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Research Article

Impacts of process parameters optimization on the performance of the annular single chamber microbial fuel cell in wastewater treatment

Energy harvest from optimized annular single chamber microbial fuel cell (ASCMFC) with novel configuration, which treats chocolate industry wastewater, was investigated. In this study, optimization of operational parameters of the ASCMFC in terms of efficiency water-soluble organic matter reduction and capability of electricity generation was evaluated. During the experiment, effluent from the anode compartment was examined through current and power density curves for variation in temperature and pH, chemical oxygen demand (COD), and turbidity removal, and substrate concentration. The performance analyzed at different temperature ranges such as 25, 30, 35, and 40°C, which showed 88% increase by uprising temperature from 25 to 35°C. The ASCMFC was used to produce electricity by adjusting pH between 5 and 9 at resistance of 100 Ω . Under the condition of pH 7 power density (16.75 W/m³) was highest, which means natural pH is preferred to maximize microbial activities. Wastewater concentration with COD of 700 and 1400 mg/L were investigated to determine its affection on current production. Reduction of current density was observed due to decrease in wastewater concentration. Significant reduction in COD and turbidity of effluent were 91 and 78%, respectively. The coulombic efficiency of 45.1% was achieved.

Keywords: ASCMFC optimization / Bioelectricity / Operational Parameters / Power Generation / Wastewater Treatment

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1 Introduction

Undoubtedly, energy supply chain throughout the world is relied on fossil fuels, which have an impact on natural owing to the emission of carbon dioxide. High consumption of fossil fuels has drastically declined its resources and destroyed natural life cycle by causing atmosphere pollution and global warming. It has brought a tremendous need to discover a suitable and environmental friendly resource as a substitute. Green fuels such

as liquid hydrocarbon fuels, diesel fuels, and biofuels are alternatives for conventional fossil fuels [1–5]. Microbial fuel cell (MFC) provides new opportunity and great potential for production of energy from renewable carbon sources and achieving simultaneous wastewater treatment [6–8].

In MFCs, organic components are metabolized with the aid of microbial communities in the anodic compartment to generate energy, electrons, and protons [9]. Furthermore, in this process, microorganisms fulfill the catalysts role instead of abiotic catalysts such as platinum, manganese oxide, and polyamine. An electron acceptor (commonly oxygen) is reduced to water in the cathode compartment for complete oxidation and reduction processes by consuming electrons and protons obtained from the anode [2].

Recently, most of researches are focused on modifying the power generation and decreasing the operating costs. Single chamber MFC (SCMFC) without proton exchange membrane is easier to perform on a large scale because of smaller volume and less area requirement [10]. In addition, SCMFC is believed

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Abbreviations: ASCMFC, annular single chamber microbial fuel cell; CE, coulombic efficiency; COD, chemical oxygen demand; CODR, COD removal; MFC, microbial fuel cell; SCMFC, single chamber microbial fuel cell

to be better than other types of MFCs due to its lower internal resistance, which is resulted from lack of proton exchange membrane and caused higher power output. Simple configuration, no need to agitate in each compartment, and relatively low cost are other factors, which show SCMFCs superiority [11, 12].

SCMFCs are capable of using various porous carbon materials as the electrode, such as carbon cloth, carbon paper, and carbon rod. The electrode material with strong biocompatibility, high chemical stability, low resistance, and its proper structure can modify bacteria attachment, electron transfer, and substrate oxidation, which affect internal resistance [13–15]. Spiral shape stainless steel modified by graphite has been used as the proper anode electrode, which contributed SCMFC performance and introduced as annular single chamber MFC (ASCMFC). In addition to electrode materials, some elements, such as microbial activity, electrolyte ionic strength, and electrode spacing has an impressive impact on internal resistance and power output limitation [16].

The operating parameters, which can influence the performance of MFC, are extensively explored in the literature but the interaction of various factors is ignored by almost all of them. A series of tests were experienced by Liu et al. [17] supporting effects of operational parameters such as ionic strength, temperature, and cell design, on power generation, which showed possibility of the power density increment by changing these variables. Hence, the mentioned experiment and wide range of other researches show the importance of them in all aspects of MFCs procedures.

In the best of our knowledge there is not any investigation to specify consequences of operational parameters on ASCMFC outputs. In this study, examinations are performed on the ASCMFC, which is obtained from the previous study with optimized electrode spacing [18]. The present paper was aimed to evaluate the performance of the system subjected to variation in temperature, pH, and wastewater chemical oxygen demand (COD) concentration, which led to obtain optimum values of each of these parameters. Furthermore, it was also necessary to evaluate the COD and turbidity to elucidate wastewater treatment success.

2 Materials and methods

2.1 ASCMFC design and construction

An air-cathode ASCMFC in cylindrical shape was fabricated and assembled as reported in our previous study (Fig. 1) [18]. An approximately 91 mm diameter coaxial cylinder was created with 71 mm diameter inner cylinder and 30 mm length. The intersection was used as the main body of the ASCMFC with an effective volume of 90 mL. A 2 cm (height) $\times$ 57 cm (length) stainless steel with 300 meshes (preconditioned in acetone, 0.1 mol/L hydrochloric acid solution, and deionized water) was used as anode electrode. The preconditioned anode was coated by graphite in order to increase its conductivity, which also could improve interaction between electrode and bacteria. A 3.5 cm (height) $\times$ 11 cm (length) 30 wt% wet proofed carbon cloth (Type A, E-TEK, Somerset, NJ, USA), with 0.5 mg/cm² of Pt on the liquid-facing side, was placed in the inner cylinder as

Figure 1. A schematic view of the optimized ASCMFC instruction, illustrating anode compartment with spiral shape anode electrode.

the cathode electrode [19]. The design was modified by fixing electrode spacing at 0.7 cm and the optimized ASCMFC was acquired by considering our previous work [18]. The system was monitored using a data acquisition system at an interval of 15 min to record the voltage outputs of the ASCMFC. The electrolyte was a mixture of 50% v/v of wastewater and growth media solution, which contained the following components for per liter: KCl (0.13 g/L); Na₂HPO₄ (4.09 g/L); NaH₂PO₄ (2.544 g/L); NH₄Cl (0.31 g/L); vitamins stock solution (5 mL) and 12.5 mL trace minerals element solution. All chemical materials used in current investigation were supplied from Merck (Darmstadt, Germany). The ASCMFC was inoculated with 3 mL of anaerobic sludge collected from the activated sludge reactor treatment plant (Qaem-shahr, Mazandaran, Iran). In this experiment, 3 mL of anaerobic sludge, 42 mL of growth media, and 45 mL of wastewater were injected into the anode chamber for the startup of the ASCMFC. Fuel for the cell was the chocolate industrial wastewater discharged from Farmland Chocolate Company (Tehran, Iran). The chocolate manufacturing industry wastewater was selected due to its high organic loading such as carbohydrates, fatty acids, and high COD, which can be characterized as environmental pollutants. The wastewater, which stored at 4°C and its COD did not markedly change during storage, was used as the inoculum. To prevent clogging, oily particulates and solids of the wastewater were removed apart by physical (filtration) methods. Then, the oil phase of the wastewater was separated via gravity-based settling and coagulation processes. The characterization of the wastewater before use is summarized in Table 1.

2.2 Experimental procedure

As the literatures illustrate, more flexibility in the control of the substrate concentration was allowed via fed-batch mode in the culture medium [20]. The reactor was inoculated with 3 mL of mixed culture, which was adopted with a mixture (1:1) of

Table 1. Chocolate industry wastewater initial characterization

Parameter	Values
pH	5.02
COD (mg/L)	1400
Turbidity (NTU)	139.68
Conductivity ($\mu\text{S}/\text{cm}$)	3160
TDS (ppm)	1.57
Salt (ppt)	1.55

wastewater and growth media solution in the fed-batch mode once each 2 days for over 2 months. A series of experiments were performed in order to evaluate optimized electrode spacing in the ASCMFC configuration by described procedure in the previous study [18]. The external resistances in the circuit (open circuit, 1000, 800, 600, 500, 300, 200, 150, 100, and 50 Ω) were investigated separately with the optimized cell, based on multiple-cycle method [21, 22]. By the aid of polarization and power density curves, interesting data can be obtained about the potential and influential parameters of the ASCMFC. Hourly averaged sustainable voltages at the permanent stage of each batch of the examination were used to plot these curves. The maximum output power density of 22.898 W/m^3 and current of 6.42 mA were resulted from the mentioned optimized configuration [18]. Experiments were divided into three series in order to evaluate the interaction of operational conditions on the cell, which presumptively affect the electricity generation and some wastewater characterizations, including substrate concentration, pH, and temperature operation, were examined. To explore the influence of different anodic pH on the ASCMFC, the system was controlled at five different pH of acidophilic (pH 6), neutral (pH 7), and alkaline (pH 8, 9) after adjusting the feed pH to the mentioned values using 0.1 M HCl and NaOH solutions. Sequential examinations performed on the chamber with vary temperatures (25, 30, 35, and 40°C) and produced current was monitored to extract the optimum operation temperature from the results. Experiments were carried out at different COD substrate of 1400 and 700 mg/L to evaluate the effect of substrate concentration on the cell performance. COD, turbidity removal efficiency, and power output were considered as indicators to assess the permanent performance of the ASCMFC.

2.3 Analyses and calculations

The cell voltages were collected through a data acquisition system, which was connected into PC across the electrodes, with on-line monitoring and recording at 15 min intervals. The current was evaluated by using previously measured voltages based on Ohm's Law and converted to power after collection according to the following relation: $P = I \cdot V$; where, P is power (W), I is current (A), and V is voltage (V). The power density was calculated by dividing the power by anode liquid volume (W/m^3). Polarization curves were plotted by changing the external resistance in the circuit according to the previously described multiple-cycle method. Coulombic efficiency (CE) was defined by $\text{CE} = \frac{M \int I dt}{n F V_{\text{An}} \Delta \text{COD}}$, where M stands for the molecular weight of oxygen (32 g/L), n stands for the number of electrons transferred

Figure 2. Maximum power density and current generation as a function of pH, $R = 100 \Omega$.

per mole of oxygen and F is for Faraday's constants, ΔCOD illustrates differences in COD concentration in a batch cycle, I is the generated current, and V_{An} is the volume of anode chamber. The pH values were measured using a pH meter (HI 211; HANA Instruments, Woonsocket, RI, USA), which was calibrated with buffers (pH of 4 and 7). The turbidity was evaluated by a probe (HI 93703; HANA Instruments, DJ110E, 457260, Romania). The bioreactor was operated via batch mode and the temperature was maintained constant in different values (25, 30, 35, and 40°C) using an incubator (311 DS; Labent, USA). The influent and effluent COD were monitored according to standard methods and COD removal (CODR) (%) was calculated based on the initial and final COD [23].

3 Results and discussion

3.1 The effect of pH variation on the maximum power output

It is presumed that pH of aqueous environment has a significant influence on the rate of treatment and power density in MFCs, by affecting the physiology of bacteria [2]. In a dual-chamber MFC with synthetic wastewater, investigations showed that suitable pH for maximizing performance was 6.5 [24]. However, in another study a pH around 9.5 was obtained as the optimum pH for an air-cathode MFC treating domestic wastewater [25]. In order to find the optimum pH, which was expected to help both the required activity of microorganisms and current intensity, previously described examinations performed and data are presented in Fig. 2. As the graph demonstrates, power density and current were reasonably low at pH 5 and followed by a drastically increased pH range between 6 and 7. The power density increased (30.21 %) from 12.84 W/m^3 at pH 6 to 16.72 W/m^3 at pH 7, respectively. However, it was not further increased at pH higher than 7; therefore, power density dropped to 11.37 W/m^3 at pH 9. A similar trend was also followed for the current.

As the earlier study has indicated, not only the pH has impact on the consistency of cell membrane, but also influenced absorption and solubility of nutrients, which impressed directly in the microbial community structure involved in biodegradation [26]. The pH ranges below 6 and higher than 8 cannot

Figure 3. Exploration of effects of different temperatures on (A) power densities and (B) current generations. All tests were carried out in ASCMFC at R_{ext} of 100 Ω .

support stability of enzymes and suppress activity of required microorganism. All microorganisms are active in a certain pH range, due to their growth and replications, which will be inhibited under other conditions [26,27]. It can be understood from the illustrated data in Fig. 2 that habituated mixed anaerobic consortia are suitable for living in a neutral environment at pH 7, which is also ideal for wastewater treatment.

3.2 Effect of temperature variation on the maximum power output

Temperature is known as one of the important parameters that can be used to control growth and metabolism of microorganisms and consequently enormously affects performance of MFCs. As a matter of fact, kinetics of bacterial growth, cathode oxygen reduction, and proton transport rate through the anolyte are factors that relied on temperature changes [17]. The ASCMFC was tested in batch mode at four different temperatures ranging from 25 to 40°C to figure out the optimum temperature while other parameters were kept constant; results were examined in terms of output power density and current illustrated in Fig. 3.

Power density and current at temperature of 25°C were at their lowest scale of 5.4 W/m² and 2.1 mA, respectively, which

might be caused by the low growth rate of bacteria under low temperatures (Fig. 3). By stepwise increasing operating temperature from 25 to 35°C with step increments of 5°C, drastically increase in power density from 5.42 to 16.29 W/m² (Fig. 3A) and current from 2.01 to 3.78 mA (Fig. 3B) were resulted due to increase in metabolic and enzymatic activity of biological cells. Temperatures beyond 35°C might damage important ingredients such as proteins, nucleic acids, and others temperature-sensitive components irreversibly, which would lead to reversing the curve and fall in cell function. It was probably the main reason of decrease in maximum power output when temperature became higher than 35°C. The most studies and documents showed that suitable temperature for mixed microflora is varied, based on their tolerance and type such as psychrophilic, mesophilic, and thermophilic.

3.3 Effects of COD and turbidity reduction on wastewater treatment

The ASCMFC configured in the current investigation also showed significant removal of substrate (COD) along with simultaneous power generation, respective to the experimental conditions used. The wastewater treatment strength was evaluated in terms of CODR. The effect of the COD concentration on the liquid bulk of the anodic compartment over the batch operation time for assessing the performance of wastewater treatment was investigated (Fig. 4A). Initial substrate with COD of 1400 mg/L was injected into the anode compartment and 100 Ω external resistance was applied.

As it can be seen in Fig. 4A, maximum CODR efficiency after 96 h operation was 91.2%; which shows a decline in COD from 1400 to 123.2 mg/L. This decline in COD concentration indicates the high potential and function of the ASCMFC as an alternative for the conventional anaerobic wastewater treatment processes. Other parameters assessed for ASCMFC operation evaluation as a modern technology for wastewater treatment was the efficiency of turbidity reduction. The reduction of turbidity in the effluent from the system followed the trend as shown in Fig. 4B. The initial turbidity has reduced by 79.66% in 96 h (Fig. 4B); the evaluated reduction in turbidity was among the highest rate of reduction reported in the literature. This efficacy in turbidity removal by the MFC system was noticeable. There are several probable reasons for this fact such as degradation mechanisms of colloidal and decomposition of organic matter.

3.4 Current production at variable substrate concentration

To examine the efficiency of substrate concentration concerning to power and current generation, 100 Ω of external resistance and the most proper obtained value of other variables were applied to the optimized ASCMFC. The system was executed in a batch cycle for each primary effluent of 1400 mg/L and diluted one of 700 mg/L. As it is illustrated in Fig. 5A, the current value was decreased from 3.77 to 2.76 mA by reducing the substrate COD concentration. Sufficient substrates

Figure 4. Variation of (A) COD removal and (B) turbidity reduction efficiency (%) in anode chamber with time in one cycle continuing for 4 days.

are vital for high bacterial activities, so that lack of organic contents in the anolyte justifies the decline in current generation and proves the close relation between current and effluent concentration.

Two separate batch cycles of 30 to 20 h, in the ASCMFC were compared (see Fig. 5B). Figure 5B shows that not only lower COD concentration decreased the highest generated current, but also it has shortened the batch time and remained at the stationary phase and declining phase for shorter period of time.

3.5 CE

CE was an essential factor for evaluating the accurate performance of MFCs. The CE is defined as the ratio of electrons extracted as current versus amount of electrons in the starting organic material. Respective data of COD concentration of the effluent are shown in Fig. 4A and current evaluation was measured while 100 Ω external resistance was fixed. Maximum CE of 45.1% was achieved by exhibiting an opposite trend with the increase of COD concentration, which was more than 1.67-fold of the value reported by Mardanpour et al. [13]. The opposite trend is probably caused by the competitive substrate consumption by microorganism community, which leads to more change in COD concentration

Figure 5. Impacts of different COD concentrations (1400 and 700mg/L) on current generation (A) during multiple batch cycles and (B) a single batch cycle.

irrespective of current generation and consequently lower CE [28].

4 Concluding remarks

Present study revealed the influence of key process parameters affecting performance of the optimized ASCMFC with the activated sludge microbial population. Effect of pH on electricity generation of the system was observed at a feed solution pH between 5 and 9. The optimal pH of 7 was reported (power density of 16.72 W/m³), which was proper for microorganism and accelerated treatment of the wastewater. It is noteworthy that electricity generation was significantly correlated with temperature as an important parameter in treatment efficiency. It is resulted that the electroactive anodic consortia developed in the MFC was more sensitive to behave in the appropriate temperature range between 35 and 40°C. The COD and turbidity removal efficiency of 96 and 79.66%, respectively, were significant outputs, which show that wastewater treatment is well done by the ASCMFC. The highest CE obtained in the current study was about 45%, which was 1.67-fold of previously reported values with the same cell configuration. To the best of our knowledge the resulted ASCMFC illustrated the highest performance in compare to similar SCMFCs. Also, it is mentioned that the higher COD concentration in anolyte causes the longer batch time, declining and stationary phase of the system.

Practical application

MFCs are promising technology with raising industrial interest for evaluating the simultaneous effect of wastewater treatment and electricity generation. This paper is targeted on demonstrating positive impacts of optimum key process parameters on the performance of the optimized innovative annular structured MFC, for the first time. Successful reduction in COD and turbidity by the current AS-CMFC shows that this novel technology can potentially be used; also separately used for industrial treatment usages.

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5 References

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