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Novel microbial fuel cell design to operate with different wastewaters simultaneously

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ABSTRACT

A novel single cathode chamber and multiple anode chamber microbial fuel cell design (MAC-MFC) was developed by incorporating multiple anode chambers into a single unit and its performance was checked. During 60 days of operation, performance of MAC-MFC was assessed and compared with standard single anode/cathode chamber microbial fuel cell (SC-MFC). The tests showed that MAC-MFC generated stable and higher power outputs compared with SC-MFC and each anode chamber contributed efficiently. Further, MAC-MFCs were incorporated with different wastewaters in different anode chambers and their behavior in MFC performance was observed. MAC-MFC efficiently treated multiple wastewaters simultaneously at low cost and small space, which claims its candidature for future possible scale-up applications.

Introduction

Microbial fuel cells (MFCs) hold a promising future in wastewater treatment as an emerging system, capable of removing contaminants and producing electricity simultaneously. These are the devices, which convert chemical energy of chemical compounds, directly into electrical energy using catalytic activities of microorganisms. The essential components of a MFC include: an anode, a cathode, an electrolyte medium that connects the two electrodes, an external circuit, and microorganisms (Cheng et al., 2006a; Mathuriya and Sharma, 2009a; Mathuriya and Yakhmi, 2014). A prototype two-chamber MFC has been studied most extensively (Cheng et al., 2006a, 2006b; Shukla et al., 2004; Mathuriya and Sharma, 2009b). This MFC consists of anaerobic anode and aerobic cathode chamber separated by a cation/proton exchange membrane. At the anode, microorganisms generate electrons through degrading organic compounds. Electrons travel through external circuit to cathode while protons through membrane. At the cathode, they react with oxygen to form water. Therefore, the electricity generated by MFC can be harvested by an external resistor placed between the anode and the cathode (Jiang et al., 2010).

Over the past few years, MFCs witnessed intense research and development and proved to be superior over other competing conventional wastewater treatment technologies, in many aspects, including enhanced conversion efficiency due to direct conversion of substrate's chemical energy into electricity; capability to treat low-strength wastewaters that are not suitable for anaerobic digestion (Rittmann, 2008; Watanabe, 2008); safe and quite performance; ability to operate at ambient temperature (Mathuriya and Sharma, 2009a); and generation of 50%–90% less solids to be disposed of (Du et al., 2007). Moreover, MFCs produce mainly carbon dioxide (CO_2) that has no useful energy content and comparatively less harmful, thus not requiring much further treatment (Jang et al., 2004). In recent years power densities in MFCs reached over 4200 mW/m^3 (Sharma and Li, 2010) and chemical oxygen demand (COD) and other contaminants removal up to 100% (Luo et al., 2011).

The structural parameters draw crucial effects on the overall performance of a fuel cell (Larminie and Dicks, 2000).

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In past, many MFC designs have been tested to increase power density or wastewater treatment efficiency (Kaewkannetra et al., 2011; Qian et al., 2011; Huang et al., 2012). In additions, various attempts have been made to minimize MFC operational costs (Buitrón and Cervantes-Astorga, 2013). Novel configurations *viz.* single-chamber (Yokoyama et al., 2006), column (Powers et al., 2011), tubular, (Rabaey et al., 2005) and high efficiency electrode materials (non-platinum coated cathodes, brush anodes, granular activated carbon anodes) have been developed till date (Rabaey et al., 2005; Cheng et al., 2006a,b; He et al., 2007; Logan et al., 2007; Zou et al., 2008; Sharma et al., 2008). However, most MFCs studies were conducted at lab scales (less than 1 L) and it was observed that the power density decreased during scale-up (Keller and Rabaey, 2008). In addition, these MFCs were able to treat only one type of wastewater at a time (Mathuriya and Sharma, 2009a; Rabaey et al., 2005). In order to make MFCs suitable for practical applications, it is critical to achieve high power density at large scale along with real time wastewater management capabilities. In present investigation, a novel multiple anode chamber and single cathode chamber MFC design (MAC-MFC) was fabricated which operated as a fed batch system to optimize power output from wastewater and its performance was compared with the standard single-anode/cathode chamber MFC (SC-MFC) for power production and chemical oxygen demand (COD) removal. Further, efficiency of MAC-MFC was studied with different wastewaters in each anode chamber to prove its ability in treating different wastewaters simultaneously, a possible situation in many waste treatment plants.

1. Materials and methods

1.1. Wastewaters

Dairy wastewater was collected from primary effluent collection tank from a local dairy plant at Agra, India. Potato wastewater was collected from local potato chip unit Agra, India. Paper wastewater was collected from a paper processing plant at Agra, India. The artificial wastewater was prepared by modifying previous method (Jang et al., 2004). The composition was (g/L): 15.0 g glucose, 450.0 mg $NaHCO_3$, 100.0 mg NH_4Cl, 10.5 mg K_2HPO_4, 6.0 mg KH_2PO_4, 64.3 mg $CaCl_2{\cdot}2H_2O$, 18.9 mg $MgSO_4{\cdot}7H_2O$, 10.0 mg $FeSO_4{\cdot}7H_2O$, 6.0 mg $MnSO_4$, 0.5 mg $ZnSO_4{\cdot}7H_2O$, 20.0 mg $CoCl_2{\cdot}6H_2O$, and 0.65 mg $CuSO_4{\cdot}5H_2O$. Spot samples of all wastewaters were transported to laboratory for physicochemical analysis. These parameters include pH, total dissolved solids (TDS), total suspended solids (TSS), volatile suspended solids (VSS), color, odor, COD, and biological oxygen demand (BOD). Each sample was left undisturbed for 24 hr at 4°C under anaerobic conditions to settle the solid particulate contents. Wastewater samples were kept in refrigerator at 4°C, when not in use. The plain wastewaters (without any modifications such as addition of nutrients, mediator, and any other microbial inoculum or trace metals) with constant COD value of 1500 mg/L were used as the inoculum for all MFC tests (except as indicated). COD values of various wastewaters were adjusted by diluting wastewaters with de-ionized water. Experiments were conducted at 30°C, pH 7.0 and stagnant condition (without stirring).

1.2. MFC designs

Standard single chamber MFCs (SC-MFCs) were constructed from two glass chambers with total inner volume of 3000 mL and working volume of 2100 mL. The anode and cathode chambers were separated using a glass plate frame having 6 × 6 cm hole. The hole was tightly sealed by a proton exchange membrane (PEM-Nafion™ 117, DuPont Co., USA). Plain carbon paper (7 × 7 cm) and graphite plate (7 × 7 cm) were used as anode and as cathode (Fig. 1a). The electrodes (both anode and cathode) were connected to copper wire and exposed copper metal surface at the joints, were tightly sealed with non conductive epoxy resin. Both anode and cathode were suspended in their respective chambers. The anodic chamber was filled with 2100 mL dairy wastewater. The anodic chamber was continuously flushed with a mixture of N_2/CO_2 (80:20, V/V) to maintain anaerobic conditions. On the other hand, cathode chamber was filled with 700 mL of 100 mmol/L phosphate buffer and pH was maintained to 7.0 by 0.5 mol/L NaOH. Air was percolated in the cathode chamber through a 0.45 μm pore size filter to provide molecular oxygen as electron acceptor for cathode.

Multiple anode chamber MFC design (MAC-MFC) was constructed from three media bottles and one water bottle. Each media bottle had a total working volume of 1000 mL while water bottle was of 5000 mL capacity. The media bottles were developed as anode chambers and water bottle as cathode. All anodes and cathode bottles were joined by a non-conductive resin with 6 × 6 cm PEM, separating the passage between the bottles (Fig. 1b). Electrode arrangements consisting of plain carbon paper (7 × 7 cm) as anode and three parallel graphite plates (7 × 7 cm) as cathode were used. The anodes and cathodes were suspended from the top cover, which was tightly sealed. The anodes were continuously flushed with N_2/CO_2 (80:20, V/V) to maintain anaerobic conditions. Cathode chamber was filled with 100 mmol/L, 2100 mL phosphate buffer and pH adjusted to 7 by 0.5 mol/L NaOH. Cathode chamber was provided with air that was passed through a 0.45 μm pore size filter to provide molecular oxygen as electron acceptor for cathode. The electrodes were attached to copper wire with all exposed metal surfaces sealed with a nonconductive epoxy. One sampling ports were tapped in the side of the anodes and cathode chamber to permit withdrawal and addition of medium solution.

1.3. MFC operations

After the attachments were completely dried, both the cathode and anode electrodes were soaked in deionized water for 1 hr before assembling the MFCs. The anode chambers were filled (700 mL) with wastewater for study. Initially MFCs were inoculated with artificial wastewater containing glucose as carbon source. After two cycles, feed solution containing 50% artificial wastewater and 50% dairy wastewater sample, inoculated into MFCs separately. After four cycles, feed solution was switched to dairy wastewater sample.

The experimental setup was run in fed-batch mode with dairy wastewater as anolyte except as indicated. The performance of all the MFCs was evaluated by measuring current, current density, potential, open circuit voltage (OCV), and power density along with COD removal efficiency. Stable voltage output was

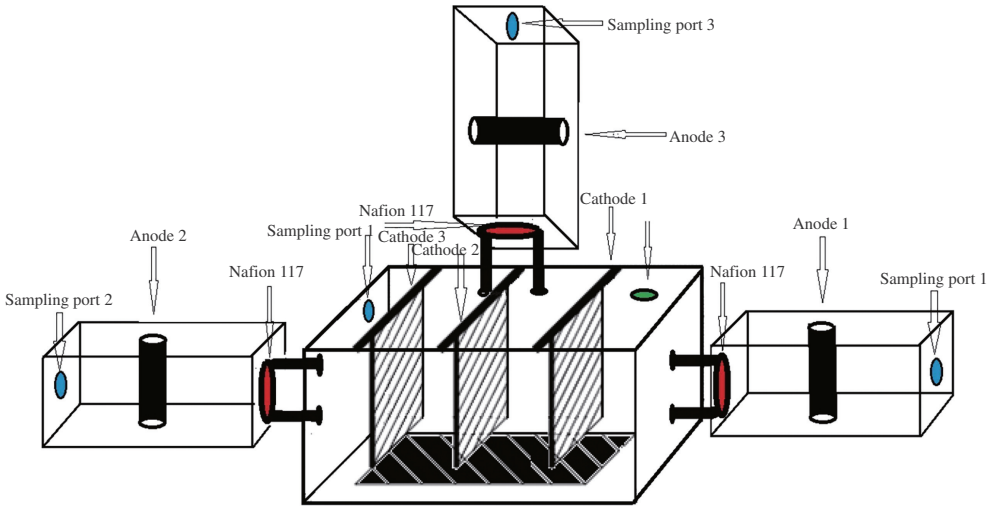

Fig. 1 – Schematic representation of standard single anode/cathode chamber microbial fuel cell (SC-MFC) and multiple anode chamber and single cathode chamber microbial fuel cell (MAC-MFC) design.

achieved after two cycles (30 days each). Constant substrate (COD) removal efficiency and voltage output were considered as indicators to assess the stable performance of the MFC. Electrode fouling was not observed and the electrodes could be used in further experiments without remarkable activity loss. Before changing the feed, inoculum was allowed to settle down (1 hr) and exhausted feed (350 mL) was replaced with fresh feed under anaerobic condition. The anode chamber was sparged with oxygen-free N_2 gas for a period (4 min) to maintain anaerobic microenvironment after every feeding event. A steady increase in voltage generation was observed with additional feed.

1.4. Polarization curve measurement

Polarization curve for an MFC dictates its sustainable performance under specific operating conditions. For each test run, when the MFCs reached steady state (*i.e.*, effluent COD concentration remained little change and obtained current didn't showed much fluctuation), a polarization curve experiment was carried out. For polarization, current generation was monitored at various external resistances connected for a few minutes and readings were noted after stabilization of voltage. The polarization curve measurement was conducted to determine the power generation at different external resistors, R_{ext} (Ω). The R_{ext} was changed from 100 to 1000 Ω during the measurement and the voltage on each R_{ext} was recorded by a digital multimeter (Model–108, Kusam Electrical Industries, India). In MAC-MFC, all three anodes were connected in series during polarization experiment.

1.5. Electricity measurement

Current (I) and voltage (V) measurements were done by connecting with 200 Ω external circuit. OCV was measured in an open circuit, without any external resistance. The power output (P) generated was calculated according to $P = V^2 / R$ and plotted with respect to R_{ext}. The maximum power output was derived from the polarization curve. Power density, PD (mW/m^2), was calculated by multiplying the current by voltage and dividing with electrode surface area.

1.6. COD measurement

COD measurements were conducted using standard methods (Greenberg et al., 1992). All samples were filtered through a 0.22 μm (pore diameter) membrane filter prior to COD measurements. COD (mg/L) removal was calculated as $E_{COD} = (COD_{in} – COD_{out} / COD_{in}) \times 100\%$, where COD_{in} (mg/L) is the influent COD and COD_{out} (mg/L) is the effluent COD.

1.7. Statistical analyses

All experiments were conducted in triplicate either using separate MFCs or the experiments were repeated at least 3 times, when single MFC was used. The results were presented as average values.

2. Results and discussion

2.1. Wastewater characterization

All wastewaters were characterized to observe their suitability to act as anolyte in anode chambers of MFCs. The characteristics of the wastewaters were observed as shown in Table 1. The results show that these wastewaters are cause of pollution in the environment. In addition, those can be utilized as anolytes in MFCs due to their high COD values.

2.2. Characterization of MFC performance

Fig. 2 represents the polarization curves of both MFC designs, as a function of current density, potential, and power density measured at variable resistances (100 to 1000 Ω). Under low resistances the fuel cell circuit allowed more electrons to flow and to neutralize the protons (H^+) present at the cathode,

Table 1 – Characterization of different wastewaters.

	Dairy	Potato	Paper
pH	7.9	6.4	7.8
COD (mg/L)	2057	1735	2839
BOD (mg/L)	1827	234	1528
TSS (mg/L)	2867	256	385
VSS concentration (mg/L)	1945	1276	2458
TDS (mg/L)	13878.5	11794.1	19889.9
Odor	Foul	Foul	Foul and intolerant
Color	Brownish	Brownish	Dark brownish

COD: chemical oxygen demand, BOD: biological oxygen demand, TSS: total suspended solids, VSS: volatile suspended solids, TDS: total dissolved solids.

in comparison to higher resistance, which resulted in rapid stabilization of potential at higher resistances. In similar manner, decreasing current generation trend with increase in the resistance was observed in both MFC types and at 1000 Ω relatively less current generation was recorded. This typical current and potential decreasing trend with increase in resistance was found to be in consistent with earlier studies (Kaewkannetra et al., 2011; Oh et al., 2004), and thus represents typical fuel cell behavior. Higher performance was observed in MAC-MFCs than SC-MFC during polarization studies, which indicate better substrate diffusion and less internal losses in MAC-MFC. MAC-MFC showed 1.23 times higher power density (356 mW/m^2) than the power density obtained from SC-MFC (289 mW/m^2). These results encourage the candidature of MAC-MFC particularly for further large scale applications.

2.3. Electricity generation in MAC-MFC and SC-MFC

After successful stable start-up, performance was measured by running the MAC-MFCs and SC-MFCs without any external resistance for 60 days under identical conditions. MFCs started generating power soon after inoculation and a gradual rise in the OCV was observed, which might be due to readily degradable components in wastewater. The voltage dropped when the easily degradable contents of anolyte were exhausted, yet the presence of other degradable components supported microbial metabolism and less OCV was observed. When the voltage output dropped remarkably, 50% of fresh wastewater was replaced with old, into MFCs to maintain the COD of 1500 mg/L. A linear increase in OCV output was observed with every additional feed.

When compared with SC-MFC, MAC-MFCs exhibited higher performance. The voltage output of MAC-MFC was 1.21 times (986 mV) higher than SC-MFC (810 mV). While highest current density of MAC-MFC was 288 mA/m^2 in comparison with 196 mA/m^2 of SC-MFC. The MAC-MFC showed less fluctuation in the voltage as well as current response in comparison with SC-MFC (Fig. 3). This might be due to multiple anode chambers in MAC-MFC which acted as co-backup to other anodic chamber. In addition, distribution of anolyte among three chambers provides much scope for microbial metabolism and substrate distribution.

2.4. COD removal efficiency

As the main aim of present study was to observe the efficiency of MFCs as waste treatment system, therefore during the operation MFCs were continuously monitored for waste (as COD) removal. COD value was fixed at 1500 mg/L in both MFCs. Both the systems showed their potential for COD removal indicating the function of microflora in metabolizing the waste in wastewater as electron donors. Microbes enriched for 60 days in an MFC removed organic contaminants in wastewater almost completely, with the concomitant generation of electricity. MAC-MFC showed a higher and stable COD removal efficiency than SC-MFC (Fig. 4), during 60 days of operation.

2.5. Simultaneous treatment of different wastewaters in MAC-MFC

Many real waste treatment sites generate more than one type of wastewaters which hinders the installation of standard MFC which treats only one type of wastewater in one time. In an attempt to meet this challenge, MAC-MFC was operated with three different wastewaters (dairy, potato and paper industry) in each anodic chamber to study its efficiency as multiple wastewater treatment system. The COD concentration of each wastewater was optimized at 1500 mg/L. COD removal efficiency was checked separately in every anodic chamber, while OCV measured in complete system to check stable voltage output. 50% fresh wastewater was replaced with old, when COD level dropped remarkably. Highest and

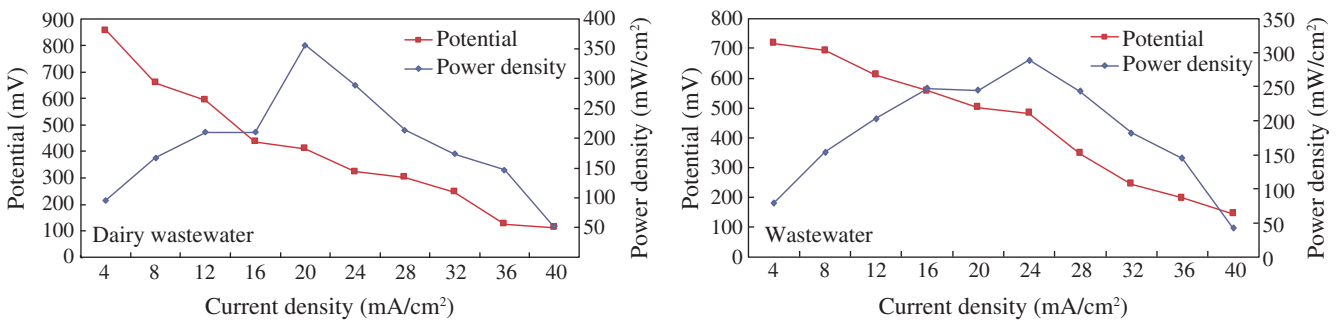

Fig. 2 – Polarization curve obtained from MAC-MFC (a) and SC-MFC (b) operating on dairy wastewater. MAC-MFC: multiple anode chamber and single cathode chamber microbial fuel cell; SC-MFC: standard single anode/cathode chamber microbial fuel cell.

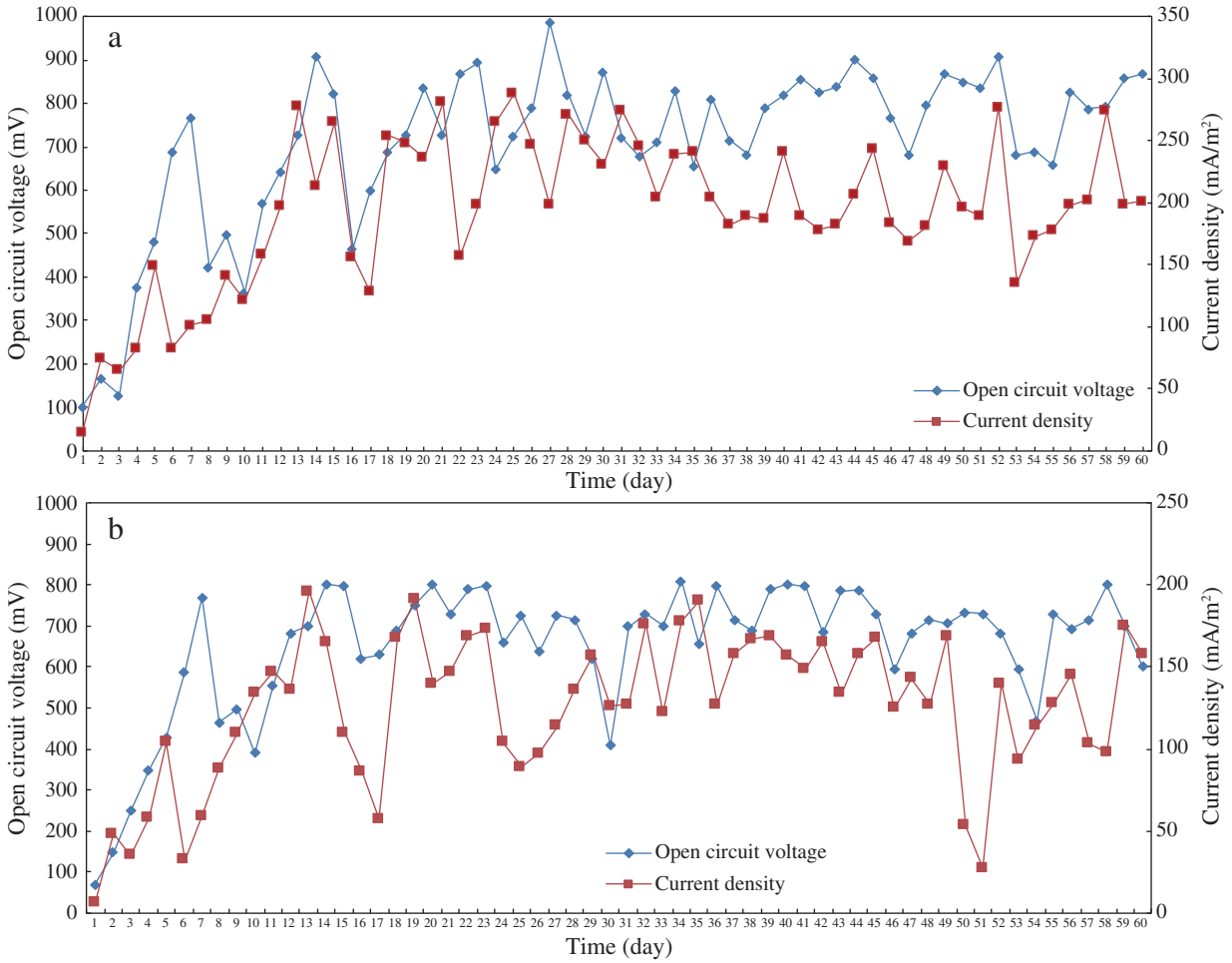

Fig. 3 – Open circuit voltage and current density obtained from MAC-MFC (a) and SC-MFC (b) operating on dairy wastewater.

rapid COD removal efficiency was shown be associated with potato wastewater. Paper industry wastewater showed a fluctuated current response throughout, and it was replaced 3 times less than potato wastewater and two times less than dairy wastewater. It was due to less COD removal in paper industry wastewater than other two wastewaters. Although

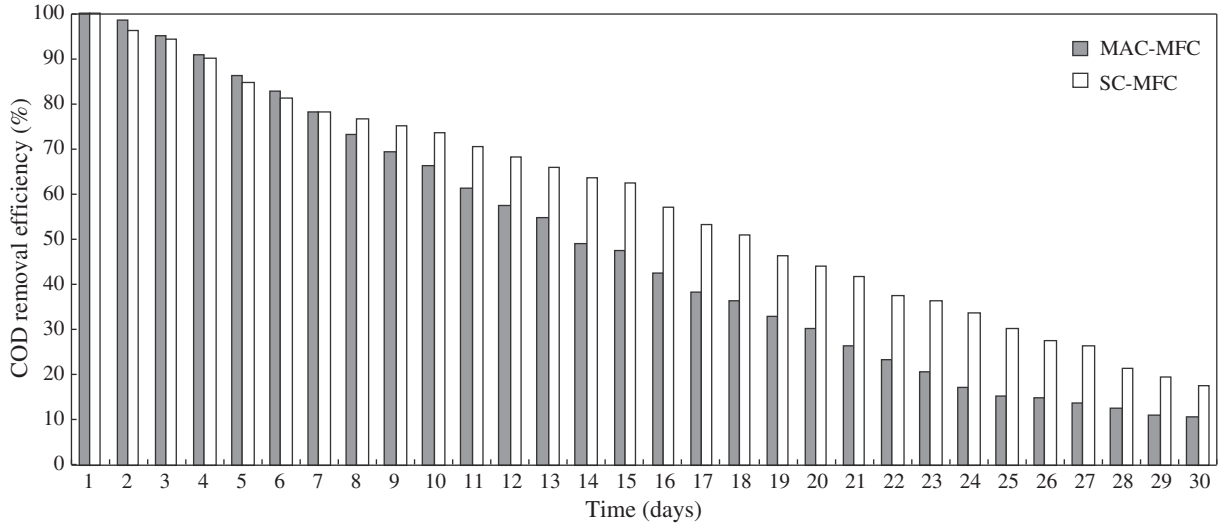

Fig. 4 – Chemical oxygen demand (COD) removal efficiencies in MAC-MFC and SC-MFC.

the COD removal trend was different yet the voltage output was almost stable, which clearly indicates that all three chambers were associated with each other and acted as backup for another anode chamber.

In some earlier studies, it was observed that the cathode was among major limiting factors for lower power output of MFCs (Jiang et al., 2010; Oh et al., 2004; Zhao et al., 2006; Zuo et al., 2007; Jiang and Li, 2009). Increase in the number of cathodes might increase the power output due to higher oxidation rate on the multiple cathodes. This concept was applied in present study that multiple cathode surface area would provide more reaction sites and thus would result in an increase in the voltage output (Oh and Logan, 2007; Rismani-Yazdi et al., 2008). However, ohmic loss and mass transport loss also limit the high voltage yield in spite of large cathode area (Rismani-Yazdi et al., 2008). The possible reason may be the scarcity of oxygen due to less liquid volume in comparison to number of electrodes (in SC-MFC). This problem was illustrated in present study that power density is also dependant of available oxygen at cathode present in catholyte. Catholyte limits the power density even if multiple cathodes are present. The configuration of multiple anode chambers, single cathode chamber (with multiple electrodes in large volume of catholyte) in one MFC reactor, thus maintained the high power generation.

MAC-MFC was found to be more efficient than SC-MFC, in terms of electricity generation and COD removal. The primary reason may be the distribution of anolyte in separate chambers, which facilitated more scope for substrate diffusion. Here, it may consider that MAC-MFC and SC-MFC are stagnant-batch systems, thus substrate diffusion may be the limitation. Second, MAC-MFC offers high membrane:anolyte contact ratio due to multiple anode chambers, which allows higher transfer of H^+ to cathode, thus offers higher efficiency.

Further, different wastewaters were tested in MAC-MFC to check its suitability to treat a large domain of wastewaters. Wastewaters were selected on the basis of the nature of major biodegradable component present in wastewater (dairy wastewater: proteinous; potato wastewater: carbohydratic; and paper industry wastewater: cellulosic nature). Dairy effluent contains dissolved proteins, fats, and sugars and is a good source of nutrition for microorganisms of broad category (Mathuriya and Sharma, 2009b). The potato wastewater is considered as rich source of starch and sugar (Mathuriya and Sharma, 2009b, 2010). Paper wastewater contains mainly cellulose or lingo-cellulosic materials which are difficult to metabolize by microbes than that of low molecular carbohydrates, or of the storage carbohydrate starch, as the ß (beta)-glycosidic bonds of the structural carbohydrate cellulose are highly resistant to hydrolysis (Mathuriya and Sharma, 2009a). Different results were obtained in terms of COD removal which suggests that performance of every anode chamber was independent and it followed more or less same pattern as was in author's previous study (Mathuriya and Sharma, 2009b). Further the stable voltage output was observed due to variation in degradation pattern of different wastewaters, which apparently acted as backup for MFC to generate sustainable voltage output. Although MAC-MFC consumed must cost due to expansive nafion 117, yet the replacement of low cost separator with nafion 117 may solve the problem.

3. Conclusions

The core objective of this study was to describe a proof-of concept MAC-MFC system with special focus on multiple wastewater treatment with energy generation. This prototype successfully demonstrated the defined objective. During parallel operation with SC-MFC, MAC-MFC exhibited superior and stable performance and all anode chambers in MAC-MFC acted as co-backup to others. Yet the performance of MFCs is influenced by several other factors *viz.* microbial activity, anolyte, PEM, internal resistance, and cathode electron transfer efficiency. All of these require careful study in order to develop MFC as an economical power production device and are of further scope.

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