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Performance of continuous flowing membrane-less microbial fuel cell with a new application of acrylic beads separator

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ABSTRACT

The performance of a membrane-less microbial fuel cell (ML-MFC) was evaluated to treat actual domestic wastewater. A novel application of inexpensive acrylic beads was examined to separate the anodic and cathodic compartments. The ML-MFC was alternatively inoculated with activated sludge and anaerobic aged sludge. Results revealed that the removal efficiencies of COD and BOD from actual wastewater were 79 and 60%, respectively, using the activated sludge as inoculum. Higher removal efficiencies up to 81 and 70% were obtained for COD and BOD removal, respectively, in ML-MFC inoculated with anaerobic sludge. Upon changing the inoculum type, a variation of current density and power generation was clearly observed. Within less than four weeks, the system generated a maximum stable current of 55.65 and 67.50 mA/m^2 for ML-MFC inoculated with activated sludge and anaerobic aged sludge, respectively. Effect of inoculum type was evaluated under variable external resistance.

Keywords: Microbial fuel cell; Wastewater; Bioelectricity generation; Activated sludge; Biomass granulation

1. Introduction

Among the new technologies to convert biomass to clean energy, microbial fuel cells (MFCs) have gained lots of attractions in recent years [1]. MFC is a device that directly converts the metabolic power of micro-organisms into electricity using electrochemical technology [2]. MFC is a promising green method to treat organic effluents and produce electricity at the same time [3]. Traditionally, MFC consists of two compartments, anode and cathode, separated by cation exchange membrane (CEM). Micro-organisms oxidize the substrate and produce electrons and protons in the anode chamber. Electrons, collected on the anode, are transported to cathode by external circuit and protons are transferred through the membrane internally. Electrons and protons are consumed in the cathode chamber by reducing oxygen, usually from water [4]. One of the most commonly used components in MFCs is the proton exchange membrane which permits the transfer of protons and other small cations, while limiting the crossover of organics or oxygen between the anodic and cathodic chambers [5]. However, the high cost as well as the bio-fouling and the associated high internal resistance could limit the power generation and the application of MFCs on

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large scales [6,7]. In order to make the process economically feasible as well as eliminating fouling problems, recently several experimental results have been reported in the literatures describing the performance membrane-less MFCs [8–12].

In this regard, several types of materials applied as separators between anode and cathode were in membrane-less microbial fuel cells (ML-MFCs). Jang et al. [8] reported the application of double-layer separators which consisted of glass bead layer supported by a glass wool layer in an up-flow twin-chambered ML-MFC. Within four weeks, a stable current of 2 mA and 90% COD removal were obtained in this ML-MFC fed with synthetic wastewater and inoculated with activated sludge. An ML-MFC was modified by Moon et al. [9] and used for continuous power generation and COD removal from artificial wastewater. The modified ML-MFC used a perforated polyacrylic plate as a separator fixed between the anode and cathode. Rodrigo et al. [13] studied the oxidation of the pollutants contained in an actual urban wastewater using a membrane-less two-chamber MFC separated by salt bridge using an anaerobic pre-treatment of the activated sludge. The power density generated was found to depend mainly on the organic matter contain (COD) but not on the wastewater flow-rate. Ghangrekar and Shinde [4] evaluated the performance of an up-flow dual-chambered mediator-less and membrane-less microbial fuel cell to treat synthetic wastewater and actual sewage. The two compartments in the ML-MFC were separated by glass bead layer supported by a glass wool layer. Maximum power density of 10.9 mW/m^2 and COD removal of 88% were recorded. Rodrigo et al. [14] studied the production of electricity and oxidation of organics in a synthetic wastewater fed with glucose and peptone of soybean as carbon sources to a membrane-less MFC consisted of a dark-glass basin separated into two identical compartments by a porous graphite plate to prevent the trespassing of organisms from the anodic to the cathodic chamber. This porous carbon separator can be considered to operate as a bipolar electrode. Han et al. [15] reported the performance of a dual-chambered ML-MFC with simulated intestinal fluid for continuous power generation to provide secure power for implantable medical devices. A carbon paper with thickness of 0.29 mm was used as separator between the two chambers to reduce the oxygen diffusion from cathode chamber which would affect the growth and electricity generation reactions of anaerobic micro-organisms in anode chamber. Ahn and Logan [16] developed a single-chamber membrane-less MFC fed with acetate-based synthetic solution. The scalable single chamber was designed with multiple graphite fiber brush anodes and a single air-cathode chamber alternatively separated from the anode using a glass fiber separator of 1.2 mm thick or textile (46% cellulose and 54% polyester) separator of 0.3 mm thick.

In the present study, an up-flow dual-chambered ML-MFC was designed and constructed to: (1) examine a new application of inexpensive acrylic beads to separate the two chambers in the ML-MFC; (2) evaluate and compare the effect of two different types of inoculum on the performance of ML-MFC fed with actual domestic wastewater in terms of COD and power generation; and (3) determine the efficiency of ML-MFC for phosphate and nitrogen removal from actual wastewater.

2. Materials and methods

2.1. MFC construction and configuration

An up-flow mediator-less and membranes-less microbial fuel cells were designed and set up for the purposes of this study. The ML-MFC was made of polyacrylic plastic material consisted of two compartments, the anode at the bottom and the cathode at the top of the rectangular shaped bio-electrochemical reactor having side dimensions of 9.4×9.4 cm (Fig. 1(a)). The total height of the reactor was 60 cm, and the distance between the two compartments was 6 cm. A 4 cm thick layer of acrylic beads supported by 2 cm thick glass wool layer was sandwiched between two perforated glass sheets (Fig. 1(b)), and placed at the upper side of the anode chamber to separate the two compartments. Graphite plates, each of dimensions $5 \times 5 \times 0.5$ cm (MGM-carbon Industrial Company, China), were used as the anode and cathode electrodes. The apparent surface area of the anode was 60 cm^2. The fuel was supplied to the bottom of the anode and the effluent left the MFC through the cathode compartment at the top. The electrodes were connected with platinum wire through an external resistance of 300 Ω. The wastewater was continuously fed via a peristaltic pump at a flow rate of 2 cm^3/min, and then the flow rate was reduced to a constant value of 0.1 cm^3/min. The bio-chemical reactor was running with a total hydraulic retention time of 30 h.

2.2. Wastewater

Actual domestic wastewater was utilized in this study to feed the mediator-less and ML-MFC. The raw wastewater was obtained from the outlet of the main sewer pipe, Al-Kut city. The freshly collected

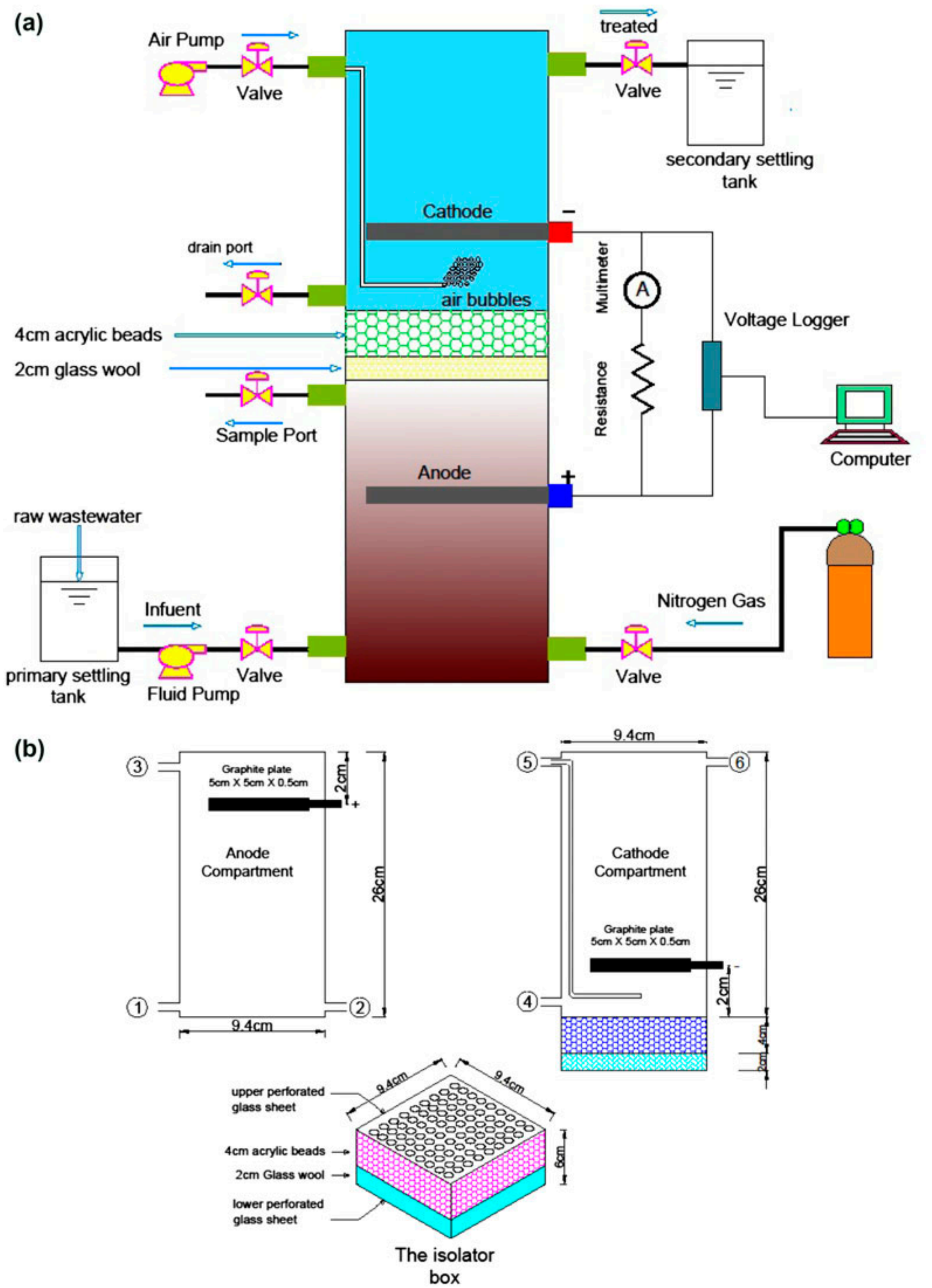

Fig. 1. Schematic diagram of ML-MFC; (a) the general layout of ML-MFC, (b) the detailed design of the anode and cathode chambers.

wastewater was primarily clarified and settled before entering the ML-MFC. The composition of the primarily clarified actual wastewater performed in this study with their constituent average concentration expressed in (mg/L) is as follows: BOD (200), COD (380), total phosphorus (12.8), nitrate nitrogen (9), ammonium nitrogen (22), chloride (30), sulfate (200), and pH (7.2).

2.3. Inoculums for the ML-MFCs

The mediator-less and ML-MFCs were used for enrichment of electrochemically active organisms originated from two different sources. Anaerobic aged sludge was collected from a local septic tank deep bottom, and an activated sludge freshly collected from the biological reactor of a local sewage treatment plant, Baghdad, Iraq. The ML-MFCs were alternatively inoculated with 1,000 mL of activated sludge and anaerobic aged sludge.

2.4. Conditioning stage

To start up the process, the anodic chamber was flushed with nitrogen, and then 1,000 mL of the freshly collected activated sludge was placed in the anodic compartment of the ML-MFC without aeration for 7 d in order to favor the enrichment of the cultures. The wastewater was not pumped into the MFC during this conditioning period, thus the only substrate available for the micro-organisms was obtained from the endogenous metabolism. After 7 d, the ML-MFC was fed with actual wastewater, at the same time the air was injected through the cathode chamber. Dissolved oxygen concentrations were measured in the anodic and cathodic compartments. The measured values of dissolved oxygen in the cathodic chamber indicated saturated concentrations. On the contrary, in the anodic chamber, the oxygen concentration was observed to be very low indicating anaerobic conditions in this compartment. An external resistance of 300 Ω was placed between the anode and cathode. Similar procedure was implemented for the ML-MFC inoculated with anaerobic aged sludge.

2.5. Measurements and analysis

The potential was measured using a multimeter (Keithley Instruments, Inc., Cleveland, OH, USA) and recorded every 1 min to a personal computer through a data acquisition system current and voltage data loggers (Lascar Electronic UK). The COD values were measured using a thermo-reactor type CR 2,200 & WTW photo lab S6. The BOD values were determined using WTW, OxiTop® IS 6/IS 12. The dissolved oxygen (DO) concentration, pH, and conductivity were measured using portable meters type ProfiLine Oxi 315i, ProfiLine pH 315i, and ProfiLine Cond 315i, respectively. PO_4^{-3}, NO_3^-, NH_4^+, Cl^-, and SO_4^{-2} were measured according to the *Standard Methods* [17]. Measurements were carried out in triplicate, and the given results represent the mean of three measured values.

3. Results and discussion

3.1. Performance of the ML-MFCs in terms of COD removal

Micro-organisms used in the mediator-less MFC typically have electrochemically active redox enzymes such as cytochromes at out membranes that can transport electrons to external materials [18]. ML-MFC alternatively inoculated with the anaerobic aged sludge and activated sludge was operated at initial COD concentrations in the range of 325–380 mg/L. After a time period of 26 d operation, a steady state condition was achieved in the ML-MFC inoculated with the anaerobic aged sludge and a percentage reduction of COD up to 81% (Fig. 2) was achieved. Similar profile for COD reduction was observed in ML-MFC inculcated with activated sludge. A maximum percentage removal of COD—79%, was achieved after 29 d operation. The slight difference in COD removal efficiencies is probably due to the variations in the inoculum quality in terms of the active micro-organisms. These results are in a good agreement with

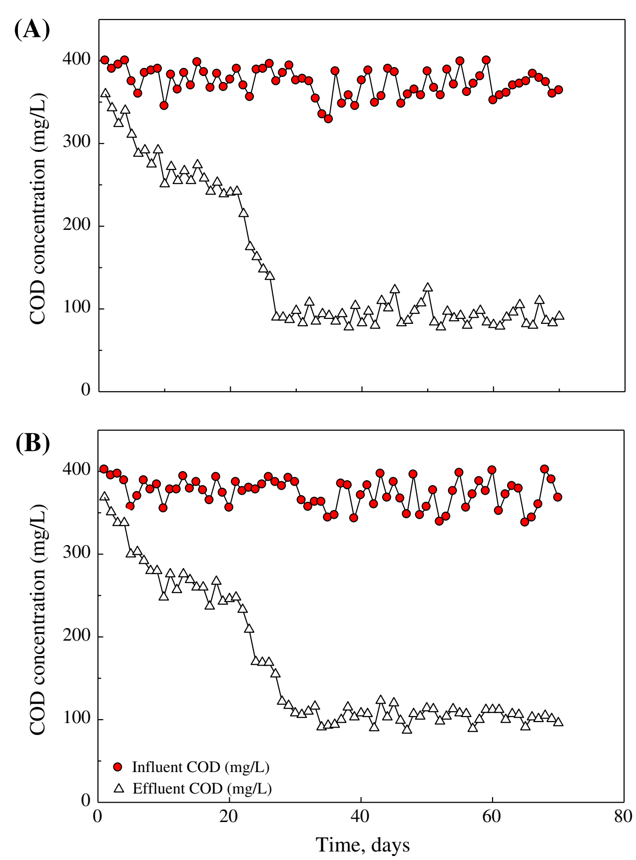

Fig. 2. COD removal with time in a membrane-less microbial fuel cell, (A) anaerobic sludge, (B) activated sludge.

those previously reported by Lobato et al. [19] regarding the evaluation of aerobic and anaerobic sludges as start-up material for MFC systems. However, the observed COD removal efficiencies were comparable to the previously reported efficiencies in the ranges of 80–90% [4,8,20–22] in spite of the fact that the range of values published in these literatures were observed for synthetic wastewater. This indicates that the presence of soluble and suspended species normally found in wastewater did not have significant effects on the ML-MFC performance in terms of COD removal.

On the other hand, BOD removal efficiencies were up to 60 and 70% for ML-MFC inoculated with activated sludge and anaerobic aged sludge, respectively (Fig. 3). The efficiencies of BOD removal are relatively lower than 78 and 86% reported by Liu et al. [20] and Ghangrekar and Shinde [4], respectively, for ML-MFC operated with synthetic wastewater and inoculated with sewage sludge. This alteration could be attributed to fact that the compositions of actual wastewater are normally different than synthetic wastewater. Real wastewater may contain components that affect the activity of micro-organisms and consequently the MFC performance. Also, the geometric design of MFC, type and material of electrodes, as well as the source of consortium may significantly affect the general performance of MFCs. However, the overall efficiencies observed for COD and BOD in this study indicate an effective wastewater treatment process.

3.2. Power generation

The performance of the ML-MFCs was evaluated in terms of current generation, voltage, and power density. For the ML-MFC inoculated with anaerobic sludge, the current increased rapidly for the first 20 d, followed by a slow increase for about 5 d, and then a constant maximum value of 0.38 mA was observed (Fig. 4). The current was maintained stable for more than 70 d under the given conditions; the open-circuit potential was about 0. 61 V and the maximum closed-circuit voltage drop across continuous external resistance 300 Ω was 0.23 V. For the ML-MFC inoculated with activated sludge, the current increased rapidly for the first 10 d, and then the current increased in a slower rate for the next 19 d up to a maximum constant value of 0.32 mA and was recorded as shown in Fig. 4. The current was maintained stable for over 70 d under the given conditions, and the open-circuit potential was about 0.58 V whereby the maximum closed-circuit voltage drop across the continuous external resistance 300 Ω was 0.20 V. Maximum power density of 21.9 and 17.0 mW/m^2 were obtained for the ML-MFC inoculated with anaerobic sludge and activated sludge, respectively. These results are comparable to the previously reported values of power densities estimated for continuously operated dual-chambered ML-MFCs as given in Table 1 taking into consideration that the effect of substrate source and concentration, inoculum source, electrode type, and MFC geometry could significantly affect the MFC performance. However, the power density values observed in this study are relatively higher than those reported by Ghangrekar and Shinde [4] and Jang et al. [8] although they used ML-MFCs having comparable systems as in this study with different type of substrate.

3.3. Influence of external resistance

The current was recorded with different resistances across the anode and cathode to determine the

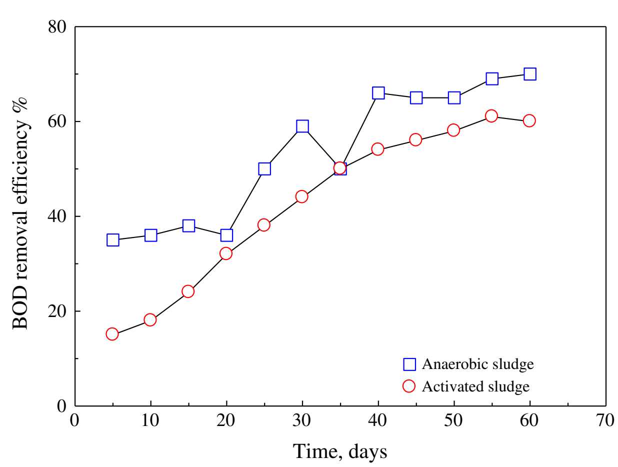

Fig. 3. BOD removal efficiency the in membrane-less microbial fuel cell.

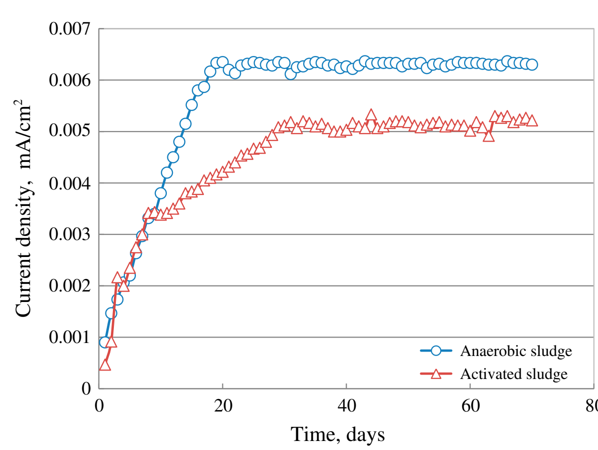

Fig. 4. Current generation with time.

Table 1
Reported values of power densities estimated for continuously operated dual-chambers MFCs

Substrate	Inoculum	Maximum power density (mW/m^2)	Types of separator between the two compartments	References
Synthetic wastewater	Activated sludge	1.30	Glass beads	[8]
Synthetic wastewater	Activated sludge	11.0	Porous graphite plate	[14]
Actual domestic wastewater	Activated sludge	17.0	Acrylic beads	Present work
Synthetic wastewater	Anaerobic sludge	10.9	Glass beads	[4]
Synthetic wastewater	Anaerobic sludge	26.34	Separated chambers with suitable tubing	[7]
Synthetic wastewater	G. sulfurreducens	24.33	Separated chambers with channel	[27]
Actual domestic wastewater	Anaerobic sludge	21.6	Acrylic beads	Present work

relationships between the current and power density as well (Fig. 5(A) and (B)). At low resistance, the electrons move more easily through the circuit than at high resistance, oxidizing electron carriers of the micro-organisms in the anode. Higher substrate oxidation by the micro-organisms is well expected with the high ratio of oxidized electron carriers of the culture at low resistance. Thus, this bio-electrochemical reactor can be operated at a low resistance to remove organic contaminants at a high rate. The variation of voltage observed across the variable external resistance is presented in Fig. 5(C). When increasing the external resistance, an increase in the voltages was observed. At the same external resistance of 2,000 Ω, the maximum obtained voltages were up to 0.28 and 0.47 V for ML-MFC inoculated with activated sludge and anaerobic sludge, respectively. Taking into consideration similar operating conditions, the difference between the observed voltages could be due to the effect of the inoculum type, probably its content of electro-active micro-organisms.

3.4. Polarization curves

In the present study, the relationships between the cell voltage and the power densities as a function of the cell current densities for the ML-MFCs were investigated. The power output from a certain MFC was a function of the circuit load, using a periodical increase in the external variable resistor. The voltage of the circuit was recorded and converted to power densities and the current was calculated using Ohms law based on the cell voltage. To determine the profile of the polarization curve for ML-MFC inoculated with anaerobic sludge, a range of variable resistances from 25 to 2,300 Ω was applied, maximum power density of 21.6 mW/m^2 and a current density of 60 mA/m^2 at external resistance of 1,000 Ω were obtained as shown in the polarization curve plot (Fig. 6).

However, these results are potentially favorable compared to a previously reported power density of 10.9 mW/m^2 at external resistance of 900 Ω produced by ML-MFC inoculated with anaerobic sludge collected from septic tank bottom, and fed with sucrose-based synthetic wastewater [4].

For ML-MFC inoculated with activated sludge, a wide range of variable resistances from 25 to 2,300 Ω was applied. From the polarization curve plot illustrated in Fig. 7, maximum power density and current density of 17 mW/m^2 and 81.34 mA/m^2, respectively, were obtained at an external resistance of 400 Ω. The observed power density for this ML-MFC is significantly favorable compared to power density of 1.3 mW/m^2 and current density of 9 mA/m^2 at external resistance of 100 Ω reported by Jang et al. [8] for a ML-MFC fed with synthetic wastewater.

The internal resistance can be calculated from the slope of the polarization curves. Accordingly, the internal resistance values are 833 and 167 Ω for ML-MFCs inoculated with anaerobic sludge and activated sludge, respectively.

3.5. Granulation of biomass in ML-MFCs

The biomass granules formation was visible in the anode chambers. After 70 d operation, the ML-MFCs

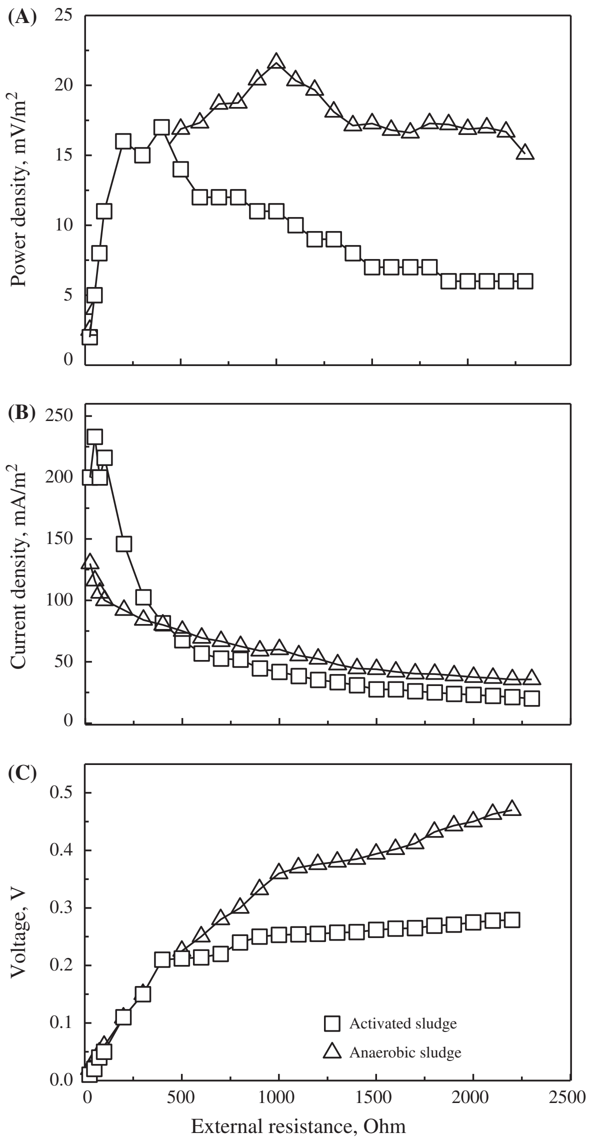

Fig. 5. Variation of external resistance with (A) power density, (B) current density, (C) voltage.

were emptied and small portions of each biofilms were scratched from the electrodes surfaces and collected for microscopic examination from anode chamber. The granulated anodic biofilms of activated sludge source had a typical color of dark gray, whereby, the granulated anodic biofilms sourced from anaerobic septic tank sludge were black; both were partially granulated having 1–2 mm size of granules. The SEM images (Fig. 8) of these biofilms revealed their porous and spongy structure with few on the surface. Similar observations relative to the type of biofilms were previously reported [4,23]. They suggested that the porous structure of granules with

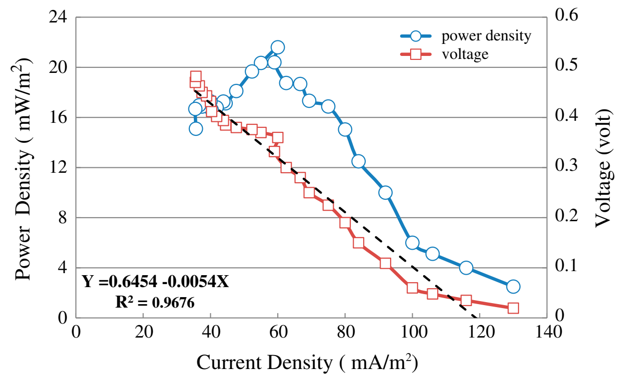

Fig. 6. Polarization curves for ML-MFC inoculated with anaerobic sludge.

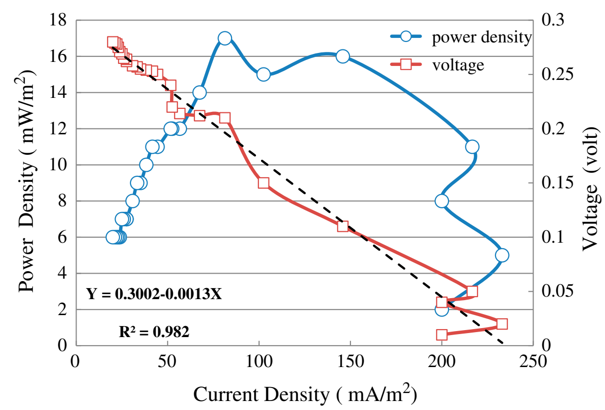

Fig. 7. Polarization curves for ML-MFC inoculated with activated sludge.

multiple cracks on the surface is likely to facilitate the passage of nutrients and substrates as well as the release of hydrogen, which had a very limited solubility of 1.58 mg/l in water, thus these granules did not exhibit a layered structure because of the simplicity of the acidification process.

3.6. Phosphate and nitrogen removal in ML-MFC

During the evaluation of ML-MFCs performance, phosphate removal of 71 and 78% was observed in MFC inoculated with activated sludge and anaerobic sludge, respectively. These results are in a good agreement with the previously published investigations. Ichihashi and Hirooka [24] reported 70–82% removal of phosphorus in the air-cathode single-chamber MFCs operated with swine wastewater.

Nitrogen in the actual wastewater was observed in the forms of NH_4^+ and NO_3^-. Removal efficiencies of

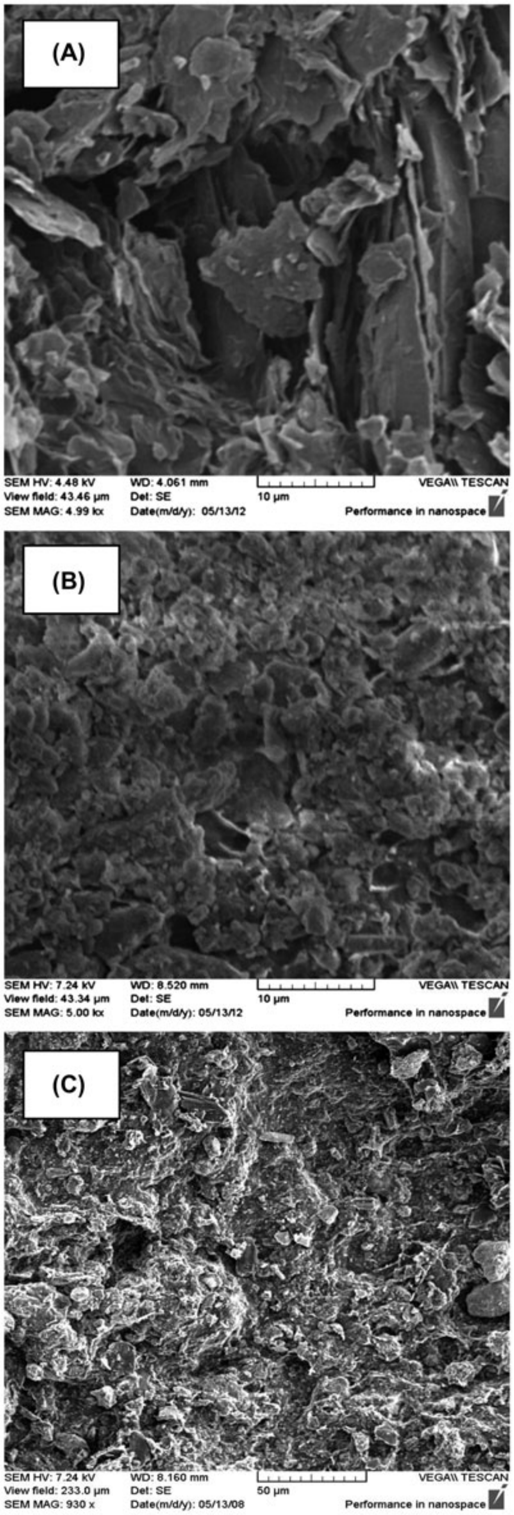

Fig. 8. SEM image for anode surface, (A) before biomass granulation, (B) and (C) after granulation of biomass for anaerobic sludge and activated sludge, respectively.

NH_4^+ and NO_3^- were up to 71 and 73%, respectively, in the ML-MFC inoculated with activated sludge. A relatively higher removal efficiency of NH_4^+ and NO_3^- up to 77 and 78%, respectively, were obtained in the ML-MFC inoculated with anaerobic sludge. Ghangrekar and Shinde [4] suggested that major removal of NH_4^+ occurs in the cathode chamber, which could be attributed to the ammonia stripping due to aeration, and the occurrence of simultaneous nitrification and denitrification in the cathode chamber. The diffusion of oxygen from the cathode to the anode chamber may slightly contribute to the removal of NH_4^+ in the anode chamber in spite of the anoxic conditions in this section. The initial concentration of NO_3^- assumed to be reduced in the anode chamber by denitrification process under anoxic conditions when oxygen levels are depleted and nitrate becomes the primary oxygen source for the facultative heterotrophic bacteria. However, the residual low concentration of NO_3^- in the treated effluent is almost due to the conversion of NH_4^+ to NO_3^- by nitrification process in the cathodic compartment. Virdis et al. [25] suggested a removal efficiency of total nitrogen up to $94.1 \pm 0.9\%$ by a simultaneous nitrification, denitrification, and carbon removal in a MFC that consisted of two compartments separated by CEM and filled with granular graphite as the anode and cathode materials. Kunthe et al. [26] reported the recovery of ammonium in two-chamber MFC at a rate of $3.29\,\mathrm{g\,N\,d^{-1}\,m^{-2}}$ which was achieved at a current density of $0.50\,\mathrm{Am^{-2}}$ from synthetic and real urine. The dissimilarity of results regarding the performance of MFCs with respect to nitrogen and phosphate removals and recovery could be attributed to the type and design of MFC, the existence or absence of membrane, as well as the type of fuel or substrate and the source of inoculum.

3.7. Prospects of acrylic beads application

One of the most commonly used components in MFCs is the CEM that is selective for transporting protons and other small cations across the membrane. However, the high cost of CEMs, their potential biofouling, and the associated high internal resistance could eliminate the power generation and limit the practical use of MFCs [5]. Therefore, several studies have designed new types of MFCs by removing the CEM or using low-cost materials as given in Table 1. In the membrane-less microbial fuel cells (ML-MFCs), the application of several types of separators including but not limited to glass beads, glass wool, perforated polyacrylic plate, porous carbon separator, and glass fiber separator have been studied during recent years.

However, none of the previous studies concerned of using acrylic beads as a separator between the anode and cathode. The significance and beauty of these acrylic beads is being inexpensive, non-biodegradable, long life and light weight materials, and the most important being non-conductive with a electrical resistivity of 10^{16} Ω cm (ASTM D257) compared to <30 Ω cm^2 for CEM (CMI-7000). Also, less fouling was expected to occur on acrylic beads compared to other materials due to their smooth and slick surfaces preventing undesirable matters to be settled and stabilized on the acrylic beads' surfaces.

Further work is being conducted to study the performance of ML-MFCs with separators consisted of non-uniform shaped and sized acrylic beads. The outcome may affect the performance of the ML-MFCs compared to the currently tested uniform acrylic beads.

4. Conclusion

This work demonstrates successfully the simultaneous wastewater treatment and energy production from real domestic wastewater using a mediator-less two-chamber ML-MFC alternatively inoculated with activated sludge and anaerobic aged sludge. The study evaluated the performance of an up-flow dual-chambered ML-MFC in the presence of acrylic beads as a separator between the anode and air cathode compartments. Results from the proposed ML-MFC configuration revealed that domestic wastewater is a potential renewable resource to produce electricity. In this study, maximum power density of 21.9 and 17.0 mW/m^2 were obtained for ML-MFC inoculated with anaerobic sludge and activated sludge, respectively. The use of inexpensive light weight acrylic beads in ML-MFC can improve the economic and feasibility application of MFC in large-scale wastewater treatment, since it eliminates the use of membrane to minimize the high initial cost and fouling problems of membrane.

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