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Effect of coconut shell salt bridge on the performance of microbial fuel cells for wastewater treatment and bioelectricity generation

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Abstract. Dual-chamber salt bridge microbial fuel cell (DCSBMFC) is an innovative technology for the treatment of wastewater and generation of bioelectricity through bacterial oxidation. Coconut shell powder (CSP) was tested as a potential proton exchange membrane (PEM) alternative to Nafion for enhancing the performance of the salt bridge. This study evaluates the impact of CSP addition on DCSBMFC performance in terms of wastewater treatment and bioelectricity generation. Inoculated samples and 12 cm long salt bridges both with and without CSP under anaerobic conditions were tested. Results showed that the DCSBMFC in the absence of CSP performed better, with chemical oxygen demand (COD) and ammoniacal nitrogen (AN) removal efficiency of 62.59% and 61.76%, respectively, compared to 53.26% and 58.5% for the system in the presence of CSP. Electricity generation in the absence of CSP recorded a maximum voltage of 118.2 mV, while the system in the presence of CSP produced a maximum voltage of 91.5 mV. These findings demonstrate that the DCSBMFC operates efficiently and reliably without the addition of CSP, indicating that the original design remains effective for both wastewater treatment and bioelectricity generation.

1. Introduction

Wastewater treatment is central to environmental sustainability, which aims to reduce the damage inflicted upon aquatic ecosystems by harmful contaminants and pollutants [1]. Traditional wastewater treatment works well but at the expense of high energy requirements and high cost, which drives quests for new, environmentally friendly technologies. Among the emerging alternatives, microbial fuel cells (MFCs) have drawn enormous interest due to their double capability of wastewater remediation and power generation [2].

Malaysia, a rapidly developing country with an estimated 32.7 million population as of 2023, has seen robust economic and demographic growth. In its goodness, the development has resulted in long-term environmental challenges such as increasing levels of untreated wastewater, water resource tensions, and higher energy consumption [3]. he country still retains its reliance on fossil fuel particularly coal and natural gas to produce electricity, which not only causes concerns of depleting resources but also causes significant harm to the environment in terms of pollution. The non-renewable resources are likely to deplete over time, further pointing to the importance of utilizing alternative sustainable energy resources [4].

The increasing volume of wastewater production in Malaysia has beyond the capabilities of existing treatment facilities, illustrating a worldwide concern where more than 80% of wastewater is discharged without adequate treatment [5]. The increasing need for clean water and energy necessitates the development of integrated and sustainable solutions. MFCs provide an effective solution by facilitating wastewater treatment and energy generation, hence enhancing infrastructure resilience and sustainability.

Dual chamber microbial fuel cells (DCMFCs) generally employ membrane separators or proton exchange membranes (PEMs), frequently constructed from materials such as Nafion, owing to its advantageous chemical and mechanical durability as well as superior proton conductivity. Nafion, however, has various drawbacks, such as fuel crossover, water evaporation at high temperatures, and severe swelling. Additionally, its high cost and non-biodegradability raise concerns about environmental impact and waste management [6,7].

Low power output and how to scale up the power output have always been the challenges faced by MFCs [8]. Electricity is generated due to the flow of electrons in the system. However, protons play an important role in completing the electrochemical process. Therefore, salt bridge plays an important role as it acts as the PEM. The addition of new materials to the salt bridge may possibly help to increase the proton conductivity of the salt bridge and might potentially increase electricity generation.

2. Methodology

2.1 Preparation of salt bridge

Coconut shell powder (CSP) was added to the salt bridge. The shells were sourced from a flea market in Kangar, Perlis, then washed, cleaned, and dried. Their surface roughness was removed using sandpaper before being ground into powder using grinder. The CSP was collected and stored in a container. To prepare the salt bridge, 10% agarose and 4% potassium chloride (KCl) were mixed with the CSP. The mixture was dissolved, heated, and stirred in a water bath, then poured into a 12 cm-long, 2 cm diameter PVC pipe sealed at one end with cellotape. After cooling, the pipe was refrigerated for 24 hours to solidify the salt bridge.

2.2 Preparation of empty fruit bunch (EFB) polypropylene biofilm carrier

EFBs and polypropylene bio balls served as carriers for microbial adherence. The EFBs were obtained from a palm oil mill at UOP Sdn. Bhd., Nibong Tebal, Pulau Pinang, whereas the bio balls were bought from SH Aquatic and Pet Centre, Kangar, Perlis. The EFBs were sterilized by soaking, washing 3 to 4 times with distilled water and rinsing to eliminate contaminants. The samples were subsequently dried in an oven at 60°C for 24 hours. The bio balls underwent a similar sterilizing procedure, which included soaking, rinsing twice with distilled water, and drying in an oven. The sterilized EFBs were firmly affixed to the polypropylene bio balls for subsequent utilization.

2.3 Inoculum preparation

The wastewater was obtained from palm oil mill effluent (POME). POME was mixed with synthetic wastewater in a ratio of 3:2 and this mixture will act as the inoculum. This inoculum was used for inoculation of the carbon felts and the polypropylene bio balls tied with EFBs by soaking them in the mixture. They were inoculated for 20 days under anaerobic condition.

2.4 Bioreactor configuration set up and operation
Two 1.6L plastic containers served as the anode and cathode chambers of the MFC (figure 1). Both were sterilized with distilled water and methanol. A 2 cm opening was made in each container, and a PVC pipe containing the salt-agarose-CSP mixture was secured between them using silicone sealant, functioning as a salt bridge to facilitate proton transfer. The anode chamber was filled with 1L of synthetic wastewater, while the cathode chamber contained tap water, each with its respective graphite electrode. Inoculated polypropylene bio balls wrapped with EFBs and carbon felts were added to the anode chamber. To create anaerobic conditions, nitrogen gas was injected before sealing the chamber. An air pump supplied oxygen to the cathode chamber. The system was maintained indoors at ambient temperature for 30 days.

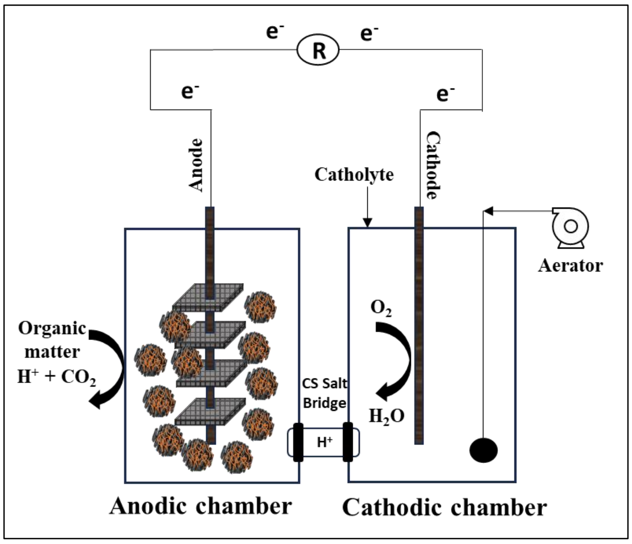

Figure 1. Configuration of DCSBMFC.

A 1000Ω resistor was connected between the cathode and anode using crocodile clip wires, and the resistor was attached to a data logger. The experiment was performed for 30 days in a salt bridge, initially without CSP. Chemical parameters of wastewater were analyzed, and electricity generation was recorded. The experiment was repeated in a salt bridge with 5% CSP to test its effect on wastewater treatment and also on bioelectricity generation.

2.5 Analytical quantitative methods
This section describes the quantitative analytical methods used for wastewater of the treatment from the microbial fuel cell system. Quantitative analysis was used and conducted in a method that has been proven and validated.

2.5.1 Chemical oxygen demand (COD)
The COD of synthetic wastewater was determined as per American Public Health Association (APHA) Method 5220D. 2.5 mL of wastewater and 2.5 mL of distilled water as blank were put into 2 different vials, followed by 1.5 mL of potassium dichromate ($K_2Cr_2O_7$) digestion solution and 3.5 mL of sulphuric acid (H_2SO_4) reagent. The vials were subjected to a thermal treatment of 2 hours at temperature of 150°C within the DRB200 reactor which had been preheated before experiment.

The blank was placed in the DR2800 Spectrophotometer after cooled to room temperature and read using the photometer to see the zero absorbance. The blank was replaced with the sample and read to measure the absorbance. The result obtained was the direct COD concentration reading of the sample measured as mg/L.

2.5.2 Ammoniacal nitrogen (AN)
The AN of the sample was analysed by following the APHA Standard Methods 4500-NH_3 Nessler Method. A 25 mL mixing cylinder was filled to the 25 mL mark with distilled water and another 25 mL of cylinder was filled with sample. Then, three drops of mineral stabiliser were added to each cylinder and the cylinder was homogenized completely. Three drops of polyvinyl alcohol dispersing agent and 1.0 mL of Nessler reagent were added to each cylinder. Each solution was poured into different 10 mL square sample cell and the blank was placed into the cell holder when the time beeped. The soft key (spectrophotometer) Zero was pressed and the display showed 0.00 mg/L NH_3-N. The blank was replaced with the sample and the AN reading was obtained after pressing the "Read" key.

2.5.3 Total suspended solid (TSS)
Total suspended solids (TSS) analysis was conducted by first pre-weighing a crucible containing filter paper. The wastewater sample was homogenized for 10 minutes and transferred to a measuring cylinder. A vacuum pump was connected to a vacuum flask, onto which the crucible was placed. The homogenized sample was then filtered through the crucible under vacuum. After filtration, the crucible was dried in an oven at 105°C for 24 hours, then cooled to room temperature. The crucible, now containing the retained solids, was weighed using an analytical balance. TSS was calculated using equation (1).

$$TSS\left(\frac{mg}{L}\right) = \frac{M_2 - M_1}{V} \tag{1}$$

Where, M_1 is the weight of crucible with filter paper only (mg), M_2 is the weight of crucible with filter paper and suspended solid (mg), and V is the volume of sample filtered (L).

2.5.4 Voltage
The voltage of the system was measured using a data logger connected to the system. The voltage generated by the system with a resistor of 1000 Ω was recorded for a period of 21 days.

3. Result and discussion

Chemical parameters such as COD, AN, dissolve oxygen (DO) and oxygen reduction potential (ORP) and electricity generation are discussed to evaluate the performance of DCSBMFC with and without CSP.

3.1 Characteristic of raw palm oil mill effluent (POME)
The characteristic of the raw POME used during inoculation phase are listed in table 1. Chemical characteristic of the wastewater such as COD, TSS, AN and pH are tested on the wastewaters.

The wastewater was analyzed for key chemical properties, including pH, COD, TSS, and AN. The pH value of 4.44 indicates that the POME is acidic. Since they contain organic acids in complex form which can serves as carbon sources for microbial manipulation. Therefore, they were used as inoculum to inoculate the materials used in the operation of MFC. On the other hand, the COD concentration of 39,180 mg/L signifies a high level of organic pollutants, requiring significant oxygen for chemical oxidation. According to Moreno-Andrade et al. [9], COD is widely used in anaerobic treatment processes as a measure of the susceptibility to oxidation of the organic and inorganic materials present in the raw and anaerobic POME. Additionally, the TSS value of 29,950 mg/L reflects a large amount of suspended solid particles, which may impact treatment efficiency. The presence of AN at 520 mg/L highlights the level of ammonia and ammonium compounds in the wastewater, which can influence biological treatment processes.

Table 1. Characteristic of raw POME.

Characteristic	Raw POME
pH	4.44
COD (mg/L)	39180
TSS (mg/L)	29950
AN (mg/L)	520

3.2 Voltage production

The performance of DCSBMFCs was investigated with and without the incorporation of CSP, revealing distinct differences in electricity generation capacity. As illustrated in figure 2, the DCSBMFC operated without CSP exhibited superior performance, achieving a maximum voltage output of 118.2 mV on the final day of observation. In contrast, the DCSBMFC incorporating CSP recorded a lower peak voltage of 91.5 mV. This discrepancy suggests that the presence of CSP may have adversely influenced the electrochemical activity within the system. The inhibitory effect could be attributed to potential clogging or alteration of microbial activity due to organic or particulate interference from CSP, thus limiting the efficiency of electron transfer mechanisms.

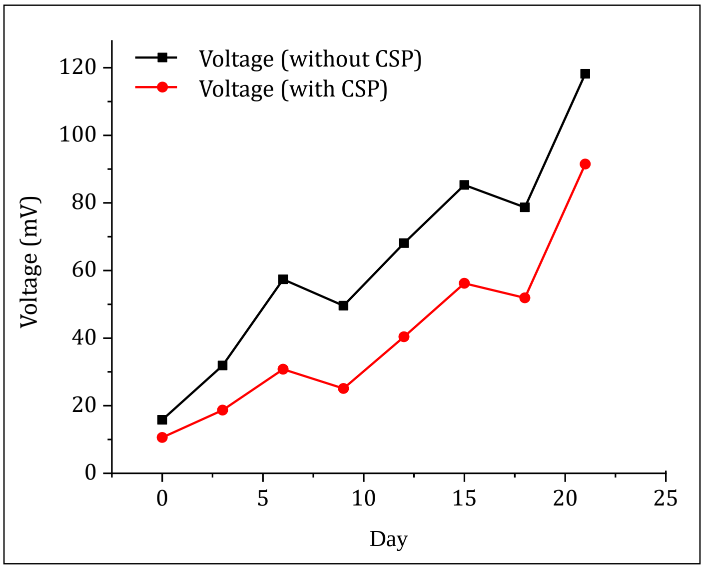

Figure 2. Voltage production in DCSBMFC with and without CSP.

The study also investigated the effect of agarose concentration in the salt bridge on MFC performance at the same time. The results showed that there was a nonlinear relationship between agarose concentration and electricity generation. An increase in agarose from 7% to 10% improved the efficiency of the system, with the best voltage recorded at 0.95 V at 10%. This is likely due to the optimum gel structure at this concentration, which supports efficient proton conduction while being mechanically strong. Increasing concentrations above 10%, for example in 11% and 12%, led to drastic reductions in voltage to 0.57 V and 0.29 V, respectively. These losses are caused by the over-polymerization of the agarose matrix which creates a denser and less permeable gel that suppresses proton conductivity [10]. These findings suggest the delicate balance between mechanical stability and ion transport efficiency in designing effective salt bridge systems for MFCs. The lower voltage output of DCSBMFC with CSP is most likely due to increased agarose gel polymerization with CSP addition, also inducing higher internal resistance.

All these results highlight the crucial role of operation conditions and material composition in regulating the performance of MFCs. While CSP addition would present unfavorable conditions for microbial activity or electron transport, optimal salt bridge conductivity by varying agarose content is required. Physicochemical interaction between added material and microbial populations must be investigated in further studies to better understand the mechanisms responsible for the disparities in performance.

3.3 Chemical oxygen demand (COD) removal

COD removal is one of the significant parameter to be evaluated in determining MFC performance in wastewater treatment. As seen in figure 3, both systems record a decreasing COD trend over the 20 days operation time. For the DCSBMFC without CSP, COD decreased from 858 mg/L to 321 mg/L with a 62.59% efficiency. In contrast, the DCSBMFC with CSP recorded decreasing from 843 mg/L to 394 mg/L with its corresponding 53.26% efficiency. Both systems indicated a rapid initial COD reduction, which indicates active microbial breakdown of organic matter, followed by a slowing rate as the substrate availability decreased. The addition of CSP may have impacted microbial activity or electron transfer, leading to lower COD removal efficiency.

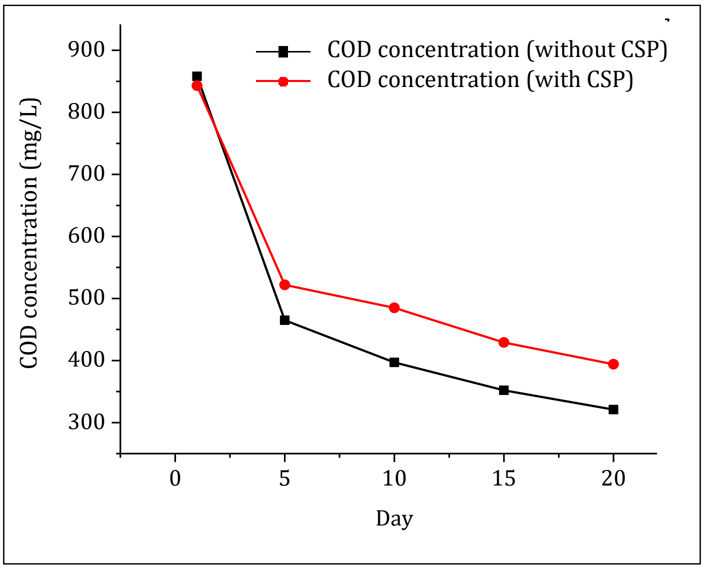

Figure 3. COD removal in DCSBMFC with and without CSP.

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A researcher found that coconut shell as a PEM in a microbial carbon capture cell (MCC) acted poorly in contrast to Nafion [11]. Cellulose in coconut shell acted as an additional substrate for bacteria and reduced COD removal efficiency. CSP in DCSBMFC in this study may play as a second substrate for microbes, dispersing bacterial energy and reducing COD removal. Generally, although both MFCs are effective in reducing COD, the system without CSP performs better.

3.4 Ammoniacal nitrogen (AN) removal

Figure 4 shows a consistent decline in AN levels. DCSBMFC without CSP achieved greater AN removal, reducing concentrations by 61.76% from 59.5 mg/L to 22.75 mg/L, compared to DCSBMFC with CSP, which reduced AN by 58.5% from 50 mg/L to 20.75 mg/L. The higher efficiency of DCSBMFC without CSP may be due to the cellulose content in the CSP salt bridge, which could serve as an alternative substrate for bacteria [11]. Additionally, AN reduction typically occurs through nitrification, an aerobic process. However, since the anode chambers in both DCSBMFCs operate under anaerobic conditions, the decline in AN is likely due to anaerobic ammonium oxidation (anammox), where nitrite serves as an electron acceptor, allowing ammonium to convert directly into nitrogen gas without oxygen [12].

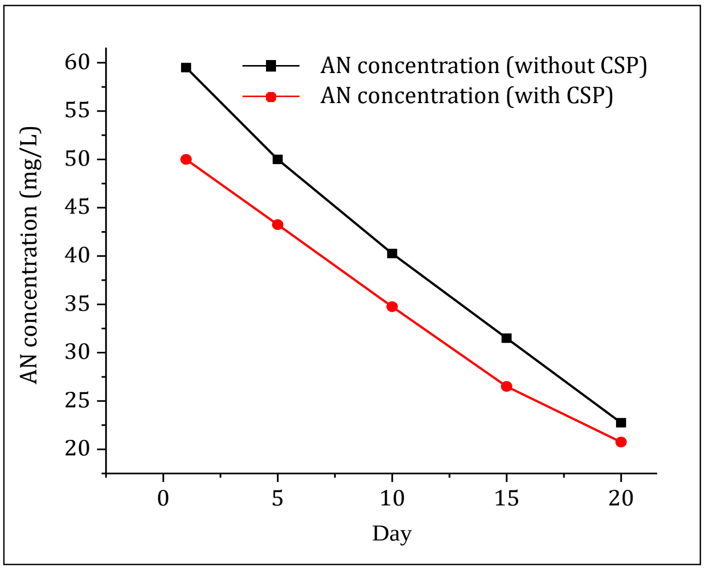

Figure 4. AN removal in DCSBMFC with and without CSP.

4. Conclusion

In conclusion, this study demonstrates that the DCSBMFC is an effective system for both wastewater treatment and bioelectricity generation under anaerobic conditions. Although CSP was explored as an alternative material in the salt bridge, the results showed that the DCSBMFC without CSP exhibited superior performance, with higher COD and ammonium nitrogen removal efficiencies and greater voltage output. These findings suggest that the original design of the DCSBMFC, without CSP, is both efficient and reliable, making it a promising technology for sustainable wastewater treatment and energy recovery.

Acknowledgement

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