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Optimization of double chamber microbial fuel cell for domestic wastewater treatment and electricity production

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Abstract：Microbial fuel cells（MFCs）represent a new approach for treating waste water along with electricity production. The present study addressed electricity production from domestic wastewater using a mediator-less double chamber MFC. The electricity production was monitored under different operational conditions for both summer and winter samples. Optimization of the anodic and cathodic chambers resulted in a maximal current of 0.784 and 0.645 mA with the maximal power intensity of 209 and 117 mW/m^2 in power duration of 24 h for the summer and winter samples，respectively. Scanning electron microscopy showed that the bacterial biofilm formation on the anode was denser for the summer sample than that when the winter sample was used，so was the total bacterial count. Therefore，samples taken during summer were considered better in electricity production and waste water treatment than those taken during winter basically because of the high microbial load during the hot season. In parallel，there was a decrease in both biological oxygen demand（BOD$_5$）and chemical oxygen demand（COD）values which reached 71.8% and 72.85%，respectively at the end of the operation process for the summer sample，while there was no evident decrease for the winter sample. Optimizing the operating conditions not only increased the potential of using domestic waste water in microbial fuel cells to produce electricity，but also improved the quality of the domestic waste water.

Keywords：microbial fuel cell；domestic wastewater treatment；operating conditions；electricity production

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The sharp rise in the industrialization and motorization urgently demands developing new alternative energy sources because current reliance on petroleum-based fuels is unsustainable due to their limited resources. The development of environmentally friendly alternative technologies is imperative to avoid supply problems and contributions to global warming. Biochemically produced electricity is perceived as a promising alternative sustainable technology. The electricity generated is a result of the oxidation of different organic wastes using degrading bacteria[1] into CO_2 and H_2O. Dual chamber MFC is composed of adjacent anodic and cathodic champers separated by a proton exchange membrane. Electrons produced by bacteria from the organic substrates migrate to the anode and flow to the cathode trough a conductive material producing electricity[2]. Microbial fuel cells（MFCs），upon these bases，represent ideal technology for not only harvesting renewable bioelectricity but also using for treatment of many types of wastewater[3] such as brewery wastewater[4]，starch processing wastewater[5]，dye wastewater[6]，domestic wastewater[7]，palm oil mill effluent[8] and landfill leachates[9] or to degrade a single compound[10]. The aspect of simultaneous pollutant treatment and power generation acquires MFCs for their green characteristics. Municipal wastewater contains a massive amount of organic materials that can be used as fuel for MFCs.

Polymers are economical material according to their nature and chemical structure，and possess interesting characteristics，such as high chemical resistance and superior thermal stability，However，they do not have much in the way of functionality，such as electrical conductivity，aqueous swelling and ion-exchange capacity. Various approaches have been made to modify the structure of polymeric backbone in order to impart various desirable properties to employ the modified polymers in advanced practical applications. Among these approaches，grafting is a useful technique for designing new materials that not only retain most of their original characteristics but also acquire additional properties of the grafted moieties. The radiation-grafting technique is one of the preferable methods because of the uniform and rapid creation of active radical sites. In addition，it is independent of the chemical nature of the polymer. It can be applied at a wide range of temperatures and easily regulated by controlling the reaction conditions. The produced radiation functionalized polymers can

be used in different field, such as proton exchange membranes, chelating polymers, conducting polymers, etc[11,12].

Proton-exchange membrane (PEM) is a key constituent of MFC, which is responsible for proton migration from the anode to the cathode. PEMs exhibit several advantages over liquid or solid inorganic electrolytes, such as high proton conductivity, good chemical, thermal and mechanical properties. Sulfonated aromatic polymer membranes have been given attention owing to their high thermal stability as well as excellent barrier properties against fuels (methanol, H_2) and oxygen[13~15]. On the other hand, the radiation functionalized polymer can be converted into conducting polymer used as electrode, which is another key constituent of the MFC, via the deposition of highly conduction metal such as silver within the grafted polymer.

The aim of the present study was to produce electricity during the biochemical treatment of domestic wastewater. To achieve such purpose, a double chamber mediator-less microbial fuel cell was constructed via the assembly of two glass bottle and radiation developed proton exchange membrane and polymer electrode. For the maximum electricity production from two different climatic seasons of winter and summer, the anodic and cathodic operation factors, such as temperature, pH value, add carbon source, the presence and amount of ferricyanide as electron acceptor, will be optimized.

1　Experimental

1.1　Domestic wastewater

Two samples of domestic wastewater were collected from Al Gabal Al Asfar stage-2 station wastewater treatment plant (WWTP), Cairo Governorate, Egypt, where wastewater aggregated from inlet working pumping station (IWPS) stage only with mechanical treatment. Two samples were taken, one during September representing the hot season and the other during January representing the winter season in Egypt.

1.2　Chemical characteristics of raw and treated domestic waste water

Chemical tests were carried out for both summer and winter wastewater samples before and after treatment as follows. Biological Oxygen Demands (BOD$_5$) was determined according to Young and Baumann[16]. Chemical Oxygen Demands (COD) was measured according to Annual Book of Standards[17]. Total suspended solids (TSS), total dissolved solids (TDS), total hardness, Phenol, oil and grease were all performed according to Eaton et

al[18]. Heavy metal (Iron, Cobalt, Copper, Zinc and Manganese) concentrations were measured using Perkin-Elmer Model 5000 atomic-absorption spectrometer utilizing air-acetylene flame atomic absorption spectrometry[19].

1.3　Configuration of microbial fuel fell

Double chamber MFC was consisted of two 300 mL bottles (anode and cathode) containing each a silver electrode with a surface area of 10.7 cm × 5.1 cm totally immersed in the anode and cathode solution. The bottles were joined by a glass bridge held by a para film between the flattened ends of the two glass tubes. The glass bridge contained a 3.5 cm diameter hole which was covered with proton exchange membrane (PEM) for proton transport. Electricity production was measured in milli ampere (mA) against time (min).

1.4　Preparation of proton exchange membrane using gamma irradiation

Low density polyethylene (LDPE) films were thoroughly washed with methanol and dried in a vacuum oven at 50 ℃ for 24 h. Samples were weighed and then immersed in glass ampoules containing styrene (Sty) solution. Ferric chloride as inhibitor was introduced in the reaction mixtures. The reaction mixtures were deaerated by bubbling of nitrogen gas for 4~7 min, sealed and then subjected to ^{60}Co gamma rays. The obtained grafted samples were removed and washed thoroughly with methanol to get rid of the formed homopolymer. The obtained grafted samples were dried in a vacuum oven at 60~70 ℃ for 24 h and weighed. The graft percentage was determined by the percent increase in weight. The low density polyethylene grafted with polystyrene LDPE-g-P(Sty) films were washed with dichloromethane, soaked for 30 min, removed and dried in an oven for 24 h. The films were sulfonated using chlorosulfonic acid solution in 1, 1, 2, 2-tetrachloroethane at room temperature for 3 h. The sulfonated grafted films were removed and thoroughly rinsed with 1, 1, 2, 2-tetrachloroethane and dichloromethane in order to remove the excess of chlorosulfonic acid. The obtained membranes were neutralized with 0.5 mol/L KOH solution overnight and regenerated by boiling with 1 mol/L hydrochloric acid for 2 h. The membranes were thoroughly washed with deionized water in order to ensure complete removal of acid and finally stored in the sealed bottles at ambient temperature[20].

1.5　Development of polymeric electrodes

The graft copolymers were prepared by direct radiation grafting of styrene/maleic anhydride (Sty/

Man) binary comonomer system onto LDPE using [60]Co γ-rays. The low density polyethylene grafted with poly (polystyrene/maleic anhydride) binary LDPE-g-P (Sty/MAn) films were removed and washed thoroughly with acetone to extract the residual monomers and the homopolymer which may have accumulated on the film. The films were dried in a vacuum oven at 50 ~ 60 ℃ for 24 h and weighed. The anhydride groups of LDPE-g-Sty/MAn copolymer were converted into maleic acid disodium salt when treated with sodium hydroxide and converted into maleic acid when treated with HCl to obtain high chelating ability[21].

The chemical deposition of silver within the functionalized LDPE sheets was achieved through successive reduction process of $AgNO_3$ chelated by the grafted polymer using sodium borohydrate as a reducing agent. In brief, the grafted LDPE sheets were soaked in 0. 1 mol/L $AgNO_3$ solution for 2 h. The chelate sheets were immersed in 10% aqueous borohydrate solution at room temperature for 5 min. The above mentioned reduction process was repeated three times to increase the amount of the deposited Ag and to ensure the homogeneity of the silver deposition.

1.6 Operation of microbial fuel cell

All the upcoming experiments were performed in batch mode for both summer and winter samples.

1.6.1 Optimization of the operation conditions

Domestic wastewater (300 mL) was used as the source of bacterial consortia and medium in anode chamber. The anodic chamber was closed with a tight lid to prevent any passage of oxygen. To optimize the conditions in the anodic chamber, different parameters, that may affect the electricity production including operation temperatures, the presence of different carbon sources and operation pH values, were tested. The cathode chamber (aerobic chamber where oxygen was used as electron acceptor for the electrode) was filled with only 250 mL of potassium phosphate buffer. To optimize the operating conditions for the cathode chamber, the effect of ferricyanide addition (25, 50 and 100 mM in the presence of 100 mM phosphate buffer) was investigated.

1.6.2 Monitoring electricity

The electric current (mA) of the microbial fuel cell during each experiment was measured by a digital multi meter (Model number DT-3900). Readings were taken at 0, 30, 60, 90, 120, 150, 180, 210 and 240 min for all experiments unless stated otherwise. At the end of the optimization process, the voltage was measured in mV and the power intensity was obtained in mW/m^2 under optimized conditions for both summer and winter samples. The reading was taken until the power reached a plateau or declined. All experiments were repeated at least 3 times to ensure consistency of the reported results and the results were represented as average values.

1.7 Biofilm formation on the anode and total bacterial load

At the end of the MFC operation time, the anodic electrode was used to monitor biofilm formation for both summer and winter samples. A strip was carefully cut from the anode in each case using sterile forceps and scissors. The strips were left to dry in air and were glued on metal stubs using a double sided adhesive tape and were coated with a thin layer of gold under reduced pressure. The images were captured using JEOL JMS 5600 scanning electron microscope at magnifications of 7 500 × using an electron beam high voltage of 30 kV. At the end of the MFC operation for both samples, a serial dilution was performed. 0. 1 mL of the cell suspension was spread over Nutrient Agar plates (NB) (Oxoid) which were incubated at 37 ℃ for 24 h. Total bacterial colonies were counted and dilution factors were applied to obtain CFU/mL values.

2 Results and discussion

Microbial fuel cells were considered a very practical approach to counteract the diminishing power. The advantages were doubled if waste water was used, and thus waste water treatment came hand in hand with electricity production[22]. One of the most commonly used MFCs in the laboratory was consisted of an anode chamber and a cathode chamber being separated by a proton exchange membrane (PEM).

Electrodes that represent essential component of the MFC was developed via the chemical deposition of Ag onto radiation modified LDPE. The preparation and characterization of LDPE-g-Sty/MAn were carried out according to our previous work[21]. The conductivity (resistivity) of the silver plated electrode was found to be 3. 06 MΩ. This value was observed to be steady throughout the whole operation process, which allowed the use of Ag plated LDPE-g-Sty/MAn as electrode in MFC. Such electrode possessed low cost, flexibility, good mechanical properties of the LDPE as well as the excellent electrical conductivity of silver. On the other hand, the use of a costly proton exchange membrane (PEM), the other essential component of the MFC, would result in an expensive technology to hinder the application process[23]. Therefore, a new PEM was prepared

using radiation grafting technique as one of the most promising tools for the modification of polymeric materials to produce high performance polymer electrolyte membrane（PEM）. The characteristic proton exchange properties of the prepared sulfonated LDPE-g-Sty such as hydration number, ion exchange capacity and proton conductivity were investigated and submitted for publication .

2.1　Optimization of MFC operation conditions

The performance of a microbial fuel cell can be influenced by several factors. The chemical constituents to be added to the anodic and cathodic champers and the optimization of some physical parameters such as incubation temperature and pH value would ensure the maximum electricity production. The optimum of each of the abovementioned parameter will be investigated independently first and then the electricity production process will be assessed later under optimized conditions.

To find out the optimum operation temperature of the MFC[3], the produced current was monitored as function in the incubation temperature of the anodic chamber. Figure 1 describes the time dependent current production from a mediator-less MFC at different operation temperatures of 20, 25, 30 and 37 ℃ for both summer and winter samples. In general, summer sample is found to produce much higher current than winter one. The obtained data（Figure 1b）show that winter sample requires about 30 min to reach its maximum production of as low as 0. 08 mA at 25 ℃ and such current is immediately declined. On the other hand, summer sample（Figure 1a）, which also indicates low current production at 20 and 25 ℃, shows high current production at 30 ℃ and such value is reached within 30 min and kept almost stable for 240 min. Moreover, summer sample exhibits a maximal current production of 0. 27 mA within 150 min and keeps stable. The results clearly show that the microbial consortia display the highest activity and consequently attain higher electricity production at their initial temperature for both the summer and winter samples.

Carbon sources act as energy source for biological process which influence not only the integral composition of bacterial community in anode chamber, but also the MFC performance. The addition of carbon sources, such as glucose, fructose, sucrose and maltose in this study, should have resulted in faster growth rates of bacteria. Figure 2 represents the effect of time on the current production as a function of the carbon source. The use of different carbon sources proves that there could be an

increase in current production for the summer sample with a maximum of 0. 3 mA reached when 5 mM sucrose is added to the domestic waste water in the anode（Figure 2（a））. On the other hand, the current obtained for the winter sample was 0. 18 mA when 5 mM glucose was added to the domestic waste water. The other carbon sources used in the current study fail to increase the current（Figure 2（b））. Ieropoulos et al[24] stated that under the same external circuit condition, the highest power and current output was given by MFCs supplemented with sucrose, while glucose, fructose and maltose suppressed bacterial growth.
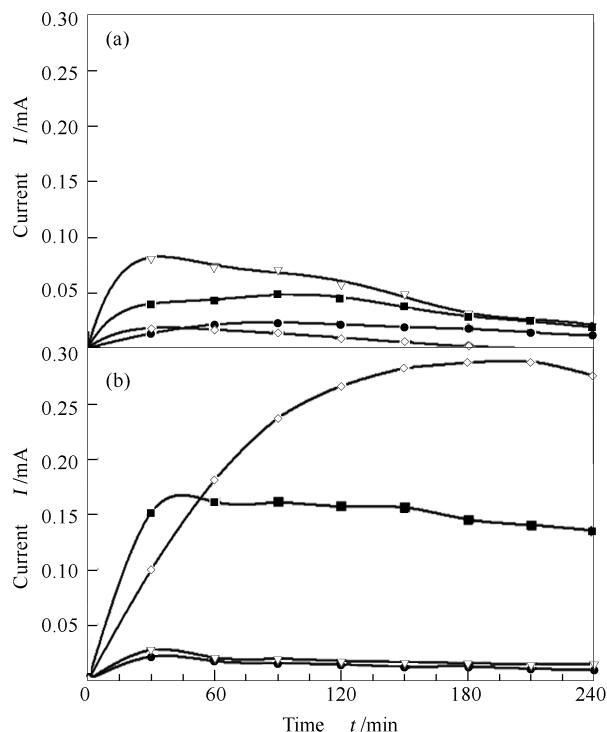

Figure 1　Effect of time on the current produced using domestic waste water; （a）winter sample and （b）summer sample at different temperatures; ●: 20 ℃; ▽: 25 ℃; ■: 30 ℃; ◇: 37 ℃

The pH value was also found to directly influence the metabolic rates of different microorganisms and consequently affecting the e⁻ and H⁺ generation mechanism. The effect of pH value in the anodic chamber for both samples was investigated to find out the pH value at which the optimum MFC operation took place. Figure 3 shows the effect of pH values within close ranges to those of the initial value on the current production from a mediator-less MFC.

The results indicate that the highest current values are 0. 28 and 0. 2 mA attained at pH value 8. 6 and 7. 9 for the summer and winter samples, respectively, i. e. , the maximum activity of the

microbial consortia is found in their original pH value. Generally, the bacteria responds to the changes in the internal and external pH value by adjusting their activity associated with many different processes including proton translocation, amino acid degradation, adaptation to acidic or basic conditions and virulence[25]. Depending on the organism and growth conditions, changes in the external pH value can bring about alterations in several primary physiological parameters including internal pH value, concentration of ions, membrane potential and proton motive force[26]. This is in agreement with our results which show that the optimal current production is attained at the original pH values for each sample.
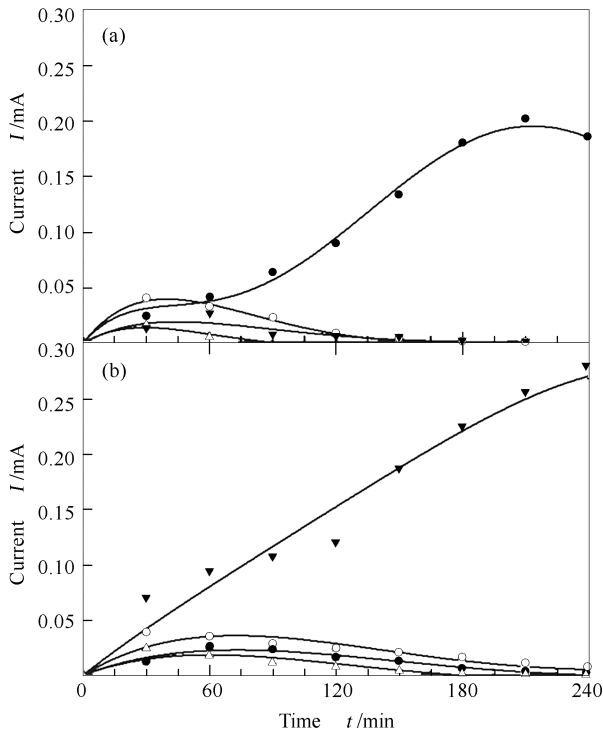

Figure 2　Effect of time on the produced current using (a) winter sample and (b) summer sample domestic waste water at different carbon source
●: glucose; ○: fructose; ▼: sucrose; △: maltose
carbon source concn. 5 mM

As an important component of MFC, the cathode has a great effect on the electricity generation characteristics. Currently, research is focused on studying the cathodic electron acceptor, the nature and type of the electrode and the catalyst on the electrode[27]. Dissolved oxygen, ferricyanide, potassium permanganate. or manganese dioxide have often been used as cathodic electron acceptors in two-chambered MFCs. Potassium ferricyanide is an excellent cathodic electron acceptor for MFCs and a high power generation can be obtained from this

system at low cost. Thus, the effect of potassium ferricyanide concentrations in the catholyte on the electricity generation characteristics of MFC is studied. The use of 50 mM ferricyanide in 100 mM potassium phosphate buffer results in an increase in current production (0.6 ~ 0.7 mA) over a period of 3 h when the summer sample is used (Figure 4).
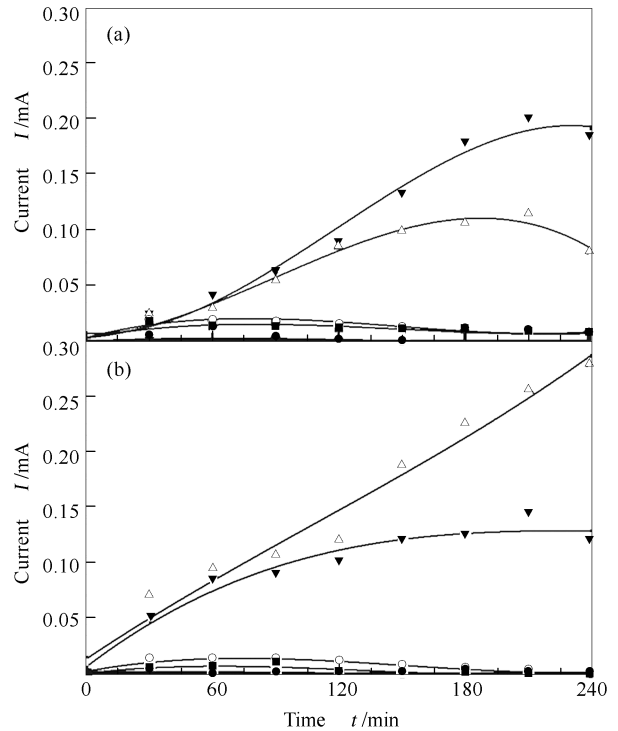

Figure 3　Effect of time on the current produced using domestic waste water; (a) winter sample and (b) summer sample at different pH value;
●: 2; ○: 5; ▼: 7; △: 8; ■: 11

While there is a lag period of current increase when the winter sample is used, the maximal current reaches 0.6 mA after 4 h when 25 mM ferricyanide is added to the same buffer (Figure 4). The use of ferricyanide as the electron acceptor in the cathodic chamber was reported to produce very high power outputs[28]. Wei et al[27] stated that ferricyanide passing through PEM to anode chamber resulted in osmotic pressure inhibition which in turn inhibited the movement of protons to the cathode chamber, decreasing the biodegradation of organic matter. It is observed that at 100 mM of ferricyanide concentration, the current production decreases, this is contrary to the results obtained by Rabaey et al[28] who claimed that high power density could be obtained from an MFC using 100 mM ferricyanide as the cathode mediator. Power generation from MFCs shows vast variation It depends on not only the type of wastewater used[29], but also the type of MFC

configuration. While Adelaja et al[10] obtained 1. 25 mW/m^2 using single microorganisms for simultaneous degradation of phenanthrene, Logan et al[30] got 39 mW/m^2 with a double chamber MFC using sediment samples. Xiao et al[31] stated that a power density obtained was varied between 46. 8 and 55. 88 mW/m^2 using alkaline pretreated sludge and that the production was changed greatly when using single or double chamber cells. Figure 6 represents a schematic diagram to summarize all the above mentioned conditions.
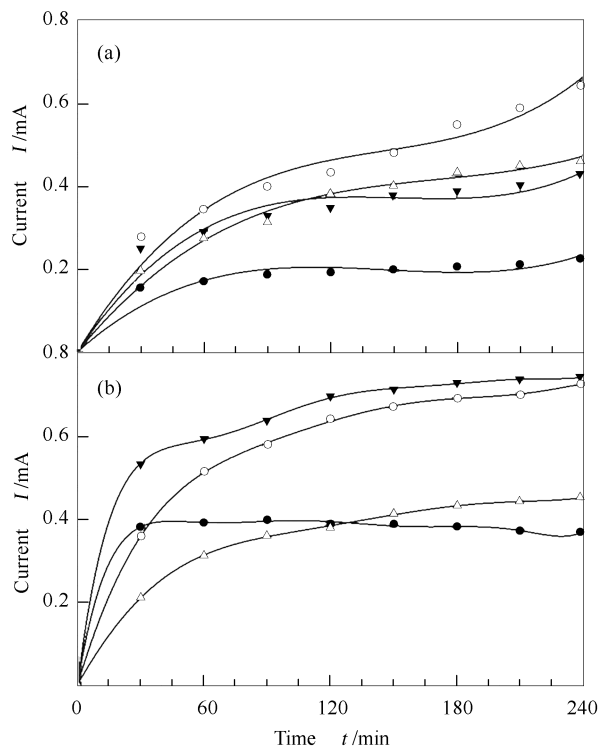

Figure 4　Effect of different ferricyanide concentrations；
●, ▼, ○, △ dissolved in 100 mM potassium
phosphate buffer pH value 7, using domestic waste
water for (a) winter sample and (b) summer sample

2. 2　Power intensity under optimized anodic and cathodic conditions

　　When the highest current values under optimized operation conditions were obtained, the power intensity was calculated. Figure 5 shows that the power increases to 202 and 117 mW/m^2 for summer and winter samples, respectively. The increasing area for the summer sample is broader in terms of time than that for the winter sample. The duration of power for the optimized summer sample achieves 120 h, while that of the winter sample declines and reaches a steady state within 24 h.

2. 3　Biofilm formation on electrodes

　　Bacterial growth on the anode is crucial to the current production. The current increase is correlated

to the increase of microbial biofilm formation on the anode and total bacterial count for the summer sample as compared to the lower levels for the winter sample. This is in agreement with You et al[32] who stated that lack of bacterial growth on the anode abolished the current production.
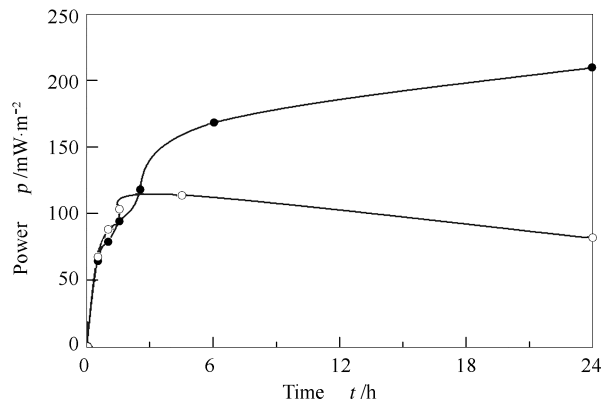

Figure 5　Obtained power (mW/m^2) under optimized conditions using domestic waste water for summer sample (a) and winter sample (b)
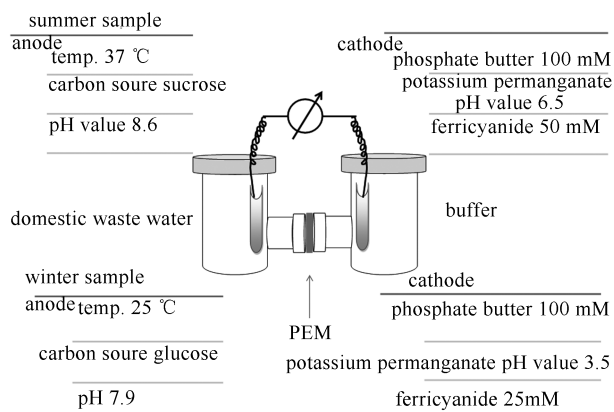

Figure 6　A schematic representation of the optimized conditions of the MFC under study for summer and winter samples

　　To analyze the relationship between electricity production and bacteria present in the domestic waste water, scanning electron micrographs were taken at the end of the operation process for both summer and winter samples. The results show that the total microbial load is 19×10^6 CFU/mL for summer sample compared with $14. 4\times10^4$ CFU/mL for winter sample. Figure 6 suggests that there are variations in the microorganisms adhering to the electrode. More rod shaped clumps are evident in the summer sample (Figure 7 (b)), while less cluster formations are found in the winter sample with Cocci in shape (Figure 7 (c)). A control electrode is used to represent the surface of the electrode free of microbial growth (Figure 7(a)).
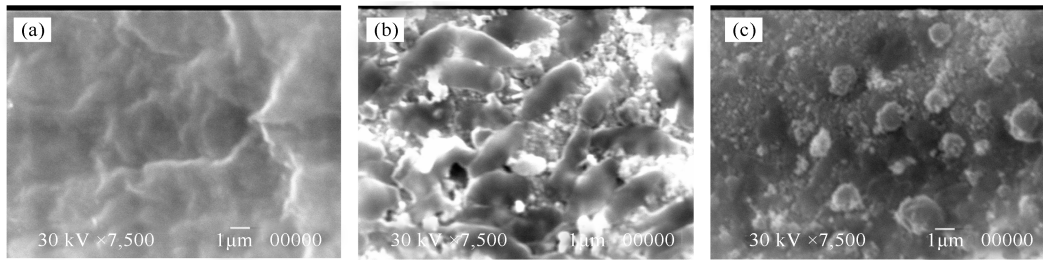
Figure 7   Scanning electron micrographs of the anode, pictures represent raw unused anode (a),
anode after the completion of MFC production process using summer sample (b) and winter sample (c)

2.4   Treatment of domestic waste water at the end of MFC operation

At the end of the operation process under optimized conditions, some chemical characteristics of the domestic waste water samples for both summer and winter seasons were analyzed and compared to those prior to operation. Table 1 represents both the results and the calculated percentage of reduction for each tested parameter. It is evident that there is a decrease in both BOD_5 and COD, which reaches 71.8% and 72.85%, respectively after using the summer sample as compared to minimal decrease when the winter sample is used for current production. TSS decreases to 15.48% and 8.45%, respectively. There is no great difference in total hardness for both samples. Fe, Co and Zn decreases to 95.37%, 75.5% and 99.99%, respectively in the winter. Mn decreases to 52.7% and 45.76% for summer and winter samples, respectively. There is no decrease in values for Cu in both samples. Total phenol content decreases to about 53.3% and 57.69% for summer and winter samples, respectively. Also the decrease in oil and grease is 99.9% for both samples.

Table 1   Change in chemical characteristics of domestic wastewater upon MFC treatment

Parameter	Summer sample			Winter sample		
	before	after	reduction /%	before	after	reduction /%
pH value	8.6	8.0	6.97	7.9	7.5	5.1
BOD_5 mg/L	160	45	71.8	40	40	0
COD mg/L	350	95	72.85	92	90	2.17
TSS mg/L	155	131	17.78	213	195	8.45
TDS mg/L	731	690	5.6	795	745	6.28
Total hardness mg//L	340	312	8.23	324	300	7.4
Phenol mg/L	0.6	0.28	53.3	0.52	0.22	57.69
Fe mg/L	0.115	0	100	2.748	0.127	95.37
Co mg/L	0	0	0	0.645	0.158	75.5
Cu mg/L	0.004	0	100	0.003	0	100
Zn mg/L	0.010	0	100	310	0.018	99.99
Mn mg/L	0.455	0.215	52.7	0.260	0.141	45.76
Oil and grease mg/L	4.8	0.002	99.9	2.3	0.0017	99.92

The same previously assayed parameters were performed for the treated waste water obtained at the end of the MFC process. It is obvious that the total bacterial load has an impact on the extent of treatment, especially for the BOD_5 and COD values which decrease after using the summer sample as compared to the winter sample. This process is probably controlled by temperature. Ahn and Logan[7] reported that COD was removed at 30 ℃ and decreased at ambient temperature of 23 ℃ for domestic waste water treatment. Moreover, the operation of the reactors under mesophilic conditions increased power output compared to those under ambient conditions.

3   Conclusions

The results demonstrated the optimization of anodic and cathodic conditions in order to obtain an increase in electricity production and power intensity. At the same time, the domestic waste water used was simultaneously treated. The electricity production and the optimization conditions showed the variation for both summer and winter samples. The high electricity production was attributed to the high microbial load of the sample taken during the hot summer season as compared to a lower count for the winter season. Moreover, high power intensity production, longer stability and higher treatment were observed for the

summer season than those for the winter season. It was our intention to follow up the spatial changes in the microbial flora under different operating conditions and to identify the key microorganism, which was responsible for both features to upgrade the MFC performance.

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