Effects of temperature on wastewater treatment in an affordable microbial fuel cell-adsorption hybrid system

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ABSTRACT

A cost effective single-chamber microbial fuel cell (MFC) integrated with adsorption system was tested under different operating temperatures to observe pH profiles, organics, solids, nutrients, color and turbidity removal and power density generation. The optimum operating temperature range was found to be ~25–35 °C with majority of the removals achieved at ~35 °C. Maximum power density recorded was 74 ± 6 mW/m^3 with coulombic efficiency (CE) of $10.65 \pm 0.5\%$ when operated at 35 °C. Present studies had successfully demonstrated the effectiveness of a hybrid system in removing various types of pollutants in POME at optimum temperature and able to fulfill the stringent effluent discharge limit. Chemical oxygen demand (COD), total solids (TS) and turbidity removals increase linearly with temperatures with removal efficiency of 0.5889%C^{-1}, 1.0754%C^{-1} and 0.7761%C^{-1}, respectively. The temperature coefficient (Q_{10}) is found to be 1.06, 1.45 and 1.09, respectively. Besides, MFC-adsorption hybrid system had demonstrated superior stability over a wide range of operating temperatures in terms of COD removal as compared to the non-integrated MFC system.

1. Introduction

Microbial fuel cells (MFCs) are bioelectrochemical devices that utilize bacteria as a biocatalyst to oxidize the organic matters present in the wastewater to produce bioenergy [1–3]. MFCs have the ability to remove various types of pollutants present in the wastewater [4,5]. They are renowned for their great capability for direct bio-energy generation from various biodegradable compounds, ranging from pure compounds to complex substrates in the wastewater [6]. There were many studies done on the typical standalone two-chambered MFC because such design is very easily constructed and typically suitable for lab scale studies. Nevertheless, standalone treatment system has limitation such as less wastewater treatment efficiency as compared to the MFC hybrid system [7]. Much studies had demonstrated the ability of hybrid MFCs in removing various types of pollutants present in the wastewater such as chemical oxygen demand (COD) [8–10], phosphorus [11], dye [12,13], suspended solids (SS) [10], total nitrogen (TN) [14], ammonia nitrogen (NH$_3$—N) [15,16], and so on. Majority of the effective pollutants removal were done by integrating MFC with other unit operation or processes. Recently, it has been reported that greater bio-energy generation and wastewater treatment can be accomplished when MFC is hybridized with unit operations or processes such as adsorption [17], membrane bioreactor [10], sequencing batch reactor [18], membrane, aeration system [19], anaerobic digester [20], aerobic system [15], biofermentor [21], anaerobic fluidized bed [22], wetland [23], anaerobic fluidized membrane reactor [24] and anaerobic sludge blanket [25]. MFC could also possibly be combined with other process such as nitritation-anammox where the research had been greatly developing in order to remove wastewater which content high level of nitrogen content [26,27].

Now, among many influencing parameters, temperature is one of the important factors which can affect the power generation as well as the water treatment performance. The definite temperature may affect the bacterial kinetics and also the types of bacteria that reside in the anodic biofilms in the wastewater fed systems [28]. Thus, while bacterial growth rate and respiration can be affected by the changes in temperature, the community development and structure can also be crucial [29].

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Nomenclature

ACFF	Activated carbon fiber felt
AFMBR	Anaerobic fluidized bed membrane bioreactor
APHA	American public health association
BOD	Biochemical oxygen demand
C_a	Total coulombs by calculated by integrating current over time
CE	Coulombic efficiency
COD	Chemical oxygen demand
C_t	Concentration of the solution at any time
D_1	Design of Behera et al. (2011)
DOE	Department of environment Malaysia
ECG	Electrocardiogram
GAC	Granular activated carbon
GFB	Graphite fiber brush
I	Current (A)
I-BAF	Immobilized biological aerated filter
MFC	Microbial fuel cell
MBR	Membrane bioreactor
$NH4^+$	Ammonium ion
$NH_3{-}N$	Ammonia nitrogen
P	Power density (mW/m^3)
PEM	Proton exchange membrane
POME	Palm oil mill effluent
Pt	Platinum
Q_{10}	Temperature coefficient
R_1	Rate at temperature 1
R_2	Rate at temperature 2
SS	Suspended solids
T_1	Temperature 1
T_2	Temperature 2
TN	Total nitrogen
TMBR	Tubular membrane bioreactor
TOC	Total organic carbon
TSS	Total suspended solids
TVS	Total volatile solids
V	Voltage (V)
ν	Liquid volume (L)

Thus far, not much research has been done on the air- cathode MFC with ceramic pot as the substitute to proton exchange membrane (PEM) except Ghadge & Ghangrekar [30] and Chatterjee and Ghangrekar et al. [31]. The reactor setups for both studies were done on standalone MFC and recently, only one had come to the effort of integrating MFC with adsorption hybrid system with ceramic pot as the PEM [32]. Besides, the effects of temperature in MFC hybrid system has never been understood especially when activated carbon is applied in the design because the performance of both MFC and adsorption system are sensitive to operating temperatures.

A recent study has been done by integrating MFC with adsorption system and such system has demonstrated great pollutants removal in the palm oil mill effluent (POME) such as COD, biochemical oxygen demand (BOD), total organic carbon (TOC), TN, $NH_3{-}N$, total solids (TS), total suspended solids (TSS), total volatile solids (TVS), turbidity and color [32]. Till now, the study of such system was only done at operating temperature of $25 \pm 1\,°C$. The effect of other operating temperature for such system has not been studied in terms of wastewater treatment and bio-energy generation.

MFCs can produce energy from various substrates and reported in literature. However, to the best knowledge of the authors, the

studies of the effect of temperatures had only been done on these few sources which includes beer brewery wastewater [33], domestic wastewater [34–36], sanitary wastewater [37], barley processing wastewater [38] and synthetic wastewater [39,40]. Municipal water treatment experiments which were done previously at low temperatures (5–15 °C) could also be used as a source of high potential MFC operation for electricity generation [41]. At present, none has come to understand the effects of temperature with palm oil mill effluent (POME) as a substrate to MFC which is known as the major contributor of the pollution loads in the rivers in countries such as Malaysia and Indonesia. POME is very rich in terms of organic matters and existing treatment method such as ponding system requires long retention time and ineffective to meet the wastewater quality standard. Since POME has very high organic matters content, therefore it can be used as a source of fuel to the MFC for bioenergy generation.

The aim of this work was to treat the palm oil mill effluent and at the same time to take the advantage from the treatment by integrating the MFC with adsorption system to generate bioenergy. Thus, the effects of temperatures in terms of power generation, pH profiles, organics, solids, nutrients, color and turbidity removal in the MFC- adsorption hybrid system were carried out systematically and presented in the present study.

2. Experimental

2.1. MFC construction

The single chamber air-cathode MFC-adsorption hybrid system with the separator fabricated from ceramic material were constructed and described in Ref. [32]. The schematic diagram of the MFC-adsorption hybrid system and the membrane electrode assembly are as shown in Fig. 1. Anode compartment consists of graphite fiber brush (GFB) (Mill-Rose Co., USA) that served as current collector and employing commercial granular activated carbon (GAC). Prior to experiment, the GFB was soaked in pure acetone overnight and heat-treated at 450 °C, and washed three times with distilled water. GAC was cleaned with distilled water several times and oven dried to remove ashes and other impurities before they were used. A specific surface area of $1000\,m^2\,g^{-1}$ (Nantong Ruibang Activated Carbon Filter Material Co. Ltd, China) activated carbon fiber felt (ACFF) was selected as the cathodes materials, which were wrapped around the ceramic pot (7 mm thick) which acted as the medium for proton exchange. Electrocardiogram (ECG) gel was applied evenly between ceramic pot and ACFF in order to increase hydration and contact area. A copper rod was selected as the circuit connector in the cathode compartment. The reactor working volume for this setup was 5.65 L.

2.2. Experimental design and operation

The MFC-adsorption hybrid system was inoculated with POME from nearby palm oil mill at Felcra Jaya, Kota Samarahan where the POME samples was taken from the second anaerobic pond and the same source was used as the substrate to the MFCs without any additions of nutrient and buffer solution for the anode chamber. Wastewater sample was kept in a cold room at 3 °C prior to use. The characteristic of the POME collected was analyzed and tested (Table 1).

The MFC-adsorption hybrid systems were inoculated at $25 \pm 1\,°C$, where $50\,\Omega$ of external resistance was installed in between the anode and the cathode segment. The systems were operated at various temperature (15 °C, 20 °C, 30 °C, 35 °C, 40 °C and 55 °C) once steady voltage was achieved and the performance were compared to the previous study which was done at operating temperature of 25 °C [32]. The performance of the MFCs at
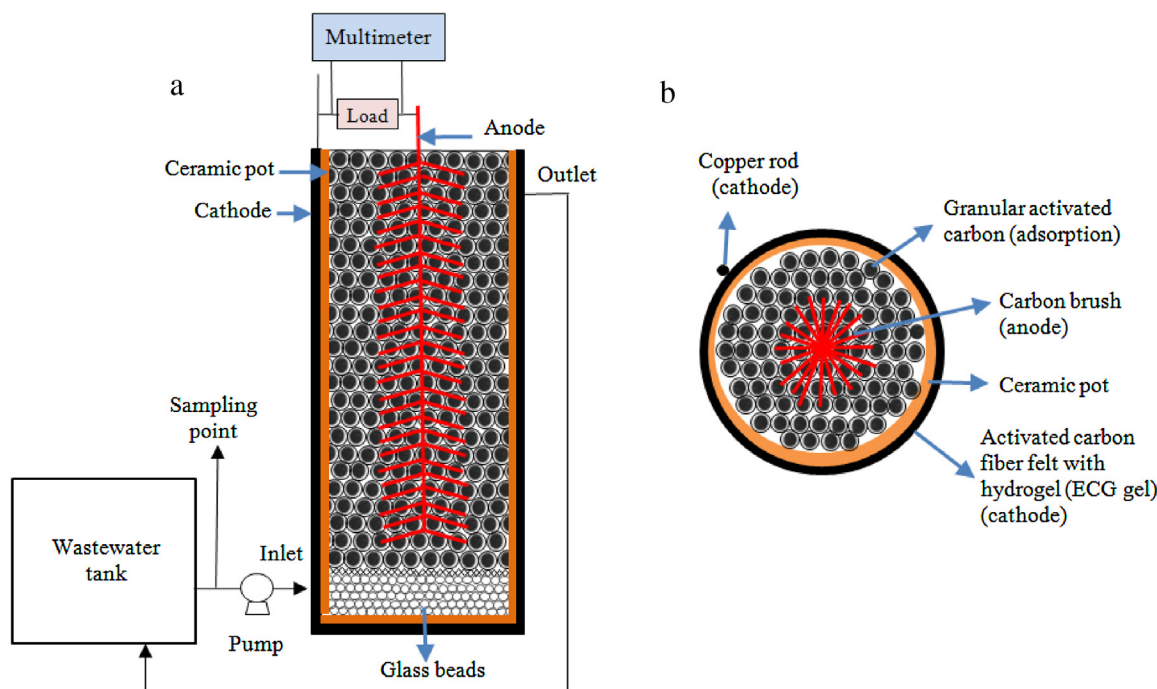
Fig. 1. (a) Schematic diagram of the (a) MFC-adsorption hybrid system (front view) and (b) membrane electrode assembly (top view).

Table 1
Characteristic of POME after the second anaerobic pond with the discharge standards as per Malaysian Department of Environment, DOE.

Parameter	Concentration	Unit	Limit
pH	7.85 ± 0.2	–	5–9
COD	884 ± 3	mg/l	a
BOD_3	233 ± 2	mg/l	100 (20)
TS	6860 ± 3	mg/l	a
TSS	539 ± 3	mg/l	400
Turbidity	203 ± 2	NTU	a
TOC	429.5 ± 2	mg/l	a
TVS	1350 ± 2	mg/l	a
Color	9017 ± 2	Pt-Co	a
TN	167.4 ± 2	mg/l	200
$NH_3{-}N$	121 ± 2	mg/l	150

[a] No discharge standard.

different temperatures was tested in parallel on individual MFCs in the closed-loop mode with fresh POME using incubators (146E, Fisher Scientific). For each temperature settings, the experiments were replicated with fresh loads of GAC.

2.3. Measurement and analysis

The pH was measured using a digital pH meter (PCSTestr 35, Eutech). COD were conducted according to HACH method 8000 which complied to the APHA standard methods with DR900 (Hach). BOD_3 were carried out according to Department of Environment (DOE), Malaysia 2011 reference method where BOD was carried out for three days at 30 °C. TS, TSS and TVS tests were implemented according to the APHA standard methods with APHA 2540 B, APHA 2540 D and APHA 2540 E, respectively. Turbidity was measured using a turbidity meter, TN-100/T-100 (Eutech) and TOC by using a TOC analyzer, TOC-LCPH (Shimadzu). Color was conducted according to HACH method 8025 which complied to the APHA standard methods with DR900 (Hach). TN was carried out according to the APHA 4500-Norg B method whereas $NH_3{-}N$ was conducted as per HACH method 8038 with

DR5000 (Hach). Experiments for each different condition were replicated twice. The wastewater samples were taken on daily basis until equilibrium was achieved and the characterization were done in triplicate for each sample obtained from the MFCs including the replicated systems. The average and standard deviation of the data for each parameter were then calculated and presented.

Voltage was continuously monitored and measured every hourly using a multimeter with data logger system (Fluke 287/289).). Polarization data were collected by varying the external resistance (varied from 100 KΩ to 10 Ω) with a variable resistance box (Tenma 72–7270) when the power production was stable.

Power density, P (mW m^{-3}) was calculated by

$$P = IV/v \tag{1}$$

where I is the current, V is the voltage, and v is the liquid working volume in the anode compartment.

The Coulombic efficiency (CE) is described as the percentage of electrons recovered from organic matter versus the theoretical maximum whereby all electrons are used for electricity production. CE was calculated by

$$CE(\%) = C_a/C_t \times 100 \tag{2}$$

where C_a is the total coulombs calculated by integrating the current over time, C_t is the theoretical amount of coulombs available from the oxidation of POME.

3. Results and discussion

3.1. pH profiles

The pH of the substrate inside the MFC-adsorption hybrid system was measured when the system was ready to be operated in the closed-loop mode. The pH was measured at the beginning of the experiment for all the MFCs operated at different operating temperature and also throughout the operation until equilibrium was achieved. Three different trends can be observed during the first 19 h: the temperatures of 15 °C, 20 °C, 40 °C and 55 °C are

under one similar trend; 25 °C under second trend; while 30 °C and 35 °C under another trend. At this point, the MFCs operated at the first trend temperatures had initial pH ranging in between 8.02 ± 0.1 and 8.31 ± 0.1. In the subsequent 19 h of operation, the pH profile dropped to pH ranging in between 7.38 ± 0.1 and 7.63 ± 0.2 before going up again and settled at pH 8.07, 8.36, 8.25 and 8.35 for MFCs operated at 15 °C, 20 °C, 40 °C and 55 °C, respectively. When the operating temperature of the MFC was 25 °C, the corresponding pH was found to be 7.85 ± 0.2 and 7.08 ± 0.3 at the beginning and at the end of experiment, respectively and the pH decreased to 6.98 ± 0.2 in the first 19 h of operation. MFCs operated at 30 °C and 35 °C having almost

similar pH profile with initial pH of 8.02 ± 0.2 and 8.07 ± 0.1 which settled at 7.40 ± 0.2 and 7.55 ± 0.2 with a pH drop of 0.62 ± 0.1 and 0.52 ± 0.1, respectively during the first 19 h of operation.

Generally from the lower part of the microcosm, organic matter found in the wastewater might had been oxidized into fatty acids under the anaerobic condition which shifted the pH towards the acidic side. With the absence of oxygen in the MFCs, pHs tend to be decreased due to the biodegradation of the organic matter in the wastewater in order to produce fatty acids [42]. Thus, this scenario explained the great fall of pH in the first 19 h of operation in MFCs operated at 30 °C and 35 °C, where the increase in microbial

Table 2
Pollutants removal efficiency under various operating conditions in between MFC-adsorption hybrid system and other MFCs.

Parameter	MFC-adsorption hybrid (Present study)	MFC-adsorption hybrid [32]	Other MFC systems	
	% removal		% removal	Refs.
COD	15 °C −82.5 ± 0.3% 20 °C − 84.2 ± 0.5% 30 °C − 91.6 ± 0.8% 35 °C − 93.5 ± 0.6% 40 °C − 83.1 ± 0.5% 55 °C −79.2 ± 0.4%	25 °C − 90.5 ± 0.3%	20 °C − 62% 35 °C − 84% 40 °C − 84% 55 °C − 58.8% 30 °C − 29.49%	[43] [44]
BOD	15 °C − 80.69 ± 0.4% 20 °C − 85.0 ± 0.3% 30 °C − 94.4 ± 0.5% 35 °C − 95.0 ± 0.3% 40 °C − 85.10 ± 0.6% 55 °C − 83.34 ± 0.4%	25 °C − 93.1 ± 0.5%	77% (temperature not mentioned)	[45]
TOC	100% for all the temperatures	25 °C − 100%	85% (temperature not mentioned) 25 °C − 75 ± 2% 30 °C − 22.82%	[46] [47] [44]
TS	15 °C − 19.9 ± 0.5% 20 °C − 27.9 ± 0.8% 30 °C − 38.5 ± 0.8% 35 °C − 41.4 ± 0.4% 40 °C − 34.0 ± 0.5% 55 °C − 29.9 ± 0.8%	25 °C −35.0 ± 0.3%		
TSS	15 °C − 82.3 ± 0.6% 20 °C − 88.7 ± 1.2% 30 °C − 97.7 ± 1.0% 35 °C − 98.0 ± 0.6% 40 °C − 88.8 ± 0.7% 55 °C − 83.65 ± 0.9%	25 °C −96.9 ± 0.2%	ambient − 86.3% 100% (temperature not mentioned)	[48] [9]
TVS	15 °C − 65.7 ± 0.6% 20 °C − 76.6 ± 0.5% 30 °C − 90.7 ± 0.4% 35 °C − 89.5 ± 0.6% 40 °C − 71.8 ± 0.5% 55 °C − 65.9 ± 0.3%	25 °C − 89.0 ± 0.2%	−	−
Color	15 °C − 92.0 ± 0.7% 20 °C − 95.3 ± 0.8% 30 °C − 96.0 ± 0.7% 35 °C − 94.0 ± 0.3% 40 °C − 91.0 ± 0.6% 55 °C − 90.0 ± 0.5%	25 °C − 98.1 ± 0.3%	29 ± 2 °C − 31.67% 30 °C − 34.16%	[52] [44]
Turbidity	15 °C − 86.6 ± 0.4% 20 °C − 91.5 ± 0.8% 30 °C − 98.00 ± 0.7 35 °C − 93.2 ± 0.5% 40 °C − 81.3 ± 0.6% 55 °C − 78.2 ± 0.3%	25 °C − 96.0 ± 0.4%	28 ± 2 °C − 96%	[53]
NH₃—N	15 °C − 0% 20 °C − 63.5 ± 0.6% 30 °C − 92.4 ± 0.4% 35 °C − 93.3 ± 0.8% 40 °C − 64.0 ± 0.5% 55 °C − 0%	25 °C − 89.6 ± 0.5%	23 °C − 30.4% 76% (Room temperature)	[54] [25]
TN	15 °C − 0% 20 °C − 52.6 ± 0.8% 30 °C − 98.00 ± 0.7% 35 °C − 87.3 ± 0.7% 40 °C − 48.0 ± 0.9% 55 °C − 0%	25 °C − 84.3 ± 0.4%	−	−

activities under these temperatures would tend to generate more fatty acids.

In comparison to the pH profiles in a non-hybrid dual chamber MFC done in the previous study, similar trend can be observed when operated in the range of 8–22 °C and 20–35 °C where the pH of the fresh feed (pH 7.4) reached the minimum value of about 6.2 and 6.7, respectively after operated for 20–25 h before the pH started to move up slowly and finally became almost constant [39]. Nevertheless, it is very difficult to compare this performance with the current study because the previous study was done in a wide range of operating temperatures whereas in the current study, the pH profiles was obtained at each specific operating temperatures. Besides, the pH profiles were not reported in the previous study for MFC operated at thermophilic temperatures (>40 °C).

3.2. Organics and solids removal

The results for all the pollutants removal efficiency under various operating conditions in between MFC-adsorption hybrid system and other MFCs were summarized in Table 2. In the previous study, MFC-adsorption hybrid system operated at 25 °C had recorded COD reduction of 90.5 ± 0.3% [32]. The performance of the COD and BOD removal percentage at various operating temperature were carried out in the present study and results were as shown in Fig. 2a. The optimum COD removal percentage was found to be at 35 °C, followed by 30 °C with the maximum removal of 93.5 ± 0.6% and 91.6 ± 0.8%, respectively. COD removal percentage at thermophilic temperatures (>40 °C) were 10.43 ± 0.2% and 14.37 ± 0.3% lower as compared to the performance at optimum temperature with removal percentage of 83.1 ± 0.5% and 79.2 ± 0.4% for MFCs operated at 40 °C and 55 °C, respectively. This scenario can be explained by the fact that there was very little mesophilic anaerobic bacteria growth which therefore inhibited the oxidation of organic matter found in the wastewater. Likewise, COD removal percentage were less effective by 11.04 ± 0.3% and 9.30 ± 0.5% when operated at 15 °C and 20 °C, respectively which

was mainly due to the lower degradation rate of influent organic matter at lower temperatures. Noted that the optimum operating temperature for effective COD removal in the present study was found to be at $\sim$35 °C which is different as compared to the previous study done in a non-hybrid dual chamber MFC (D_1) where the maximum COD removal reported was 84% when operated at 40 °C [43]. This may be due to the poor adsorption performance in the MFC-adsorption hybrid system when operated at elevated temperature (40 °C and above). As shown in Fig. 2a, the COD removal percentage gradient in between 15 °C and 35 °C in a hybrid system was 0.5889% C^{-1} as compared to 1.4467% C^{-1} in D_1 (non-hybrid MFC). This scenario proved that the pollutants removal performance of the hybrid system was less sensitive to the changes of operating temperatures. Likewise, when the hybrid system was operated at elevated temperatures, the gradient of COD removal percentage was 0.2626% C^{-1} as compared to 1.6800% C^{-1} for D_1. This phenomenon had demonstrated the importance of the MFC-adsorption hybrid system over the standalone MFC where the integrated system had proved a tremendous stability over a wide range of operating temperatures in terms of COD removal where the performance were not much affected by the temperatures. This is mainly due to the sorption characteristics and large surface area of the GAC in the anode chamber adopted in the present study which has the ability in removing the organic compounds from the substrate. Recently, a dual chamber MFC with graphite granules adsorption hybrid system had demonstrated removal of 29.49% of COD when operated at 30 °C [44]. The COD removal efficiency was however less as compared to present study due to the fact that the surface area of the graphite granules used was only 35.75 m^2/g, 96.4% lower compared to the surface area of the GAC used in the present study.

At the moment, there are no studies done to understand the removal of BOD with MFC-adsorption hybrid system at various operating temperature. Therefore, in the present study, the BOD removal percentage was understood and presented in Fig. 2a. In the previous study done with MFC-adsorption hybrid system at 25 °C, a BOD removal of 93.1 ± 0.5% was achieved [32]. However, in the present study, the highest BOD removal percentage recorded was 95.0 ± 0.3% at 35 °C, followed by 94.4 ± 0.5% at 30 °C which was therefore able to achieve the stringent effluent discharge requirement set by the DOE for the BOD parameter, where the final BOD achieved was 11.65 mg/l and 12.9 mg/l, respectively. In comparison to the previous study done in a non-hybrid semi-continuous air-cathode MFC, such system was only able to remove 77% of the BOD from landfill leachate [45]. The operating temperature was however not mentioned by the authors. Nevertheless, by comparing the BOD removal efficiency of these two different MFCs, it can be observed that integration of adsorption system to the MFC is an added advantage to the overall BOD removal performance.

Overall, it can be observed that regardless of MFCs operating temperature, the MFC-adsorption hybrid system has the ability to remove the entire TOC present in the wastewater (Fig. 2b). Tremendous TOC removals can be observed in the first 19 h of operation for MFCs operated in the range of 25 °C and 35 °C with removal of at least 90% and above. In the subsequent 24 h, at least 95% of TOC were removed from the substrate and achieved 100% of removal after operated in the MFCs for 67 h. For MFCs operated at temperature below 25 °C, a removal of 57.5 ± 0.8% and 62.3 ± 0.3% was achieved at 15 °C and 20 °C in the first 19 h and complete removal was achieved after 67 h of operation for both cases. For MFCs operated at 40 °C and 55 °C, the TOC removal rate were slower as compared to other operating temperatures where complete TOC removals were only achieved after 91 h and 115 h of operation, respectively. A non-hybrid dual chamber MFC was only able to remove up to the maximum of 85% of TOC after 96 h of
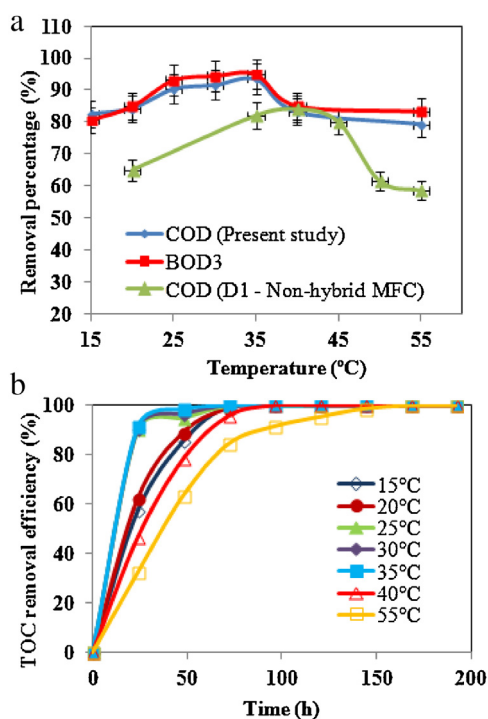

Fig. 2. (a) COD and BOD removal percentage at various operating temperature and (b) TOC removal efficiency over time.

operation [46]. On the other hand, dual reactor systems were fabricated with a single chamber MFC as a low-voltage power source to simultaneously accomplish hydrogen peroxide generation and iron (II) release for the Fenton reaction and such configuration was able to achieve 75 ± 2% TOC removal at 25 °C [47]. Thus, in the present study, the great superiority of integrating adsorption system to MFC can be observed where TOC can be removed completely at any operating temperatures.

In the previous study, the removal efficiency for TS, TSS and TVS achieved in the same system was 35.0 ± 0.3%, 96.9 ± 0.2% and 89.0 ± 0.2%, respectively when operated at 25 °C [32]. The removal percentage of TS, TSS and TVS at various operating temperature is as shown in Fig. 3a. The optimum operating temperature for TSS removal was found to be at 35 °C with removal percentage of 98.0 ± 0.6%. However, for TVS removal, the optimum performance was found to be at 30 °C with removal percentage of 90.7 ± 0.4%, followed by 89.5 ± 0.6% at 35 °C. For TS removal, the performance was generally about 20–40% lower as compared to TSS and TVS removals where the maximum removal was found to be at 35 °C with removal percentage of 41.4 ± 0.4%. Optimum temperatures for solids removal were typically ranged in between 30 °C and 35 °C, which can be explained by the fact that the MFCs anaerobic anode chamber has better tendency to reduce solids production at such conditions, where the bacterial biomass production by the MFCs system are much lower due to the anaerobic environment. A 90 L baffled non-hybrid microbial fuel cell stacked by five stacked modules was fabricated in the previous study for brewery wastewater treatment at ambient temperature and such system was able to remove SS as much as 86.3% which was about 10% less as compared to the current study [48]. Nevertheless, in comparison to another study done with MFC integrated with membrane bioreactor (MBR), such system was able to remove the entire SS present in the wastewater [9]. Therefore, in terms of solids removal, MFC-MBR hybrid system may be a better option in comparison to MFC-adsorption hybrid system because membrane type of unit operation has better solids rejection rate as compared to adsorption system.
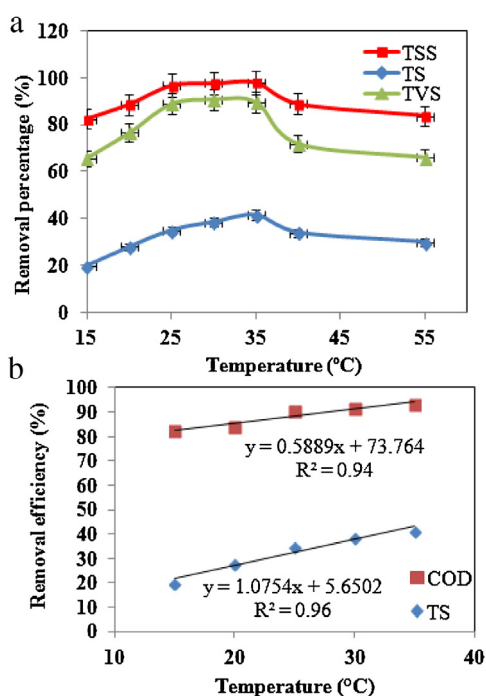

Fig. 3. (a) TS, TSS and TVS removal percentage at various operating temperature and (b) COD and TS removal efficiency as a function of temperature.

On top of that, we had discovered that there was a linear relationship between operating temperature and pollutants removal efficiency (Fig. 3b) at a rate of 0.5889% C^{-1} with regression (R^2) of 0.94 for COD removal and 1.0754% C^{-1} with regression (R^2) of 0.96 for TS removal. The removal for these two pollutants as a function of operating temperature were only applicable for operating temperatures ranging from 15 °C to 35 °C. In chemistry and biochemistry, the rate of change of a biological or chemical system as a consequence of the temperature by 10 °C is defined as below:

$$Q_{10} = \left(\frac{R_2}{R_1}\right)^{10/(T_2-T_1)} \quad (1)$$

Where Q_{10} is the temperature coefficient, R_1 is the removal rate at temperature T_1 (where $T_1 < T_2$), R_2 is the removal rate at T_2 (where $T_2 > T_1$), T_1 is the temperature at which the removal rate R_1 is measured (where $T_1 < T_2$) and T_2 is the temperature at which the removal rate R_2 is measured (where $T_2 > T_1$) [49]. Typically, Q_{10} value should fall in between 1 and 3 [50]. The COD and TS removal efficiency was taken as a measure of the rate of the electrochemical processes happening in the MFC-adsorption hybrid system and the temperature coefficient was calculated. It can be observed that the Q_{10} relationship between COD and TS removal efficiency with temperature was 1.06 and 1.45, respectively between 15 °C and 35 °C. The linear function of temperature on COD and TS in Fig. 3b were showing positive trends because the pollutants removal rate increased as a function of operating temperatures. The optimum operating temperatures can be seen to be ~25–35 °C which is about the temperature of the POME (~30–35 °C) when obtained freshly from the original source. Therefore, MFC-adsorption hybrid system can be ideally implemented in tropical countries where the tropical temperature of 30–35 °C is very close to the optimum operating temperature.

3.3. Color, turbidity, NH₃—N and TN removals

POME is generally associated with dark brown color. The presence of organic compounds such as anthocyanin and carotene pigment that were extracted from fresh fruit bunches in the sterilization process imparts this persistent color to wastewater [33]. Therefore, it is crucial for decolorization of POME to happen before it is suitable to be discharged to the river. In order to achieve this, the effect of temperature on color removal was investigated in order to find the optimum operating temperature for the removal. In the previous study done in the same MFC-adsorption hybrid system, a removal of 98.1 ± 0.3% was achieved when operated at temperature of 25 ± 1 °C [32]. In the present study, it was discovered that there was not much changes on the decolorization when operated in between 15 °C and 35 °C (Fig. 4). Maximum color removal was achieved at 25 °C and the decolorization performance were about 7–8% lower than optimum temperature when operated at elevated temperatures (>40 °C). Such behavior can be attributed by the fact that most of the color removals were responsible by the adsorption system in the MFC system where adsorption process was more effective at lower temperatures. The performance of color removal in the current MFC-adsorption hybrid system was typically attractive as compared to the removal efficiency of a non-hybrid air cathode MFC operated at 29 ± 2 °C where the removal percentage recorded was only 31.67% for distillery wastewater [52]. Therefore, adsorption system played a crucial role in removing majority of the color present in the wastewater.

Turbidity in the wastewater is typically contributed by the presence of the suspended solids. From Fig. 4a, optimum temperature for effective turbidity removal was found to be at 30 °C with the removal percentage of 98.00 ± 0.7 as compared to
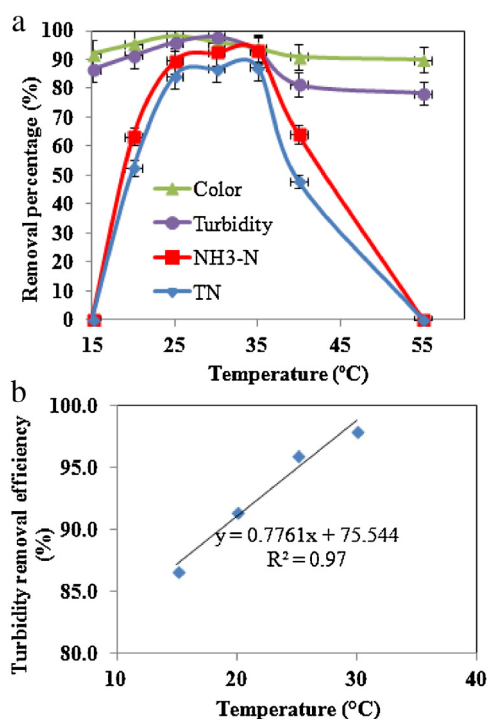
Fig. 4. (a) Color, turbidity, NH_3—N and TN removal percentage at various operating temperature and (b) turbidity as removal efficiency as a function of temperature.

$96.0 \pm 0.4\%$ when operated at 25 °C in the previous study [32]. As compared to the performance of the fuel cell which incorporated biocatalyst in the system and operated at ambient temperature (28 ± 2 °C) without the application of GAC [53], such system was able to remove turbidity maximum up to 96%, which was 2% less as compared to the MFC system in the current study. The operating temperature was found to be proportional to the turbidity removal efficiency at a rate of $0.7761\% \, C^{-1}$ with regression (R^2) of 0.97. Such relationship was only applicable for operating temperature range in between 15 °C and 30 °C. For temperature more than 35 °C and above, the performance started to decrease from $93.2 \pm 0.5\%$ at 35 °C–29.9 $\pm$ 0.3 at 55 °C. The temperature coefficient (Q_{10}) is found to be 1.09 between 15 °C and 30 °C. The linear relationship in between turbidity and operating temperature was showing positive trend because turbidity removal rate increased as a function of operating temperature with maximum removal observed at 30 °C.

The MFC-adsorption hybrid system has the ability to remove NH_3—N and TN as much as $89.6 \pm 0.5\%$ and $84.3 \pm 0.4\%$, respectively at the temperature of 25 ± 1 °C as reported in the previous study [32]. In the present study, it can be observed that the optimum temperature for NH_3—N and TN removals was at 35 °C with removal efficiency of $93.3 \pm 0.8\%$ and $87.3 \pm 0.7\%$, respectively (Fig. 4a). Such scenario may be due to the effectiveness of the ammonium ion (NH_4^+) migration in the anode chamber to the cathode which might eventually become nitrogen via further nitrification and denitrification reactions in the air cathode compartment under the optimum temperature. Similar phenomenon can also be observed in a non-hybrid dual chamber MFC operated at room temperature (~23 °C) where a total of 30.4% of the NH_4^+ migrated through the PEM to the cathode chamber for nitrification [54]. Since there was an effective removal of NH_3—N in the anode chamber when operated at 35 °C, TN concentration was greatly affected as well because NH_3—N was one of the component measured in TN, on top of other organic nitrogen,

nitrate nitrogen and nitrite nitrogen presented in the POME. In the previous study, an integration of dual chamber membrane-less MFC with adsorption hybrid system was fabricated with the anode chamber filled with graphite granules and tested for NH_3—N removal in POME [25]. Such system was able to demonstrate about 76% of NH_3—N removal after operated at room temperature for approximately 33 days. The performance of the MFC system in the present study has however demonstrated a better performance in NH_3—N removal which may be due to the difference in terms of anode fillers where GAC has better capability in removing NH_3—N as compared to graphite granules due to the high surface area of GAC. Besides, the configuration of single and double chambers MFC may have affected the NH_4^+ migration where the movement of NH_4^+ to the cathode compartment should be more effective in the single chamber MFC due to the wide cathode surface area.

3.4. Bioenergy generation

All the MFC-adsorption hybrid system were inoculated at 25 ± 1 °C until a stable voltage was achieved before switching them to respective operating temperatures of 15 °C, 20 °C, 25 °C, 30 °C, 35 °C, 40 °C and 55 °C in the closed-loop mode. At the first few hours of inoculation, the voltage generated was rather low but was however, started to demonstrate a steady increase after 100 h of inoculation in the MFCs. The low voltage generation during the first few hours might be due to the anode biofilm which were yet to be established and therefore limit the electron transfer to the electrodes. The MFCs were fed with fresh POME in a closed-loop mode after 200 h of inoculation at the rate of $6.12 \, L \, h^{-1}$ and COD loading of $884 \pm 3 \, mg \, L^{-1}$.

In the previous study, an operating temperature of 25 ± 1 °C was tested in the same MFC-adsorption hybrid system, fabricated from low cost, local ceramic material in a closed-loop mode and it had been reported that an average voltage of 106 mV was achieved after 171 h of operation [32]. In the present studies, the performance of the voltage generated from such system was further investigated at other operating temperatures of 15 °C, 20 °C, 30 °C, 35 °C, 40 °C and 55 °C and results were as shown in Fig. 5a.

From Fig. 5a, it can be observed that the voltage were moving downtrend especially for MFCs operated at temperatures of 15 °C, 20 °C, 40 °C and 55 °C until a stable voltage was achieved. The average voltage recorded were 71.74 ± 6.4 mV, 92.48 ± 4.3 mV, 64.45 ± 9.1 mV and 62.61 ± 11.5 mV, respectively. The voltage generations at these operating temperatures were however 20–45% lower as compared to the baseline studies done at 25 °C. The decreased of voltage generation at lower temperature may be due to inactive microbial activities [55]. Therefore, such phenomenon may affect the substrate utilization rate which therefore minimal substrate is being converted into bio-energy. Low voltage generation at elevated operating temperatures (>40 °C) may be due to the denaturation of enzyme which therefore, lead to low voltage generation as a result of poor electrons transfer to the electrode [37]. On top of that, when bacteria was exposed to high temperatures continuously, important elements presented in the cell such as proteins, nucleic acids or other temperature-sensitive matter may suffered an irreversible damage which therefore affect the cell function.

Operating temperatures of 30 °C and 35 °C had however achieved a more satisfactory magnitude of voltage generation with an average of 115.68 ± 8.2 mV and 125.69 ± 9.1 mV, respectively. Such phenomenon may be due to the increase in biomass growth and substrate utilization as the temperature increases which therefore, improve the microbial activities and bacterial attachment on the electrode [56]. Thus, electrons could be
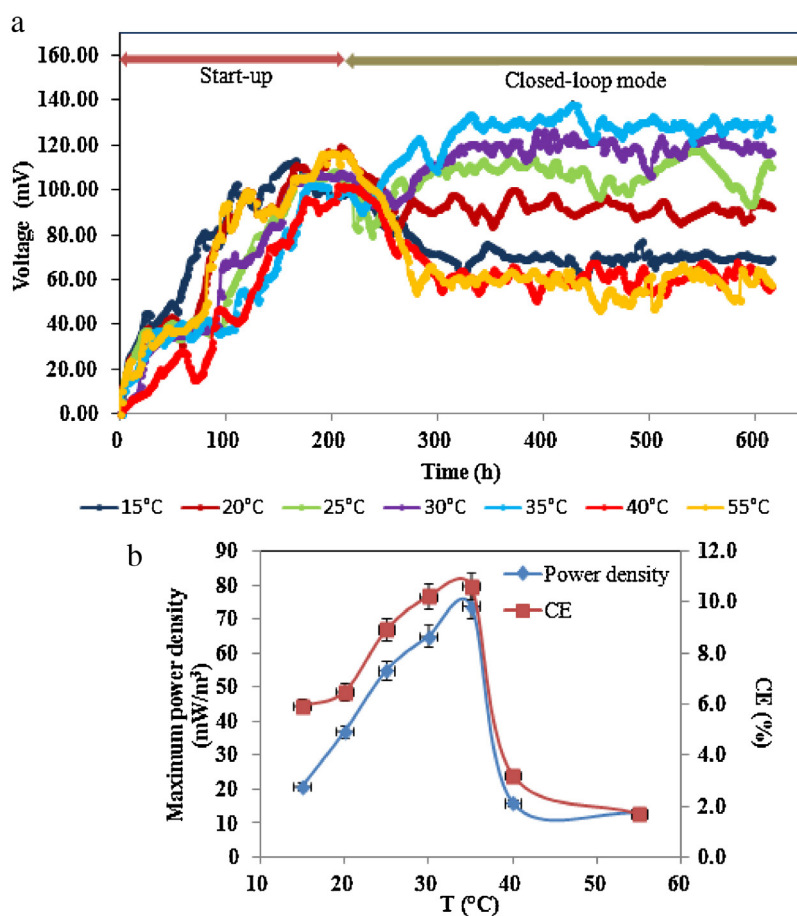
Fig. 5. (a) Voltage generation over 600 h of operation, (b) maximum power density and coulombic efficiency (CE) as a function of operating temperature.

transferred effectively to the anode electrodes which then helped to increase the voltage generation.

In the previous study, the maximum power density and coulombic efficiency (CE) was approximately $55\,mW/m^3$ and $8.94 \pm 0.3\%$ when operated at $25\,°C$ [32]. The same MFC-adsorption hybrid was tested for other operating temperature of $15\,°C$, $20\,°C$, $30\,°C$, $35\,°C$, $40\,°C$ and $55\,°C$ and the findings were tabulated in Fig. 5b. The power density and CE were calculated based on Eqs. (1) and (2). Generally, polarization curves were measured when the cell voltage become relatively stable which was after 300 h. The highest power density and CE were achieved when MFC was operated at $35\,°C$ with the results of $74 \pm 6\,mW/m^3$ and $10.65 \pm 0.5\%$, respectively. Generally, the performance at $35\,°C$ was found better as compared to the MFC-adsorption hybrid system operated at $25\,°C$ which may be due to the fact that the biofilms grown at $35\,°C$ are more electrochemically active as compared to those grown at $25\,°C$ which in turn helps in the transfer of the electrons to the anode electrode.

3.5. Comparison with other MFC-hybrid systems

MFC integrated with any unit operations or processes have the privilege of combining the advantages of each unit as compared to the standalone MFC [7]. Several studies have been done on MFC-hybrid system recently and a comprehensive comparison of these systems were done in terms of operating temperature, pollutant removal efficiency, MFC design, source of inocula, substrate and economics feasibility and presented in Table 3. Integrating MFC with membrane bioreactor (MBR) has been a trend in the recent years where it can be observed that such system is able to remove

COD with about 90% or more when integrated with tubular membrane bioreactor (TMBR) [9,10] and anaerobic fluidized bed membrane bioreactor (AFMBR) [57]. Besides, the effluent SS concentration was nearly nil with NH_3-N removal of more than 93.6% when operated at room temperatures ranging in between $14\,°C$ and $25\,°C$. There is no doubt that MFC-MBR hybrid system has the ability in removing organics and solids from the wastewater. Nevertheless, the efficiency of such system in removing other pollutants such as color, turbidity and nutrients are not reported. Besides, majority of the MFC-MBR hybrid system requires aeration system which therefore increases the operating cost and makes it impractical to scale-up. Even though no aeration system was involved in the MFC-AFMBR hybrid system, the adoption of expensive platinum (Pt) catalyst could be a major concern in terms of economic feasibility.

MFC integrated with adsorption can be an ideal combined system. On top of the typical biological electrochemical processes that normally take place in an MFC, physicochemical processes from the activated carbon happen simultaneously [32]. The activated carbon has the ability to remove a wide range of pollutants from the wastewater which may not be able to remove through the standalone MFC system. Therefore, any integration of MFC system with adsorption unit processes can be a great hybrid system to improve the wastewater quality besides deriving bio-energy from the system. Besides, the high surface area of the activated carbon can be a great platform for bacterial attachment for a more efficient electron transfer to the anode electron which improves the bio-energy generation. From Table 3, MFC-adsorption hybrid system in the dual chamber MFC [58] has reported a lower COD and color removals when operated at $30\,°C$ as compared

Table 3
Performance of MFC-hybrid system under different operating temperatures ranging from 25 °C to 35 °C.

	Dual chamber MFC-adsorption [58]	Two-stage MFC-immobilized biological aerated filter (I-BAF) [25]	Two stage MFC-anaerobic fluidized bed membrane bioreactor (AFMBR) [57]	MFC-adsorption [32]	MFC-adsorption (Present work)
Operating temperature	30 °C	35 °C	∼ 25 °C	25 °C	35 °C
Pollutants removed	COD − 71% (anode), 76% (cathode) Color − 73% (anode), 77% (cathode)	COD − 96.5% NH₃—N − 93.6%	COD − 92.5% TSS − ∼ 100%	COD − 90.5 BOD₃ −93.1 TOC −100% TSS − 96.9 TVS − 89.0 TS − 35.0 Color − 98.1 NH₃—N − 89.6 TN − 84.3 Turbidity − 96.0	COD − 93.6 BOD₃ −95.0 TOC − 100% TSS − 98.0 TVS − 89.5 TS − 41.4 Color − 94.0 NH₃—N − 93.3 TN − 87.3 Turbidity − 93.2
Anode	GAC and graphite rod	MFC 1 −Graphite granules and graphite rod MFC 2 − Carbon fiber felt and copper wire	GFB and titanium core wire	GAC and GFB	GAC and GFB
Cathode	GAC and graphite rod	Lead oxide electrode with copper wire	Carbon cloth with platinum (Pt) catalyst on water side and four polytetrafluoroethylene (PTFE) layers on the air side	ACFF and copper wire	ACFF and copper wire
PEM	Glass wool	No membrane	Two layers of textile separator	Ceramic pot	Ceramic pot
Source of inocula	Aerobic sludge from dye wastewater plant	Anaerobic digestion and activated sludge from sewage plant	Domestic wastewater from primary clarifier of wastewater plant	POME	POME
Substrate	Dye wastewater	POME	Domestic wastewater	POME	POME
Other characteristic	Low investment cost, high operating cost due to aeration. Two-chambers MFC.	Low investment cost, high operating cost due to aeration in both MFCs.	High investment cost due to Pt catalyst is expensive, low operating cost due to no aeration involve.	Low investment cost, low operating cost due to no aeration and no expensive catalyst involve. Single chamber MFC.	Low investment cost, low operating cost due to no aeration and no expensive catalyst involve. Single chamber MFC.

to the single chamber, air-cathode MFC-adsorption hybrid system when operated at 25 ± 1 °C [32] and 35 ± 1 °C (present study). Nevertheless, dual chamber MFC-adsorption hybrid system involves aeration system which can be very challenging in terms of scaling up and operating cost. Basically, the single chamber, air-cathode MFC- adsorption hybrid system operated at 35 ± 1 °C had generally demonstrated better pollutants removals as compared to the one operated at 25 ± 1 °C except for color and turbidity. POME for these studies was taken from the second anaerobic pond where the substrate temperature can range in between 30 °C and 35 °C. It is therefore practical for MFC-adsorption hybrid system with POME as the substrate to run at 35 °C for optimal pollutants removal where no further temperature adjustment is needed. Besides, such system does not involve the application of expensive catalyst as well as aeration system which makes it possible for scale-up application.

MFC integrated with immobilized biological aerated filter (I-BAF) operated at 35 ± 1 °C with POME as the substrate had generally demonstrated COD and NH₃—N removal which was almost comparable with the MFC-adsorption hybrid system operated at the same operating temperature [25]. Nevertheless, such removals through MFC-I-BAF hybrid system can only be achieved after operated for about 33 days as compared to MFC-adsorption hybrid system where the same removals can be achieved within ∼ 6 days of operation. Even though the MFC-I-BAF hybrid system was fabricated with cost effective materials, such system involved two aeration units which was consider not so practical for scale-up application.

4. Conclusions

In this work, we have presented findings from MFC-adsorption hybrid system, which was operated at six different operating

temperatures (15 °C, 20 °C, 30 °C, 35 °C, 40 °C and 55 °C) and the performance were compared to the previous work done in terms of pH profiles, bioelectricity generation, organics, solids, nutrients, color and turbidity removal. In the present study, it can be observed that the pollutants removal efficiency for POME with MFC-adsorption hybrid system is very much affected by the operating temperatures. The optimum temperature was found to be at 35 °C where such condition had demonstrated maximum removal of COD, BOD, TSS, TS, TN, NH₃—N and TOC. Besides, the maximum power density achieved was 74 ± 6 mW/m³ with CE of 10.65 ± 0.5% when operated at 35 °C. Other important findings from the present study include:

- Generally, MFC-adsorption hybrid system had demonstrated better pollutants removal efficiency as compared to standalone and other types of MFCs. This may be due to the application of adsorption system in the MFC which further improved on the wastewater quality.
- The performance of COD removal efficiency in a MFC-adsorption hybrid system was less sensitive to the changes of the operating temperatures where the performance was found to be ∼80–94% when operated in between 15 and 55 °C as compared to 60–84% when operated with standalone MFC in between 20 and 55 °C.

Such system is feasible and ideally to be implemented in tropical countries where the tropical temperature of 30–35 °C is very close to the optimum operating temperature as indicated in the current study.

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