et al., 2019; Zheng et al., 2018). Discharging of wastewater without/with minor treatment can contaminate fresh water sources which negatively affects the environmental and aquatic systems as well as human health (Kumar et al., 2019). Wastewater treatment is an effective solution to overcome discharging problems while being served as an alternative water source for agricultural and industrial demand (Akhoundi and Nazif, 2018). Traditional treatment of wastewater includes biological and physiochemical methods. Using a flocking agent to remove suspension solids by chemical precipitation generates secondary pollutants, which increases the sludge discharging load in the activated sludge process. However, the bulk volume of the generated sludge needs a proper way to be discarded while being an energy intensive process (Xia et al., 2019). Microbial fuel cell (MFC) as an environment friendly, sustainable and energy-effective solution has the potential to be considered to overcome this issue. The MFC is a bio-electrochemical system which offers wastewater treatment while producing clean electrical energy (Al-Mamun et al., 2016; Jafary et al., 2017a). It generates bioelectricity from organic compounds which are stored in the different types of wastes through biocatalytic oxidation and reduction reactions (Al-Mamun et al., 2017b, 2017a). It can also be modified to recover value added products from wastes addressing the global waste to wealth movement (Barua et al., 2019).

Microbial fuel cell and performance challenge: The first observation of bioelectricity production was conducted by Luigi Galvani when he connected frog legs to a metallic conductor in the late 18th century (Jafary et al., 2015). However, MFC was discovered in the 20th century when Potter recorded bioelectricity generation by using biotic anode and abiotic cathode. It was found that the electricity generation was due to reduced redox potential as a result of bacterial growth. The existing form of MFC, which uses the electrode as an electron acceptor or sink and oxidizes wastewater through bio-electrochemically active microorganisms was innovated by Kim et al., (Kim et al., 2004; Slate et al.,

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https://doi.org/10.1016/j.bcab.2019.101370
Received 30 July 2019; Received in revised form 14 September 2019; Accepted 2 October 2019
Available online 3 October 2019

List of abbreviations

AC	activated carbon
AC	alternating current
CE	columbic efficiency
CCP	conductive carbon paint
DMFC	direct methanol fuel cell
DA-SMFC	double anode sediment microbial fuel cell
DCMFC	dual chamber microbial fuel cell
ENR	enrofloxacin
AC^+	forward direction half-wave AC
GDL	gas diffusion layer
MFC	microbial fuel cell
NS	no separator
ORR	oxygen reduction reaction
PTFE	polytetrafluoroethylene
PVA	polyvinyl alcohol
PVDF	polyvinylidene fluoride
Ag-NP	silver nanoparticle
SCMFC	single chamber membrane-less MFC
SSM	stainless steel mesh
SP	With separator
WWTP	wastewater treatment plant

2019).

In a typical MFC, the anaerobic oxidation of organic compounds releases electrons, protons, and carbon dioxide (Lefebvre et al., 2008b, 2009). Electrons pass through the electric circuit to the cathode, while protons pass through a membrane to reach the cathode. In the cathode, oxygen -which is provided externally-is reduced by protons and electrons to form water as shown in equations (1) and (2) (Satar et al., 2018);

$$CH_3COO^- + 4H_2O \rightarrow 2HCO_3^- + 9H^+ + 8e^- \quad (1)$$

$$2O_2 + 8H^+ + 8e^- \rightarrow 4H_2O \quad (2)$$

Bio-cathode MFC is a complete sustainable system, which uses the bacteria with a self-generation ability to catalyze waste oxidation and reduction reaction in the anode and cathode chambers, respectively (Al-Mamun and Baawain, 2015; Jafary et al., 2017b). In terms of wastewater treatment, MFCs came with certain credentials: no external energy requirements for aeration, wider range of operational temperature, low rate of sludge production and low energy input for bio-electrochemical reactions in some cases (Al-Mamun et al., 2018; Palanisamy et al., 2019). However, low power generation in MFC made its application limited and brought up the optimization challenges (Nandy and Kundu, 2018). That's why, many modifications in terms of the membrane, electrode, catalyst and operation modes were needed. The main target of any type of modification was to reduce material, fabrication and operational cost and to increase the overall performance (Lefebvre et al., 2008a; Slate et al., 2019). Configuration is one of the focal points in the MFC, which directly affects the membranes, electrode and electrolyte. Dual chamber microbial fuel cell (DCMFC) was the most widely studied and used MFC configuration as shown in Fig. 1A (Palanisamy et al., 2019).

The two-chamber design consisted of one anodic and one cathodic chamber in which the oxidation and reduction half reactions occur, separated by a membrane (Jafary et al., 2018). Inserting a proton selective membrane between two chambers reduced the diffusion of oxygen to the anode and higher columbic efficiency (CE) as a result. However, using membrane caused accumulation of protons in the anodic chamber which reduced the pH of anolyte and finally deteriorate microorganisms performance as well as power generation (Yang et al., 2016). Moreover, membrane imposed high internal resistance and cost (62.5% of the capital cost) to the MFC while needing external energy input for cathodic aeration (Li et al., 2018; Nandy and Kundu, 2018; Palanisamy et al., 2019).

To overcome the aforementioned challenges by using membrane in DCMFC, air cathode single chamber microbial fuel cell (SCMFC) was introduced as illustrated in Fig. 1B (Massaglia et al., 2019). In the SCMFC, the membrane was eliminated and the air cathode was designed with a gas diffusion layer (GDL) to control the oxygen diffusion from the air towards the cathode, removing external aeration demand. Low capital cost, simplicity and high power generation made SCMFC a feasible design for large scale implementation (Liu et al., 2015). Logan et al. reported 80% increase in power generation (28 in a membrane based MFC vs 146 W/m^2 in a membrane less MFC) in an SCMFC running with domestic wastewater (Liu and Logan, 2004).

In SCMFC, the cathode is in direct contact with the air on one side and the electrolyte on other side (Vogl et al., 2016). Elimination of membrane from SCMFC design drastically reduced its capital cost as

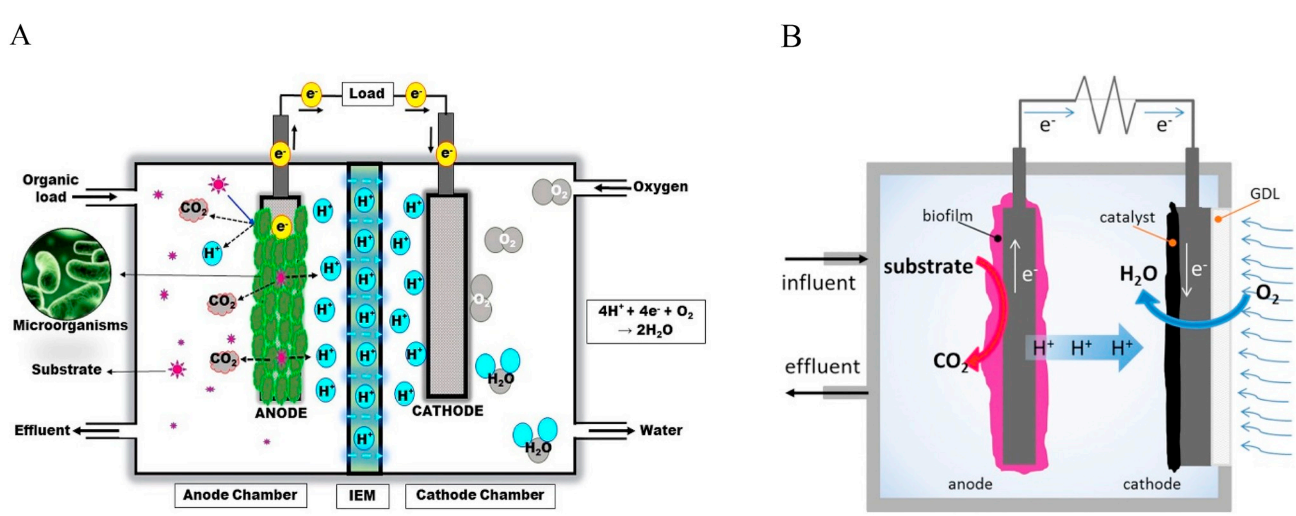

Fig. 1. A) Dual chamber (Palanisamy et al., 2019) and B) air cathode single chamber membrane-less microbial fuel cell (Massaglia et al., 2019), Copyright (2019), with permission from Elsevier.

well as the internal resistance of the bio-chemical system (Vicari et al., 2016). But, it increased the diffusion of organic matters into the cathode matrix that subsequently led to the formation of aerobic biofilm on it. Such formation of biofilm clogged the active sites of catalyst layer coated on the air cathode and consequently deteriorated the MFC performance over a long-term operation (Chen et al., 2019). Since the last decade, a large number of review articles have been published addressing various aspects of MFC design, development of new electrode materials and optimizing the system performance for high power recovery and pollution remediation. However, none of the previous studies reviewed the biofouling mechanisms on air cathode in SCMFC and summarized the mitigation strategies of biofouling. Therefore, the current study aimed to review the biofouling mechanisms and recently used physical and kinetic control strategies to prevent the formation of biofouling on the air cathode in SCMFC. The study also summarized the chemical cleaning and surface modification strategies to mitigate biofouling. At the end, the review identified the research gaps to achieve a sustainable air cathode for large-scale implementation of the SCMFC technology for power recovery and bioremediation. It was noteworthy to mention that salt precipitation (scaling) and local alkalization (inorganic fouling) on air cathode (An et al., 2017) were two other reasons for deteriorating the cathode performance in SCMFCs which were not reviewed in this article.

2. Biofouling

Carbon nature of cathode and its biocompatibility make it as a potential site for microorganisms to attach and colonize. The high potential and hydrophilicity of the catalytic layer of the cathode -which is made to allow proton transportation through the boundary interface-caused the biofilm to grow. Biofouling is defined as the attachment of microorganisms like bacteria, and fungi, more specifically heterotrophic microorganisms in form of biofilm to the surface of cathode (Gule et al., 2015; Liu et al., 2015). Biofilm can be composed of a single strain or mixed culture that can form the synergistic biofilms later on and are more recalcitrant to detach (Logan et al., 2019). Extracellular polymeric substances of biofilm play as a protection layer of sessile cells against any environmental attack (Xu et al., 2017). Microbial influenced

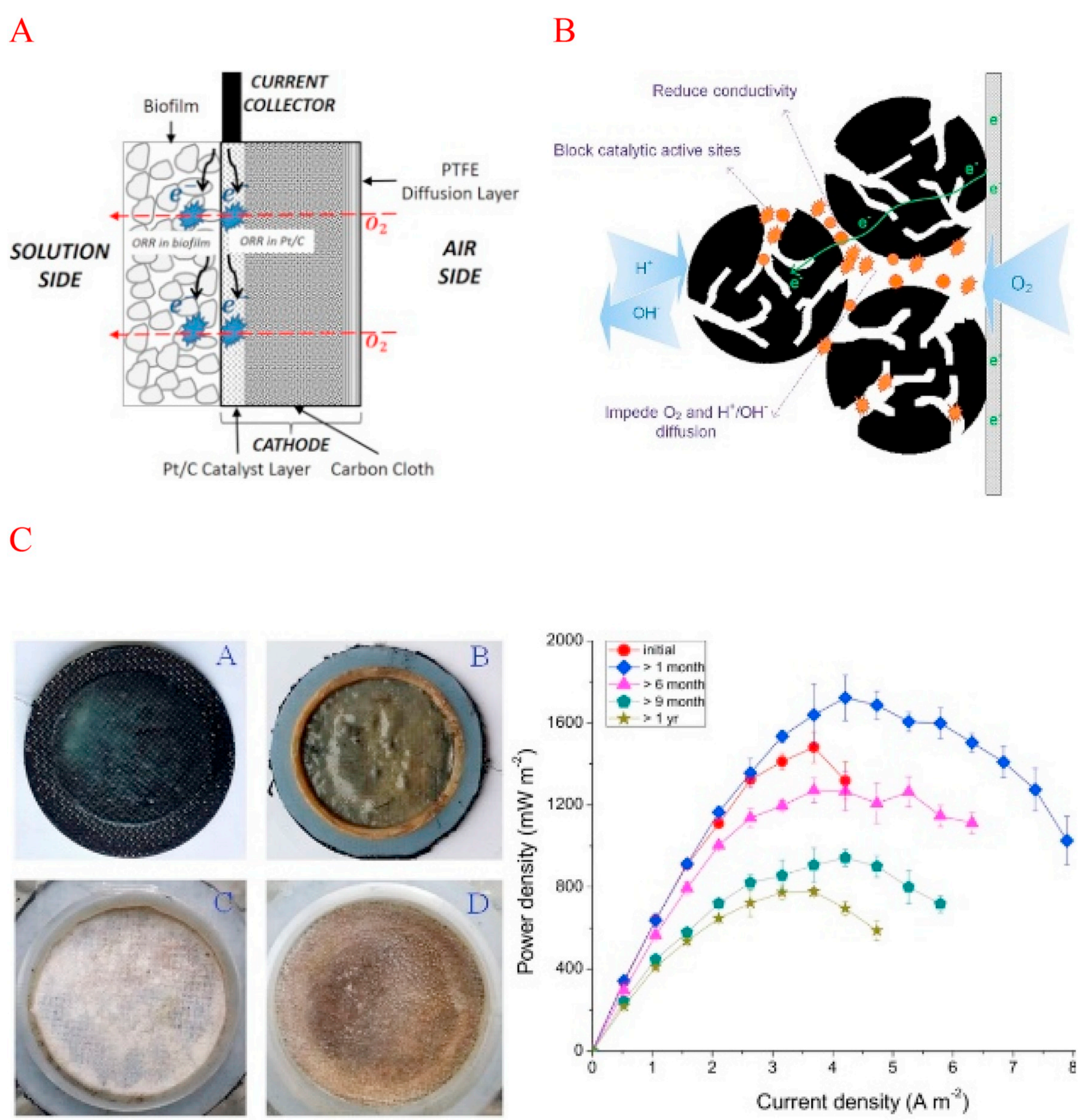

Fig. 2. Cathodic biofouling as a result of biofilm formation on air cathode SCMFC and power reduction over time, A) (Liu et al., 2018), B) (Ou et al., 2016) and C) (Zhang et al., 2017)), Copyright (2019) with permission from Elsevier.

corrosion is one the known example of biofouling as a result of biofilm formation on the surface which imposes a high cost to the oil and gas industry every year (Jia et al., 2019).

By all the aforementioned advantages of SCMFCs, however they are susceptible to biofilm formation on the air-breathing cathode and biofouling as the result. The schematic of cathodic biofouling is shown in Fig. 2 (Ou et al., 2016; Zhang et al., 2017). Eliminating the membrane in the SCMFCs, provides direct contact between the cathode and solution. It forms a layer of aerobic bacteria which blocks the access of protons and charged ions to cathode catalyst sites and also block hydroxide transport out of cathode resulting in a pH imbalance within the electrolyte and catalyst layer (Ma et al., 2015). Aerobic bacterial may also consume some parts of diffused oxygen and reduce the rate of oxygen reduction reaction on the cathode (Call et al., 2017). The extracellular compounds of the biofilm may negatively affect the physiochemical properties of the cathodic catalyst (Liu et al., 2015; Yuan et al., 2016). Biofilm formation on the cathode reduces the active catalytic sites and active area of the cathode which consequently reduces the practical MFC performance (Yang et al., 2018a,b). As the biofilm colonizes and covers the active catalytic sites, the oxygen reduction rate decreases on the cathode and the internal resistance gradually increases. Substrate competition of anodic and cathodic biofilms is another side effect of biofilm formation on the cathode. As a result columbic efficiency drops and power generation reduces (Rossi et al., 2018).

The negative effect of biofilm formation and biofouling as a result of this thick biofilm (ranged between a few μm to a few mm) was reported in different researches (Noori et al., 2019). Exchange current density was reported to decrease by 48% and power generation by 20.3% due to aerobic biofilm formation after two months of operation (Yuan et al., 2016). An et al. (2017) reported a 59% increase in charge transfer resistance caused by biofouling after two months of operation. Another report showed 40% reduction in power density of SCMFC by using carbon as a catalyst after one year of operation (Li et al., 2017). Liu et al. (2015) reported 23% of power decay and 35 mg of biomass formation after 58 cycles on SCMFC (Fig. 3A-B). Yang et al. (2019) in a very recent study showed that the power generation reduced by 52% from 744 to 354 W/m^2 and a thick biomass was formed on the air cathode over just one month of operation (Fig. 3C-D).

Different methods were proposed and studied to control (prevent or mitigate) biofouling in SMFCs. Selecting the suitable method could maintain the MFC performance high and keep the cathodic reaction stable which were discussed accordingly.

3. Biofouling control

Different controlling methods were studied to prevent and mitigate cathodic biofouling in SCMFCs. They consisted of different physical, chemical, electro kinetic and surface modification methods as stated accordingly.

3.1. Physical method

Biofouling can be controlled by physical methods. It includes the cleaning of the surface frequently as well as replacing the cathode, using separator and double anode to reduce the quantity of organic compound in the electrolyte.

As already discussed placing the membrane between the electrodes in a two-chamber design increases MFC cost and the internal resistance of the system and reduces the power generation of the system drastically. However, membrane separator applying on the air side of the cathode is a strategy to reduce biofouling and to maintain the cathodic performance by reducing the oxygen penetration. Vogl et al. (2016)

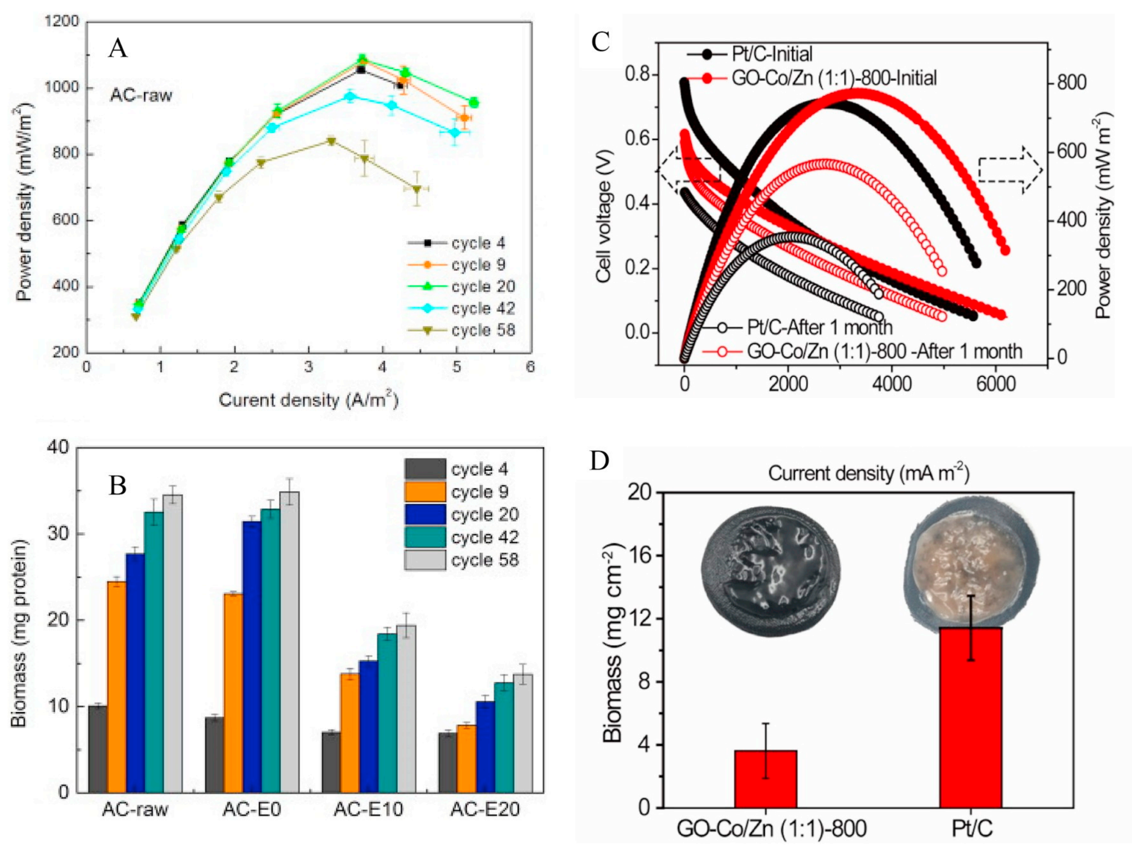

Fig. 3. A) A reduction in power density and B) an increase in biomass formation on the cathode of SCMFC at the beginning and the end of 58 cycles (Liu et al., 2015), C) a reduction in power density and D) an increase in biomass formation on the cathode of SCMFC at the beginning and the end of one month of operation (Yang et al., 2019). Copyright (2019) with permission from Elsevier.

studied the effect of applying two different membranes on the air side of the cathode to control oxygen transfer and aerobic biofilm formation. LDS 2 Silk SK (LORR) and GMM-210 (HORR) with low and high oxygen mass transfer coefficients of 0.6×10^{-3} and 5.9×10^{-3} cm/s were selected for the SCMFCs with 120 and 15 mm distance between the anode and cathode, respectively. In both cases, power outputs were higher when the membranes were applied compared to the control ones (without membranes). However, the MFC with HORR (the higher oxygen transfer and the lower distance) showed higher performance (460 mW/m^2) compared to that of LORR (the lower oxygen transfer and the higher distance) one (254 mW/m^2). A similar trend was shown for cathodic internal resistance in the membrane-protected SCMFCs and the control SCMFCs; 165 Ω for LORR vs 271 Ω for control-LORR, 27 Ω for HORR vs 51 Ω for control-HORR. Moreover, the flexibility of design for replacing the membrane separator over coatings and bound layers was noteworthy. The study proved partial protection of electrodes against biofouling on the expense of using membranes, while the importance of oxygen transfer rate and the electrode distances were also illustrated in this research. Moreover, considering the fact that membrane is exposed to fouling itself, further research on membrane fouling is needed to evaluate the feasibility of this method (Vogl et al., 2016).

A simple double-anode design in a sediment microbial fuel cell (DA-SMFC) was another suggested approach to inhibit colonization of heterotrophic cathodic biofilm (Yang et al., 2018a). The application of two anodes was explained for two reasons. A high electron production rate was observed on two anodes and the diffusion of the organic compound to the cathode was inhibited through physical adsorption of organics to the second anode placed on the sediment water interface. It was also reported that the second anode absorbed and degraded the organics in overlying water which inhibited aerobic biofilm to form on the cathode. The DA-SMFC was compared with SMFC which had the same configuration of DA-SMFC except for the position of anode II as shown in Fig. 4A. The higher cathodic charge transfer resistance (120 Ω) and biofilm protein content after 60 days of operation (0.134 µg/L) in the SMFC than those of DA-SMFC (92 Ω and 0.03 µg/L) was explained by the negative effect of the cathodic biofilm on ORR as shown in Fig. 4B-C.

An innovative SCMFC design was fabricated by Oliot et al. (2016) which allowed replacing the deteriorating air cathode with a new electrode without disturbing the bioanode by exposing it to the air as shown in Fig. 5. While the scope of the study was not cleaning or mitigation of the fouled electrode, the removable cathode design could make the cleaning process more convenient. Moreover, the removable air-cathode design could simplify studying the stages of biofilm formation without disturbing the whole system. Moreover, more studies on removable SCMFC air cathode design are needed to make the cleaning and or regeneration process simpler and more user friendly.

Rossi et al. (2018) applied a novel, cheap and scalable technique using two magnets on both sides of the air cathode to clean the cathode regularly without opening the system. By moving the magnet on the air cathode side, the magnet on the electrolyte side also moved and removed the biofilm on the catalyst side of the cathode.

They have monitored the power production under three different experimental sets at the beginning and end of one month of operation; a) inserting the magnets on the cathode but not moved to clean, b) inserting the magnets and moved daily to clean the cathode and c) not using any magnate. No cloth separator (NS) was used in this experimental sets. The results were then compared with repeating the same experiments when applying the cloth separator (Sp) which was suggested as a biofouling reducing method by (Yang et al., 2018b). The results showed that daily cleaning of the air cathode could result in a 40% higher power generation compared to no biofilm removal status. Moreover, the magnetic cleaning strategy was more successful than

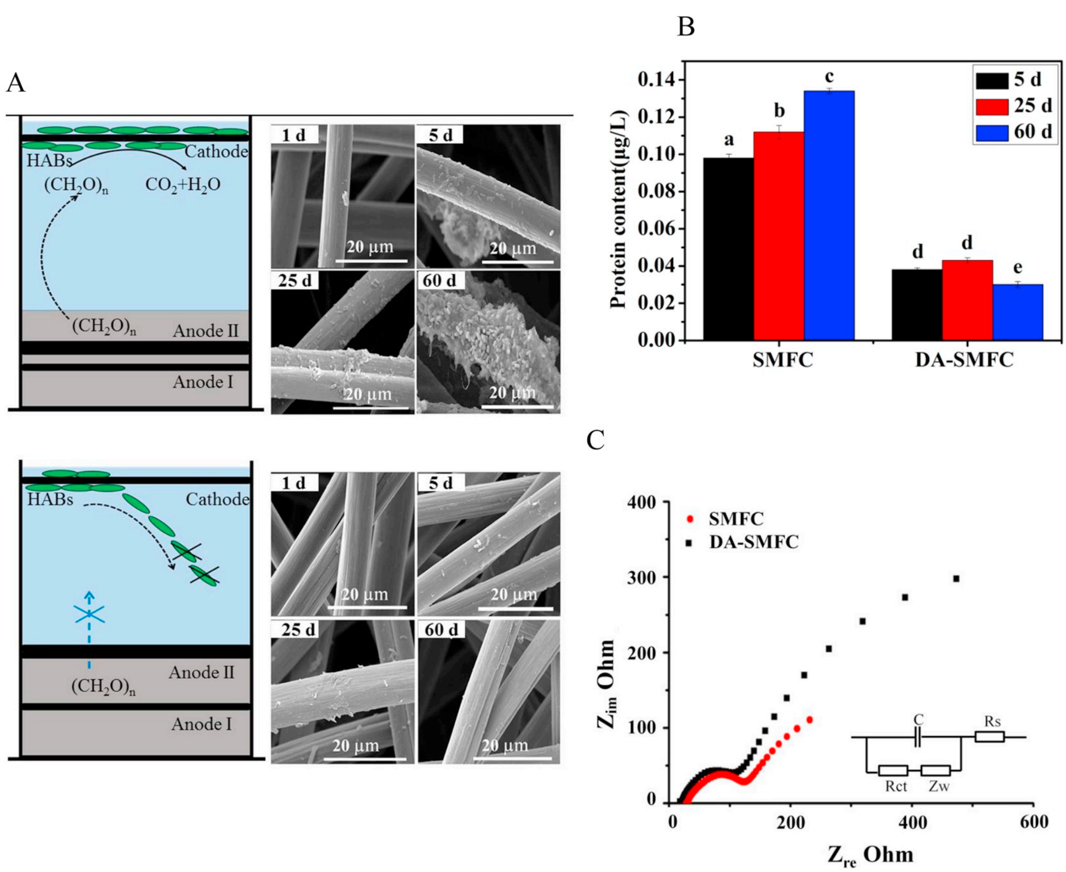

Fig. 4. A) Dual anode strategy used by (Yang et al., 2018a) to inhibit diffusion of organic matters to the cathode (SMFC top and DA-SMFC bottom) and biofouling controlling, B) protein content and C) cathodic charge transfer of the studied systems.

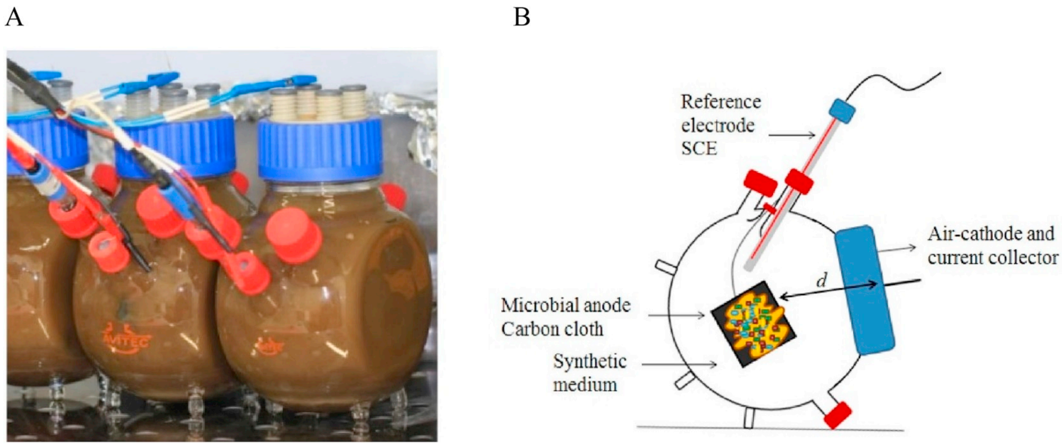

Fig. 5. A) Experimental set up and B) scheme of the removable air cathode SMFC (Oliot et al., 2016). Copyright (2019) with permission from Elsevier.

applying cloth separator. Power output was about 53% (NS) and 38% (Sp) of that recorded at the beginning of experiments (Rossi et al., 2018). However, (Yang et al., 2018b) suggested to bond the separator chemically to minimize the interstitial volume between the separator and the cathode that was believed to cause biofilm formation or accumulation of high pH solution. Nevertheless, power decay was observed in both studies but with a lower rate than of non-treated air cathodes.

As firmly discussed, the suggested physical methods for cleaning and inhibiting the biofilm formation on the cathode could recover the cathodic performance to some extent. However, since the biofilm penetrates into the inner layers of the catalyst and causes internal fouling, the performance cannot reach to its initial stage before biofouling. Moreover, physical methods may not be applicable to all configurations and for every surface area especially in the large scales. For that reason, chemical methods may be considered as the second option to mitigate the biofouling problem (Gule et al., 2015).

3.2. Chemical cleaning

Chemical cleaning is another approach to clean the biofilm. The interaction between the chemical and the biofilm can be defined through three steps; chemical adsorption on the microorganism cells of the biofilm, chemical penetration into the cells, and finally cell death due to deformation of the cell structure (Gule et al., 2015). Zhang et al. (2017) reported activated carbon (AC) as an efficient and cost effective option for the cathode material compared to Pt cathode. They have shown the effectiveness of weak hydrochloric acid cleaning of the AC cathode compared to the Pt cathode. Acid cleaning could restore the power generation to 85% of its initial value in the AC cathode SCMFC compared to 21% in Pt cathode SCMFC, after 17 months of operation. The results suggested that the chemical cleaning of cathode resulted in the loss of Pt catalysts from the cathode surface, which ultimately reduced the performance of the system. However, soaking the AC-based cathode in the weak HCL acid dissolved the salts precipitated in different diffusion layers of the catalysts besides the biofilm removal without noticeable corrosion. Liu et al. (2018) used sequent washing with solutions of 10 mM sodium dodecyl sulphate (SDS) and 5 mM sodium hydroxide (NaOH) for 2 h to clean the cathodic biofilm formed over six months of operation. The cathode was also washed additionally with another sequent of 60 mM of HCL for 2 h and the results were compared as shown in Fig. 6.

After six months of operation, the power generation dropped remarkably from 1330 to 888 mW/m^2 and the cathode internal resistance increased from 25.1 to 70 Ω. Chemical cleaning of the cathode biofilm with SDS + NaOH and SDS + NaOH + HCl could successfully retrieve 80 and 86% of the power output of the SCMFC before fouling, and reduce the internal resistance to 43.8 and 35.2 Ω, respectively (Liu et al., 2018).

Pasternak et al. (2016) achieved full recovery of biofouled and deteriorated cathode in terms of power and current generation through the alkaline lysis washing. The process was claimed as a fast, easy, inexpensive and more importantly an in-situ cleaning process. They constructed a new reactor using ceramic earthenware cylinders as both chassis and membranes which the outer surface of cylinders was covered

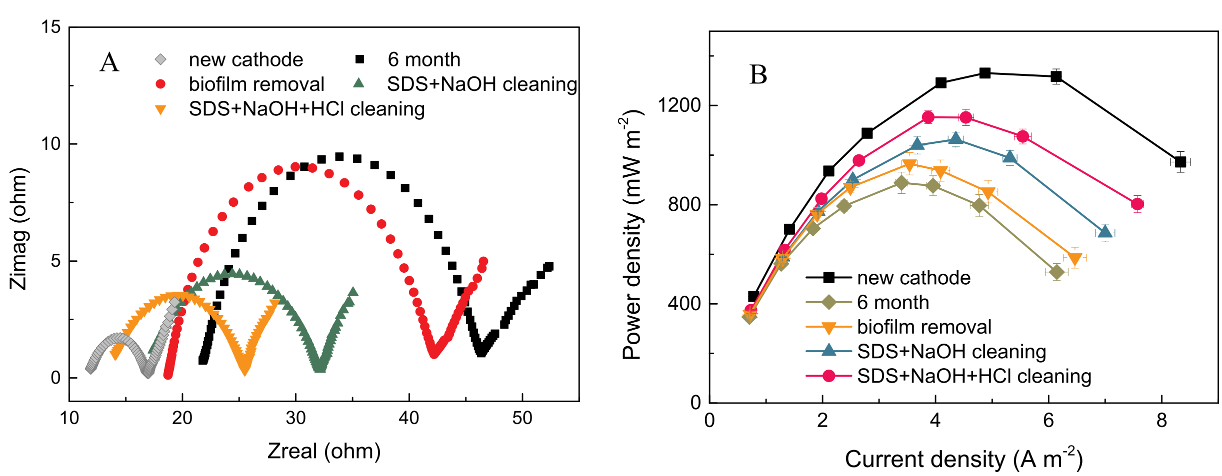

Fig. 6. A) Nyquist plots and B) power density curves of SCMFCs with the new cathode and after chemical cleaning of the cathodes over six months of operation (Liu et al., 2018). Copyright (2019), with permission from Elsevier.

by two layers of conductive carbon paint (CCP) as the cathode (Fig. 7. A). Owing to the design, they easily washed the surface of the cathodes with 0.1 M NaOH, lysis solution (0.2 M NaOH+0.1%Triton X-100, heated to 60 °C) and deionized water subsequently once the biofilm formation decreased the MFC performance. An additional step of replacing the second layer of CCP was also performed. They have also tested solely lysis washing of the cathode until no further power increase was observed. Both procedures were reported successful and could recover 100 and 107% of the power obtained when using new cathode as shown in Fig. 7. Moreover, it was suggested that the earlier treatment of the cathode by lysis solution could prevent biofouling and fouling of the cathode without further performance drop (Pasternak et al., 2016).

Rossi et al. (2019) in a very recent study evaluated the use of NaOH, ethanol, HCl, vinegar and steam cleaning to determine the best cleaning approach for restoring the performance in biofouled cathodes with polyvinylidene fluoride (PVDF) and polytetrafluoroethylene (PTFE) diffusion layers. Although the power decay in the SMFCs with PVDF (70%) and PTFE (78%) cathode diffusion layers after two months were quite different, the impact of cleaning methods was quite similar on both systems. Concentrated HCl or acetic acid could completely restore the initial power densities mainly due to removing salt penetrated into the AC catalyst layers. The study has also provided a detailed cleaning protocol for each approach, which can be employed by wastewater treatment plant (WWTP) operators. However, a detailed cost analysis is needed to compare the methods from an economical viewpoint.

Using chemical may not be the best option with respect to cost, safety and environmental impact. Moreover, it might be needed to remove the cathode for a chemical cleaning or else a very careful procedure is needed to avoid any negative effect on the anodic biofilm in an in-situ cleaning process. Moreover, the chemical which is used should be non-toxic and harmless to the environment which may limit the range of chemicals to be selected. In the case of Pt or catalyst coated cathodes, washing may lead to loss of catalyst layers from the cathode and performance drop after cleaning. However, it is noteworthy that the chemical cleaning can effectively target salts which were precipitated and penetrated into the catalyst layers for some electrode materials e.g. activated carbon (Gule et al., 2015; Zhou et al., 2018).

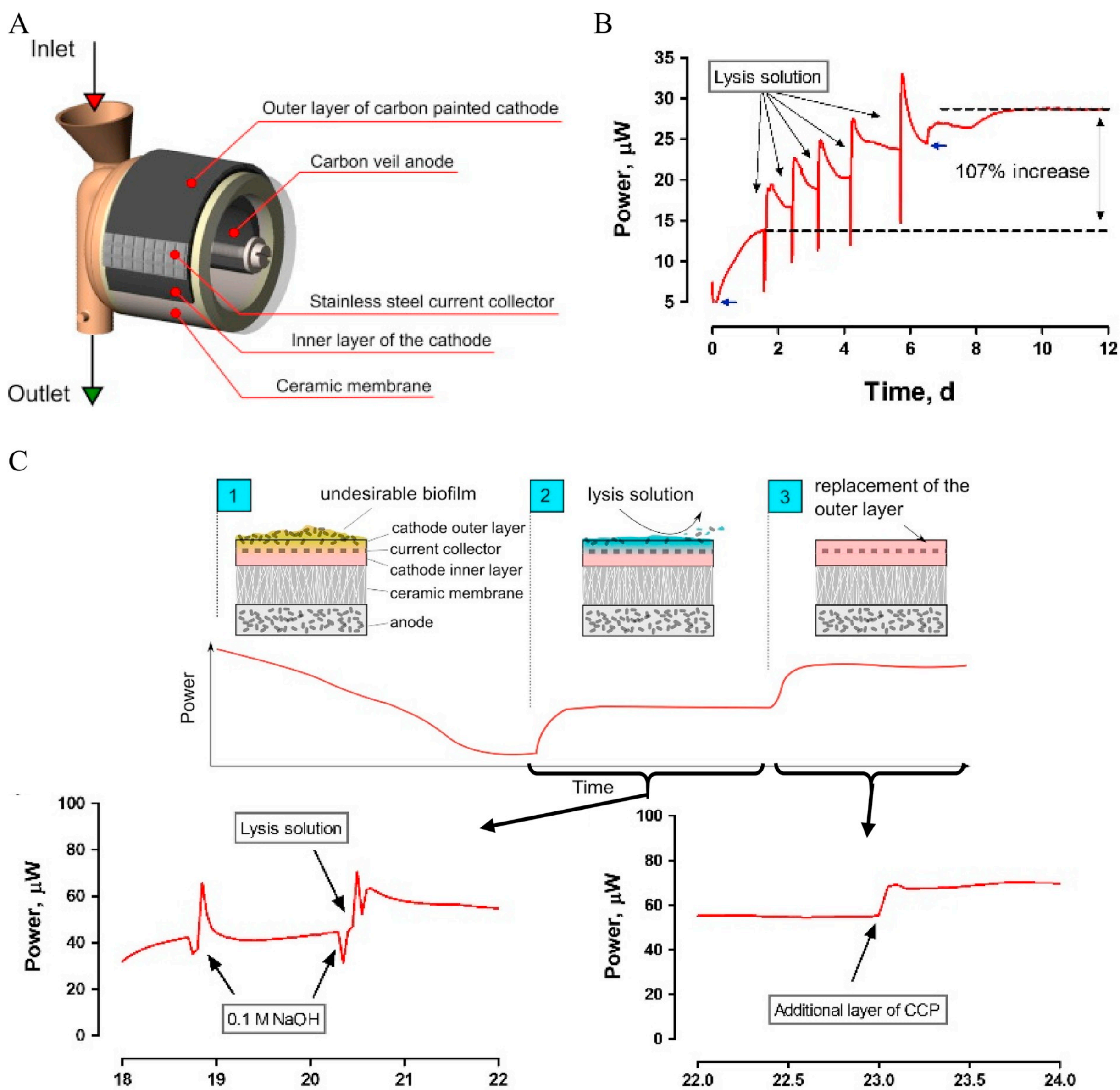

Fig. 7. A) Schematic design of the air-cathode SCMFC which the power regenerated by B) sequent lysis washing and C) 0.1 M NaOH + lysis solution (0.2 M NaOH+0.1%Triton X-100, heated to 60 °C) washing, and replacing the second layer of CCP (Pasternak et al., 2016). Copyright (2019), with permission from Elsevier.

3.3. Electro kinetic control

The electrical field has been shown as another effective way to detach biofilms from the cathode by using three main forces of electroosmotic, electrophoretic and electrostatic. Movement of ions as a result of alternating currents (AC) field could loosen the biofilm and make the biofilm detachment easier (Zhou et al., 2018). Wang et al. (2016) investigated the inhibition effect of different frequencies of AC on the biofilm activity for the first time. They have found that high frequencies of AC (1–1000 kHz) inhibited the biofilm activity temporarily while lower frequencies than 100 Hz acted more permanently by loosening the biofilm due to fluid convection in the biofilm driven by the electrohydrodynamic force. Biofilm Removal was also reported at a low frequency of AC (1 Hz) due to water electrolysis. Zhou et al. (2018) have applied a half-wave rectified alternating field (1.2 V/1 kHz) to excite the ions in pores. They have hypothesised that AC may move the ions migrated into the cathode pores and then loosen and detach the biofilm which deactivated the cathodic catalyst. They have recovered 77% of the initial power using forward direction half-wave AC (AC^+) as shown in Fig. 8. The method was promising since no external scraping or chemical cleaning was involved and both precipitated salts and biofilm were affected by the waves.

Application of electrical field may not be so preferable due to the complexity of wave form and energy consumption. Moreover, the periodical treatment is needed to avoid biofilm formation. Therefore, the recent interests are more into permanent solutions with a longer impact. Surface modification to modify the hydrophilicity of the cathodic surface and or incorporation of an antimicrobial agent into the catalyst layers to prevent biofilm formation are also promising research approaches.

3.4. Surface modification

Surface modification can be applied by fabricating materials with optimal hydrophilic properties (hydrophilicity control) or incorporating an antimicrobial agent into the catalyst layer.

Li et al. (2016) studied the effect of cathodic hydrophilicity on biofouling of the air cathode. Since the binders play the main role on the hydrophilicity of the catalyst layer, they have fabricated cathodes using different concentrations of hydrophobic PTFE and amphiphilic LA132 as binders. PTFE is a widely used binder in the air cathode MFC which showed a comparable power density to that of obtained by expensive hydrophilic nafion binder. However, the hydrophobic PTFE is more prone to biofouling, which needed further surface modifications. The application of mixed binders of PTFE and LA132 showed the positive effect of the optimal hydrophilicity on the SCMFC performance (power and charge transfer resistance) and biofilm formation (biofouling). The cathode with 67% LA132 showed the highest electrochemical activity since the optimal hydrophilicity of the catalyst layer extended the reaction interface and facilitated the oxygen reduction on the cathode. Moreover, the electronegative nitrile groups in LA132 binder formed a repulsion force which effectively reduced the bacterial adhesion and biofouling by 47% compared to the PTFE cathode (Li et al., 2016). This finding could explain why the mixture of polyvinyl alcohol (PVA) and vanillin eliminated biofouling as investigated by Chatterjee and Ghangrekar et al. (2014). Moreover, while the hydrophilicity control by using hydrophobic PTFE and amphiphilic LA 132 worked well in the first study (Li et al., 2016), the mixed binders of PTFE and hydrophilic PVA enhanced biofouling by attracting both hydrophilic and hydrophobic foulants in the other study (Chatterjee and Ghangrekar, 2014). The study showed that the components of the mixed binders are also playing an important role in biofouling. Moreover, the lack of ORR catalytic activity of vanillin restricted its application as a biocide to be incorporated into the cathode catalyst. Therefore, Noori et al. (2016) used bio electrochemically active silver nanoparticle (Ag-NP) for its antifouling properties in the SCMFC air cathode. Low charge transfer resistance, high power and enhanced CE were observed while the Ag-NP air cathode inhibited the fungal biofouling as well. Bimetallic Pt–Ag alloys were suggested in another study by Noori et al. (2018) to combine catalytic and anifouling properties of both metals to increase the SCMFC performance. They achieved high power of 999 mW/m^2 which was significantly higher than the Ag-NP cathode (147 mW/m^2) without noticing any sign of biofouling for 40 days of operation. Recent studies on cathode material proved high performance of transmission metal sulphide catalyst coated on the cathode. Silver + ferrous sulphide + partly graphitized carbon (Ag/FeS/PCG) catalyst from waste pomelo skin was reported as a promising catalyst type by taking the anti-biofouling advantage of silver for a durable high performing air cathode. Biocidal properties of silver besides to its high conductivity when alloyed with any elements suggested it as an efficient element when the surface modification was chosen for the biofouling mitigation. The power declined by 19.9% to 1090 mW/m^2 in the proposed electrode composite after 90 days of operation which was still far higher than 306 mW/m^2 in Pt/C cathode (Sun et al., 2017). However, the power output and antifouling effect were lower than their previous study on nanosilver/iron oxide/graphite carbon with 1642 mW/m^2 power output with only 4.2% power decay after 17 cycles (Ma et al., 2015). Other forms of Ag incorporated cathode were listed in Table 1. The antibacterial property of zinc oxide was also reported recently by Yang et al. (2019). Although a significant low biomass was formed on Zn/Co coated catalyst compared to Pt coated one, the study just showed the results for only one month of operation which was not sufficient to decide on the long-term stability of the Zn/Co coated cathode (Yang et al., 2019). The AC air cathode was also modified by using quaternary ammonium compound due to its bifunctional property as both catalyst and biocide (Li et al., 2014). The power was reduced by 21% after two months of operation, which was quite lower than the power decay in the plain AC (31%). Incorporation of antibiotic into the catalyst layer was another approach to inhibit biofilm growth which only studied once. Liu et al. (2015) studied the inhibitory effect of enrofloxacin (ENR) added to the AC

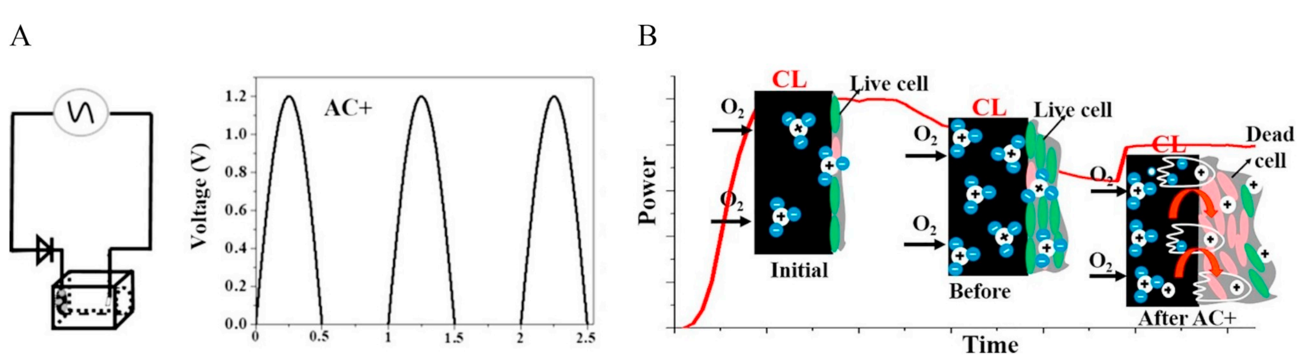

Fig. 8. A) Schematic of circuit connection and wave form for forwarding direction half-wave AC regeneration method and B) the power recovery through this method (Zhou et al., 2018), Copyright (2019) with permission from Elsevier.

Table 1
A summary of biofouling treatment and mitigation methods in SCMFCs.

Biofouling Control method	Anode	Cathode	Procedure of Control	Operation length (d)	Details of mitigation method	**Initial Power mW/m^2**	**Power After biofouling mW/m^2**	**Recovered power mW/m^2**	Ref
Physical treatment	Graphite fibre brush (GFB)	Activated carbon (AC)+ PTFE + stainless steel mesh (SSM)	Placing magnate at both sides of the cathode	30	Magnate and daily cleaned	247	132	133	Rossi et al. (2018)
	Carbon felt (CF)	Air direct methanol fuel cell (DMFC)+Pt cathode	Using separator on air cathode side	37	Using separator with low oxygen transfer rate on air cathode side	254	254	254	Vogl et al. (2016)
	Carbon fibre brush (CFB)	AC + PTFE + SSM	Biofilm removal	180	Removing biofilm	1330	888	965	(Liu et al., 2018)
	Carbon cloth	Air cathode + SSM	Air cathode movable design	25	Replacing the air cathode	700	–	1960	Oliot et al. (2016)
	CF	AC + PTFE + SS	Biofilm removal	365	Scrapping the biofilm	1722	777	834	Zhang et al. (2017)
Chemical treatment	CFB	AC + PTFE + SS	chemical cleaning (SDS + NaOH)	180	SDS + NaOH cleaning	1330.9	888	1063	(Liu et al., 2018)
	Carbon fiber veil	Conductive carbon paint on earthenware cylinder	Lysis + NaOH + CCP	86	Washing with NaOH, lysis solution and adding additional layer of CCP	105 μW	9.8 μW	105 μW	Pasternak et al. (2016)
			Lysis	–	Repeated lysis washing until stable power recorded	14	–	28.98	
	CF	AC + PTFE + SS	Washing + drying + pressing	365	AC was washed, dried and repressed with a new PTFE filter	1722	777	1800	Zhang et al. (2017)
	CFB	AC + PVDF + SS	Bonded separator + acid wash	65	Bonding the AC cathode with a cloth separator and washing with acid after fouling	380	190	230	(Yang et al., 2018b)
			Non-bonded separator + acid wash		AC cathode with a cloth separator (not chemically bonded) with acid wash after fouling	340	60	250	
	GFB	AC + PVDF + SS	Ethanol wash	60	Soaking the AC cathode in 70% ethanol	300	90	160	Rossi et al. (2019)
			HCL wash		Soaking the AC cathode in 100 mM HCl			320	
			NaOH wash		Soaking the AC cathode in 100 mM NaOH			160	
			Steam cleaning		Steam cleaning by steamer			190	
		AC + PTFE + SS	HCL wash		Soaking the AC cathode in 100 mM HCl	320	70	360	
					Soaking the AC cathode in 1 mM HCl			80	
					Soaking the AC cathode in 0.01 mM HCl			50	
			vinegar wash		Soaking the AC cathode in 5% acetic acid			330	
	GFB	AC + PTFE + SS Pt + SS + PDMS	Acid wash	480	Soaking the cathode in 60 mM HCl acid	1040 1270	860 250	1020 270	Zhang et al. (2014)

(*continued on next page*)

Table 1 (*continued*)

Biofouling Control method	Anode	Cathode	Procedure of Control	Operation length (d)	Details of mitigation method	**Initial Power mW/m²**	**Power After biofouling mW/m²**	**Recovered power mW/m²**	Ref
Electrical treatment	CFB	AC + PTFE + SS	Electric field	30	Applying forward direction half-wave on the cathode	1842	1179	1426 (77%)	Zhou et al. (2018)
Hydrophilicity control	Carbon fibre brush	AC + PTFE + SS	Hydrophilicity control	150	Hydrophilicity control of the cathode by adding hydrophobic PTFE binder	1028	–	17 mg protein formation on biofilm	Li et al. (2016)
	Carbon fibre brush	AC + PTFE + LA132 + SS			Hydrophilicity control of the cathode by mixing 67% amphiphilic LA132 and 33% PTFE binders	1171	–	9 mg protein formation on biofilm	
Surface modification	Carbon fibre brush	Carbon black (CB)+ PTFE + stainless steel + AgNP/Fe3O4/GC	Incorporation of silver/iron oxide/ graphitic carbon composite into the cathode	17 cycles	AgNP/Fe3O4/GC	1712	–	1642	Ma et al. (2015)
		Carbon black + PTFE + stainless steel + Fe3O4/GC			AgNPs/AC	1476		1004	
		Carbon black + PTFE + stainless steel + AgNP/AC			Fe3O4/GC	1328		792	
		Carbon black + PTFE + Pt			Pt–C	1210		993	
	Carbon felt	SS wire + Ag–C–Pt + PTFE + Nafion	Incorporation of silver composite into the catalyst layer	80	C–Ag3Pt	999	–	946	Noori et al. (2018)
		SS wire + C–Pt + PTFE + Nafion			Pt–C	903	–	859	
	CFB	SSM + PTFE + Ag/FeS/PGC	Incorporation of silver/ferrous sulfide/partly graphitized carbon into the catalyst layer	90	Ag/FeS/PCG	1361	–	1090	Sun et al. (2017)
		SSM + PTFE + CB			Pt–C	483		306	
	Carbon Cloth	Graphene oxide-supported cobalt and zinc oxide nanoparticle (GO-Zn/Co)+ PTFE + carbon cloth	Incorporation of cobalt and zinc oxide nanoparticle into the catalyst layer	30	GO-Zn/Co	773		566	Yang et al. (2019)
		Pt + PTFE + carbon cloth			Pt–C	744		354	
	GFB	AC + ENR	Antibiotic incorporation into the catalyst layer	90	ENR + AC	930		1069	Liu et al. (2015)
		AC			AC raw	1055		841	

catalyst layer. The broad spectrum of antimicrobial activity of ENR against a wide range of bacteria and excellent electrochemical stability within air cathode working potential made ENR a suitable option to inhibit cathodic biofilm growth. Moreover, ENR is water insoluble which makes it hard to be released into the electrolyte. No power decay was reported for ENR treated cathode for over three months of operation compared to 21% reported for the cathode treated with quaternary ammonium compound and 20% in the Ag treated cathode in other studies. Moreover, a careful procedure like a coating carrier or physical entrapment could be applied to prevent antibiotic permeation into the catalyst layer which reduced ORR catalytic property of activated carbon layer. Liu et al. (2015) suggested the joint addition of AgNP and antibiotics like ENR to take advantages of antibacterial effect of ENR and high ORR catalytic activity of AgNP to maximize the SCMFC performance without biofouling-related performance decay. However, no study has been reported in this regards since then.

4. Conclusion

Single chamber air cathode MFC is one of the most feasible, practical and cost-effective configurations for wastewater treatment over the long-term operation. However, the air breathing cathode electrode-which passively uses the air with high oxygen reduction rate-is susceptible to biofouling making the air cathode system high in cost and poor in durability. Extensive attempts have been devoted to prevent and mitigate biofouling on the air cathode SMFCs from physical cleaning to surface modification. The ultimate solution of biofouling control may involve one or integrated approaches which are reviewed in this article. While some of the proposed mitigation strategies like hydrophilicity control of the binder and surface modification could delay electrode biofouling, concentrated acid wash can be considered as a simple and efficient cleaning method to fully restore the air cathode performance. Moreover, regardless of what mitigation approach is suitable, modifying the SMFC configuration to allow simple removal and cleaning of the air cathode without exposing the anode to the air is needed to be taken into consideration. Moreover, it was seen that cleaning methods were not applicable to the cathodes coated with thin layers of catalyst e.g Pt-coated electrode, while activated carbon cathode showed high power recovery after cleaning without facing the problem of removing off the catalyst layer. Furthermore, despite all comprehensive studies on mitigation approaches, there is no specific cost evaluation study on different approaches in one unique setup to define a clear protocol to fabricate durable air cathodes with feasible cleaning procedure after power decay.

Acknowledgment

The authors wish to extend their appreciation to Sultan Qaboos University, Muscat, Oman, for financial support through His Majesty's Trust Fund (SR/ENG/CAED/17/01) collaborated with University of Alberta, Canada.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.bcab.2019.101370.

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