

Effects of electrode material and surface area on the performance of microbial fuel cells treating leachate

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ABSTRACT

In this study, the effect of electrode material and surface area on the bioelectricity generation and leachate treatment using fixed-bed microbial fuel cells (FB-MFCs) were examined for 90 d. Two materials including graphite rod (GR) and carbon cloth (CC) were used as electrode in air-cathode and anode compartments. The performance of the FB-MFCs that has been packed with granular activated carbon was investigated using a mixed microorganism culture for leachate bioconversion in batch mode operation. The maximum removal of chemical oxygen demand and total Kjeldahl nitrogen were 99%, and 99.79%, respectively when surface area of the carbon cloth was 0.036 m². The maximum power density (99.8 W/m³) and current (3.323 mA) achieved when carbon cloth has been applied as electrode and the surface area of electrodes were 0.018 and 0.036 m², respectively. This study verified the potential of FB-MFCs as an efficient system for simultaneous leachate treating and power generation. Furthermore, the results provided valuable information for the further development of a continuous MFC system using graphite rod and carbon cloth as electrode.

Keywords: Microbial fuel cell; Leachate; Fixed-bed; Activated carbon; Energy; Electrode

1. Introduction

Over the last few decades, the global need for energy has multiplied, which has pushed up fuel prices. To cope with rising fuel prices, researchers have made great efforts to find out sustainable and low-cost ways to generate electricity [1]. Among different methods, microbial fuel cells (MFCs) as a cost-effective technology with lower energy consumption for efficient operation have been proposed to address environmental concerns and energy insecurity with minimal or zero use of hydrocarbons [2,3]. Furthermore, MFCs system has a great potential for generating bioelectricity through wastewater treatment [4,5]. This technology has been used to purify different wastewaters contain

organic matter and nutrients that originated from different sources such as food waste leachate [5], landfill leachate [6], industrial wastewater [7], rejected water [8]. Among them, compost leachate encompasses high concentrations of pollutants (including organic materials and nitrogen compounds) that are seriously harmful to the environment and human health [9]. The MFC bioreactors, showed promising results for treating leachate from different sources [5,10].

However, the system has showed a notable enhancement in power density, but its full-scale application is limited due to low electricity generation and high cost [11]. The results of previous research works have confirmed that the efficiency of MFC system can be influenced by operating conditions (temperature, solution pH, electrical conductivity,

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and hydrodynamics), configuration and operational parameters (electrode materials and surface areas, nature of bacteria and types of substrate) [1,12–14]. Among different factors, electrode play key role in performance and cost of MFCs. They influence microbial attachment, electron transfer, internal resistance and rate of electrode surface reactions, which these effects are different for diverse electrode materials due to alterations in their physical and chemical properties [15]. Therefore, over the past decade researchers have examined the effect of different electrode materials on the efficiency of MFCs [16–19].

Regardless of the type of electrodes, they all should generally have good electrical conductivity, good chemical stability, high resistance and reasonable price. Carbon materials with the required characteristics have been used in MFCs [20–22]. Tsai et al. [23] used a modified carbon cloth electrode with carbon nanotube in the MFC system. The results showed that the power density and coulombic efficiency of system increased to 65 mW/m² and 67%, respectively. Zhao et al. studied the performance of three different types of electrodes (graphite foil, carbon cloth and carbon cloth fabrics) and found that carbon fabric has better performance than graphite and carbon cloth with maximum power density of 0.15 mW/m² [16].

In this study, a two-chamber MFC has been used to investigate the effects of material and surface area of electrode on the power generation and leachate treatment and it is potentially a very interesting possibility to improve power densities. Therefore, the carbon-based electrodes including graphite, and carbon cloth were investigated to determine

the best electrode. Moreover, the effect of electrode surface area ranging from 0.018 to 0.054 m² was also taken into account under the same operating conditions, when the reactor fed with real leachate from composting site.

2. Materials and methods

2.1. MFC construction and operation

A dual-chambered MFC with a working volume of 0.01 m³ for each chamber was constructed from Plexiglas tube, with 12 cm inner diameter, and 23 cm height. A cation exchange membrane (CEM) as shown in Fig. 1 separated the chambers in the H-shaped design reactor. Each chamber has been filled to a height of 20 cm by prepared granular activated carbon (GAC) from vine wood at the grain size (0.5–1 cm in diameter) as bio-carrier and third electrode. The pretreated membrane (Nafion, 117, USA) with surface area of 30 cm² has been used through a conjunction to create an electric current between the anode and the cathode. The carbon cloth and graphite were used as electrode in both chambers. The distance between electrodes was different from 1 to 3 cm. All electrodes have been pretreated prior to be used in reactor through method applied by Ketep et al. [24]. An embedded waterproof copper wire was used as a current collector through a multimeter (PROVA 903, Taiwan) from anode and cathode. An aquarium air pump (RESUN 5505 model, China) and porous stone diffusers were used for supplying continuously the aeration to the cathode chamber at a flow rate of 15 L/min (DO ~3 mg/L).

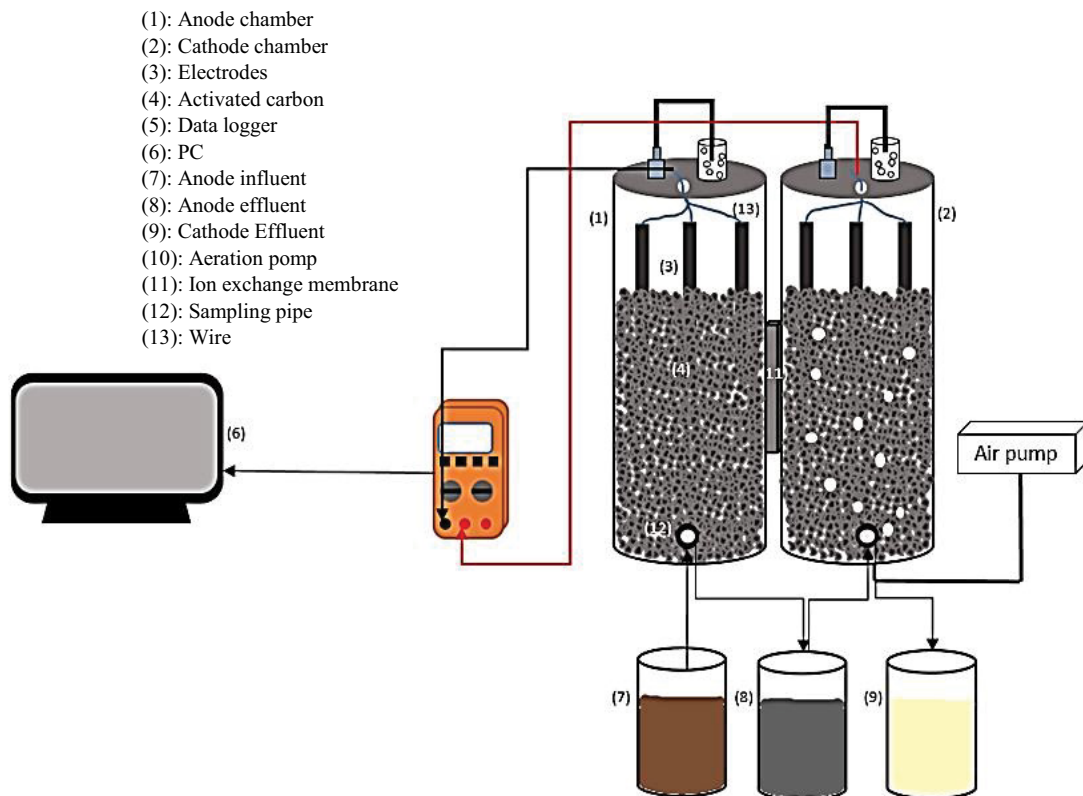


Fig. 1. Experimental set-up.

The system has been operated at batch mode (four stages) for different conditions according to the Table 1. Power data have been collected every 2 min using multimeter with data logger. The reactor temperature was kept at level of $33^{\circ}\text{C} \pm 2^{\circ}\text{C}$. The sampling carried out based on the reactor cycle from influent and effluent of the anode and cathode compartments.

2.2. Inoculum and wastewater

The sludge collected from the municipal wastewater treatment plant (Kermanshah, Iran) was used as inoculum. The biomass with MLSS $\sim 10,000$ and $3,000$ mg/L has been inoculated on the bed of activated carbon in both anode and cathode chambers, respectively. The acclimatization of microorganisms with leachate in the start-up phase, a step-by-step process with different percentage of leachate (20%, 40%, 60%, 80% and 100%) has been performed under aerobic and anaerobic conditions for one month. After appearing light brown and black brown colors on the surface of granular activated carbon, the effect of main factors after washing and reinstalling all connections and tubing on the reactor performance have been investigated.

The leachate samples were collected from the compost site (Kermanshah, Iran). All containers were stored at temperature below 4°C for future use. The physicochemical characteristics of leachate in Kermanshah compost site are summarized in Table 2. The color of leachate was dark brown and the biochemical oxygen demand/chemical oxygen demand (COD) ratio was 0.35. The COD value of raw leachate was about $120,000$ mg/L with acidic pH. The diluted leachate at desire value of pH was injected into the reactor from the anaerobic column.

2.3. Calculations and data analysis

The efficiency of system to treat leachate was evaluated through determining different parameters such as COD, total Kjeldahl nitrogen, ammonium, nitrite, and nitrate according to the standard method 21th ed. [25]. Other parameters including temperature, dissolved oxygen and

pH were measured routinely in order to control the process. A digital multimeter with a data logger was used to monitor continuously the value of the voltage (V) and current (I) at 2 min intervals. The power was calculated using Eq.1.

$$P = VI \quad (1)$$

where V is the potential (mV) and I is current (mA).

The power density (mW/m^2) can be estimated using Eq. (2).

$$p = 10 \times V \cdot i \quad (2)$$

where i is the current density (mA/m^2). The coefficient 10 is used to equalize the units [26].

The coulombic efficiency (CE) according to the Eq. (3) is defined as the ratio of total coulombs actually transferred to the anode from the substrate, to maximum possible coulombs if all substrate are available for power generation [10].

$$\text{CE} (\%) = \frac{M \int_0^t I dt}{F \cdot v \cdot 4 \cdot \Delta\text{COD}} \quad (3)$$

where M is molecular weight of O_2 (32 g/mol), I is current generated during whole batch cycle (A), F is Faraday's constant (96,485 C/mole electrons), and v is volume (L).

3. Results and discussion

3.1. Acclimation and start-up

The treatment efficiency and power generation of a MFC depends on the total population of microorganisms and the percentage of electrogenic microorganisms within this total population, respectively [27]. Therefore, suitable enrichment of electrogenic microorganisms was aim of the first phase in this study. In this stage, with the creation of suitable aerobic and anaerobic conditions, the enrichment

Table 1
Operation conditions for FB-MFCs

Parameters	Anode	Cathode
Electrode	Single graphite rod (0.0226 m^2)	
	Three carbon cloth, 1 cm wide (0.018 m^2)	
	Three carbon cloth, 2 cm wide (0.036 m^2)	
	Three carbon cloth, 3 cm wide (0.054 m^2)	
Temperature	33 ± 2	33 ± 2
Total volume	3.5 L	3.5 L
Working volume	1.2 L	1.2 L
Operation cycle	48 h	48 h
Discharge	3 min	3 min
Filling	2 min	2 min
Lag	1 min	1 min
Air flow	–	15 L/min

Table 2
Physicochemical characteristics of compost leachate

Parameters	Units	Compost leachate	Influent leachate
pH	–	4.58	7 ± 0.09
Total Kjeldahl nitrogen	mg/L	1,820	165 ± 30
NH_4	mg/L	406	100 ± 40
NO_3	mg/L	660.44	220 ± 60
NO_2	mg/L	4.976	0.9 ± 0.2
COD	mg/L	120,000	10,000
Biochemical oxygen demand	mg/L	42,000	3,500
ALK	mg/L	3,853	$3,800 \pm 400$
P	mg/L	268	2.32
Conductivity	$\text{K } \mu\text{S}$	23.3	1.94
Biochemical oxygen demand/ COD	–	0.35	0.35

of microorganisms that are responsible for the removal of organic carbon and nitrogen compounds in two separate columns have been performed. The anaerobic column was fed with an initial COD concentration of 10,000 mg/L and the system efficiency was 55%. The MLSS concentration increased from about 3,000–10,000 mg/L because of proper growth of biomass, which indicated the system's ability to function at this condition. Investigating the concentration of MLSS at the beginning and the end of the aerobic process showed that, in addition to increasing the MLSS, the system has a good effect on the COD and ammonium removal.

3.2. COD removal efficiency

The FB-MFC was operated at initial COD concentration of 10,000 mg/L with HRT of 48 h at four different runs. At the steady state conditions, the COD of effluent was 100–160 mg/L, which indicates the removal rate of 89%–99%. The results showed a high removal rate in comparison with previous studies [28–32]. The overall efficiency of the system showed a high ability for the COD removal from leachate at all four steps by different electrodes. The COD removal were 96.4 ± 4 , 95.6 ± 2.9 , 97.17 ± 1.3 and 97.43 ± 1.381 mg/L for graphite rod, carbon cloth (0.018 m²), carbon cloth (0.036 m²) and carbon cloth (0.054 m²), respectively (Fig. 2). No significant difference was observed in COD removal at different stages (A, B, C and D) with different electrodes. Along with the MFC process, the adsorption of leachate by granular activated carbon is likely to result in further COD removal [33]. Ahn et al. investigated a MFC with different configurations for domestic wastewater treatment and results showed that system has ability to decrease COD about 93% [34]. Ganesh et al. [33] compared an activated carbon-MFC and biochar-MFC for treatment of leachate. Results showed that COD removal in activated carbon-MFC (74%) significantly was higher than biochar-MFC (28%).

3.3. Nitrogen removal efficiency

The system eliminated more than 99.2% of NH₄-N and no significant difference in the concentration of NH₄-N in

the effluent of MFC for different runs (Fig. 3). The total nitrogen removal efficiency was 99.8% at final effluent of reactor. The maximum removal of nitrogen in the cathode chamber could be related to the removal of ammonia through nitrification and denitrification processes. The biomass immobilization in the attached growth reactor has been applied to improve the nitrogen removal due to different advantages such as high biomass concentration, simultaneous nitrification and denitrification, and resistance to nitrogen shock loading and other environmental changes [35]. Previous studies have shown that the removal of NO₃-N in MFC systems can be attributed to exogenous bacteria that restore nitrate to nitrogen [36]. Nitrate greatly affects the coulombic efficiency due to electron competition between the generation of electricity and the process of denitrification [37]. Nitrate can be used in the cathode chamber where biocathode converting nitrate to nitrogen [38]). Based on the nitrogen concentration, the maximum efficiency of system was 83%. Ammonium ion concentration was measured during all of the steps and its removal efficiency was 99.2% (Fig. 3). Ammonium ion in the anode chamber passed through the membrane and entered the cathode section where the nitrification process was carried out with a series of bioelectrochemical reactions. These results indicate that ammonium ions act as an electron donor in the nitrification process and are continuously removed in the cathode chamber. The nitrogen removal has been reported in previous studies using MFC [39,40]. They indicated that both nitrite and nitrate function as cathode electron acceptors to produce power [41–43].

3.4. Power generation

Fig. 4 indicates the average generated current at steady state condition during the four runs of MFCs in this research work. The results showed that all of the electrodes have suitable efficiency and the maximum electricity current for graphite and carbon cloth with different surface areas of 0.018, 0.036 and 0.054 m² were 1.901, 1.850, 3.320 and

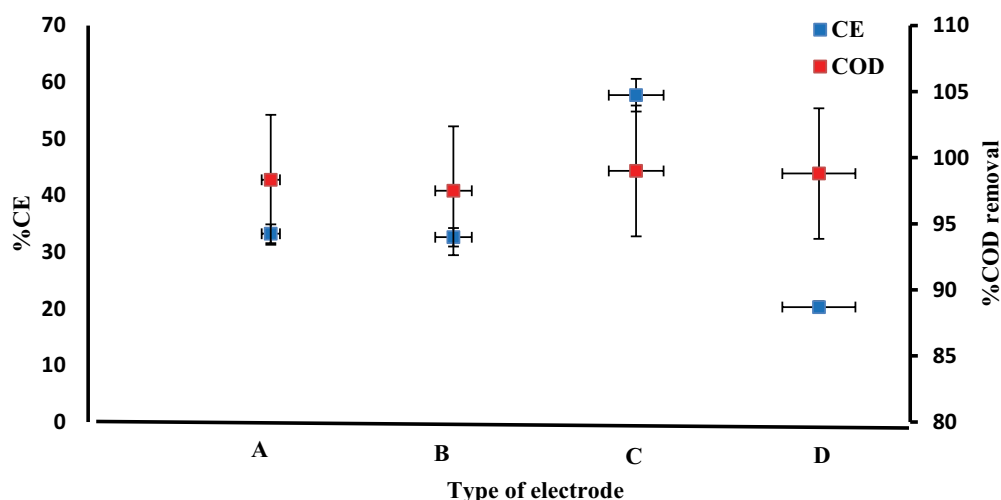


Fig. 2. Effect of electrodes on the COD removal and current efficiency (A: graphite rod; B: carbon cloth 0.018 m²; C: carbon cloth 0.036 m²; D: carbon cloth 0.054 m²).

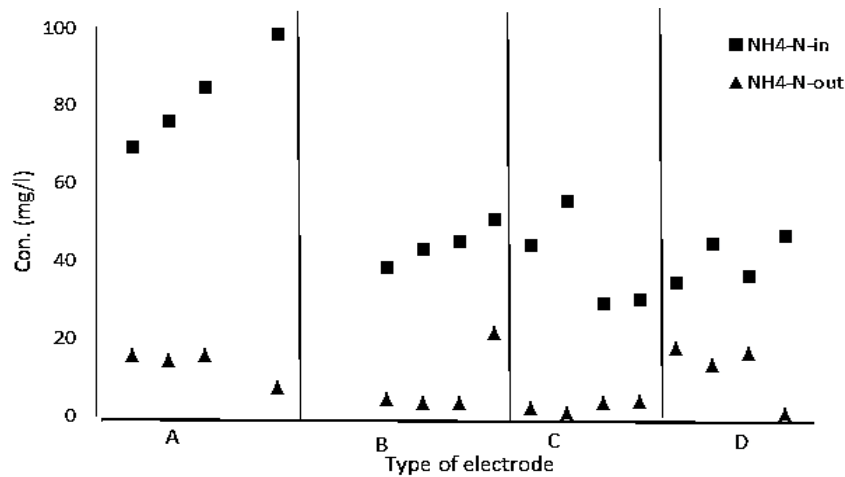


Fig. 3. The concentration of $\text{NH}_4\text{-N}$ for influent and effluent for different runs (A: graphite rod; B: carbon cloth 0.018 m^2 ; C: carbon cloth 0.036 m^2 ; D: carbon cloth 0.054 m^2).

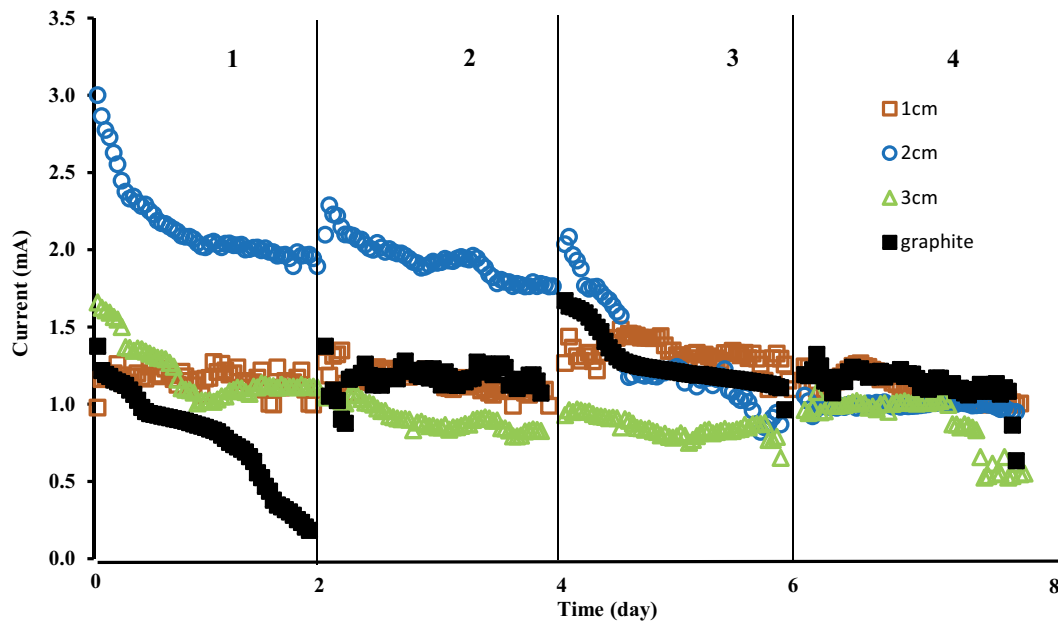


Fig. 4. Current generation in different runs with four repetition of each run.

1.170 mA, respectively. The amount of current was initially at its maximum level and then decreased gradually, and at the end of each run was produced at a relatively constant amount. The fact that at the beginning of each run there is more current is probably due to the presence of oxygen, nitrate and nitrite as electron acceptors [44]. The maximum current in the third stage with a 0.036 m^2 carbon cloth electrode was 3.32 mA and a maximum power density was obtained in the second-stage with a carbon cloth electrode of 0.018 m^2 , which was equal to 99.8 Wm^{-3} . High density can be due to the better electrochemical activity of bacteria in the reactor. The power density ($14.3\text{--}99.8 \text{ Wm}^{-3}$) is greater than that obtained in previous studies ($266\text{--}766 \text{ mW/m}^2$) [10,45,46].

Fig. 5 shows the power density, voltage and current during performance of FB-MFC treating leachate for four

runs. The coulombic efficiency of system from 20.3% to 57.7% was found to be greater than that obtained in previous studies on the leachate treatment. Ganesh and Jambeck [33], reported the maximum yield of coulombic $\sim 21\%$ when leachate with a COD concentration of $1,000 \text{ mg/L}$ was treated using a MFC reactor. The current of system with increasing the electrode surface area from 0.018 to 0.036 m^2 increased from 1.90 to 3.32 mA and further increase in the electrode surface to 0.054 m^2 was cause of current reduction to 1.17 mA . The achieved data based on the electrode surface area are similar to the results that have been obtained in previous studies [47,44]. The increase in the electrode surface area, resulting in a higher level of bacterial growth, which can improve current generation. Similar results with the use of carbon brush have been obtained in previous study [44]. Reducing current when the surface area

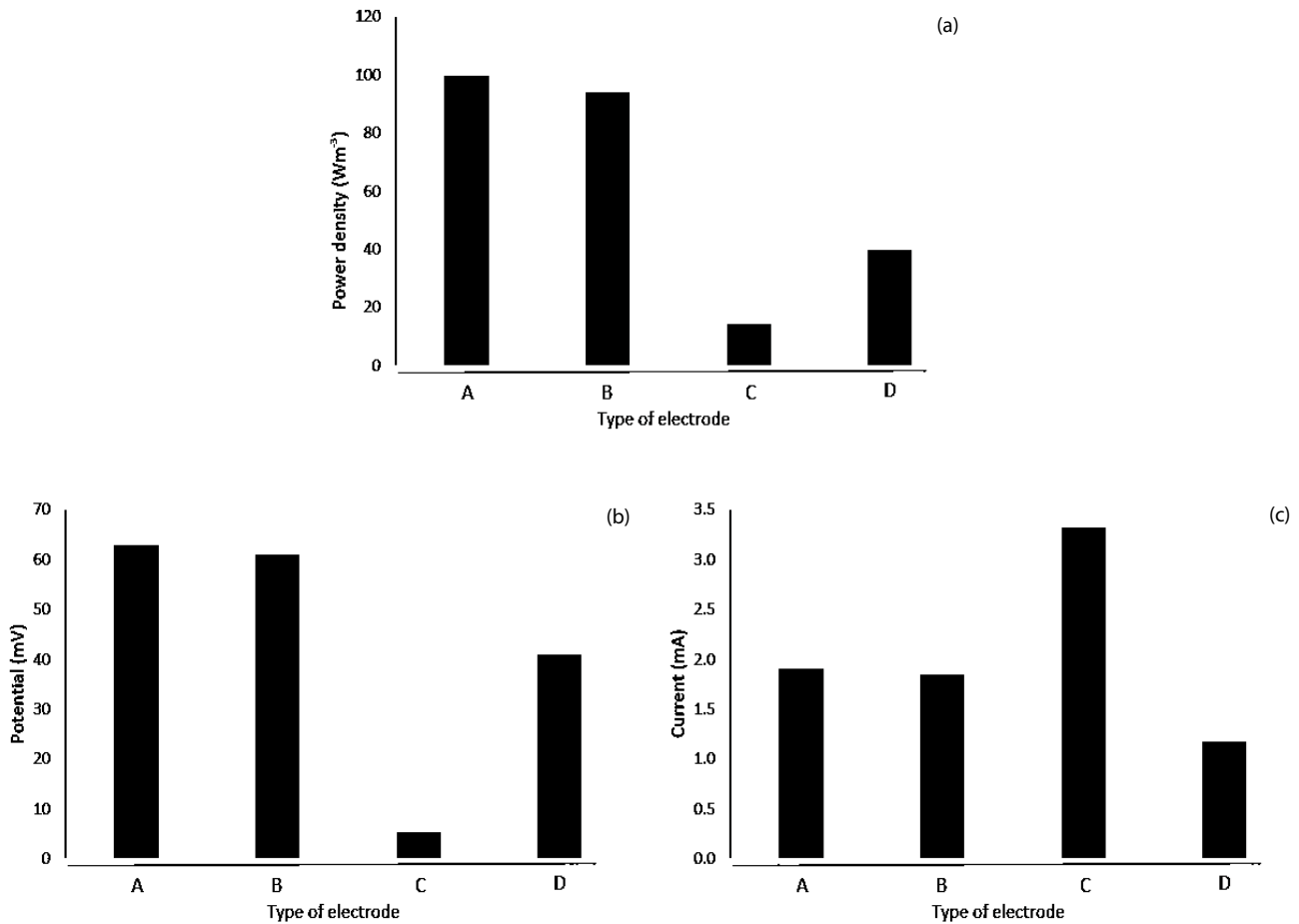


Fig. 5. Power density (a), potential (b) and current output (c) in different runs (A: graphite rod, B: carbon cloth 0.018 m², C: carbon cloth 0.036 m², D: carbon cloth 0.054 m²).

increases from 0.036 to 0.054 m² is probably due to the saturation of the electrode surface by the more volume of the sewage that causing increase internal resistance. This result is consistent with the Ghangrekar and Shinde result, in which a lower area of electrodes produces more current [2].

The coulombic efficiency (CE%) for MFC was in the range of 20%–57%, which was significantly higher for electrode with surface area of 0.036 m². The presence of nitrate in the leachate reduces the efficiency of the coulombic because it reduces electron access to the reaction in the cathode chamber by using organic matter, and on the other hand, the nitrate itself acts as an electron acceptor, which in turn reduces the efficiency of the coulombic. According to the results obtained in previous studies on the MFC and power generation, the maximum yield of coulombic with COD (1,000 mg/L), was equal to 21% and its it even reduced with increasing in COD concentration [48]. This is because electrochemical bacteria cannot convert all carbon to electricity. The results of this study were significantly higher than the results of studies by Gajaraj et al. [49] and Cheng et al. [50]. Probably because the ion exchange membrane has not been used in these studies, the oxygen penetration into the anode compartment in the absence of the membrane can reduce the coliform bacteria.

4. Conclusion

In this study, two electrode materials graphite (0.226 cm²) and carbon cloth with different surface areas (0.018, 0.036 and 0.054 cm²) were selected to investigate their effects on the performance of FB-MFCs for simultaneous leachate treatment and bioelectricity generation. These results demonstrated that the MFCs displayed the high bioelectricity generation using carbon cloth. The results showed that the maximum MFC efficiency obtained using 0.036 cm² carbon cloth electrode with COD removal of 99% and the bioelectricity generation of 3.323 mA.

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