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Ultrafiltration membrane bio-fuel cell as an energy-efficient advanced wastewater treatment system

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Summary

A novel anaerobic ultrafiltration membrane bio-fuel cell (UFBFC) was conceptualized to improve the efficiency of wastewater treatment with a reduced footprint and bioenergy recovery. The system with a working volume of 40 L reached a maximum power density of 23.3 mW/m^2 while treating synthetic wastewater with initial chemical oxygen demand (COD) of 2026 ± 61 mg/L and 19.1 mW/m^2 while treating actual fish processing wastewater with initial COD of 1915 ± 63 mg/L. The average COD removal efficiency of UFBFC was found to be almost similar (>97%) for both types of wastewaters showing the efficacy of the system in real-life applications comparable to the existing treatment systems. The cost-effective longitudinal vibrational mechanism (0.25 $/kLD of treated wastewater), with a frequency of 0.5 Hz and an amplitude of 8 mm, revealed superior anti-biofouling ability for ultrafiltration membrane placed inside the anodic chamber of UFBFC than cost-intensive conventional techniques (117 $/kLD in conventional MBR with aeration). The capacitor-based power management circuit improved the maximum normalized energy recovery (58 Wh/m^3) of UFBFC by over twice compared to UFBFC when operated with external resistance of 100 Ω (24 Wh/m^3). The harvested energy by UFBFC with capacitor-based power management system thus could successfully compensate the operational power requirement of the system, proving its practical field-scale applications as an energy-economic advanced wastewater treatment system.

Highlights

- Ultrafiltration membrane bio-fuel cell (UFBFC) for efficient wastewater treatment
- Cost-effective longitudinal vibration technique for reduced membrane biofouling
- Chemical oxygen demand removal efficiency of UFBFC was >97%
- Capacitor-based circuit improved normalized energy recovery of UFBFC
- UFBFC successfully used to treat synthetic as well as fish processing wastewater

KEYWORDS
anaerobic membrane bioreactor, biofouling mitigation, capacitor-based power management circuit, energy-efficient wastewater treatment, longitudinal vibration, microbial fuel cell

1 INTRODUCTION

As a consequence of anthropogenic activities, most of the water sources around the globe are jeopardized by the high amount of pollutant levels. That is why the wastewater treatment systems are essential for the dependent ecosystem to survive. On the other hand, the consumption of fossil fuels to provide energy for heating or power supply for operating the existing wastewater treatment systems must be minimized to reduce its negative influence on the environment.[1,2] Based on this, integrating bioelectrochemical systems (BES), viz. microbial fuel cell (MFC), along with membrane bioreactor (MBR), could be an important development in wastewater treatment added with a simultaneous bio-energy recovery for onsite wastewater treatment applications worldwide (Figure 1).[3,4]

Different combination of MFC-MBR systems have been experimented extensively to offer an attractive option for wastewater treatment with simultaneous bioenergy recovery in recent years.[5] The incorporation of MBR could be a game-changer to overcome the limitations of MFC, achieving improved biomass retention and organic matter removal efficiency.[6] On the other hand, the energy harvested by MFC can partially or wholly offset the energy requirement for the aerator or the filtration assembly in the MBR.[7] Although, the effective merging of these two technologies into a single unit for pilot-scale applications was challenging,[8] a few research works are being performed worldwide to address these issues of effective agglomeration of these two technologies.[9,10] According to the literature, MFC can be a pre- or post-integrated part of MBR to harvest bioenergy and treat wastewater effectively.[11,12]

Other than the said configurations, another standard format of this system is MBR as a post-treatment unit to MFC. This kind of system can significantly reduce membrane fouling and the deletion of the additional cathodic compartments. Wang et al. demonstrated such a combination where the dissolved oxygen contributed through the aeration tank of MBR act as a terminal electron acceptor .[3] An average current and maximum power density of 1.9 ± 0.4 mA and 6.0 W/m^3 were produced by this system, respectively.[3] Furthermore, the two-stage method of MFC and anaerobic fluidized-bed MBR has been shown to generate quality effluent with a minimum energy demand elsewhere.[13] An average chemical oxygen demand (COD) removal efficiency of 92.5% was reported for a hydraulic retention time (HRT) of 9 h with almost

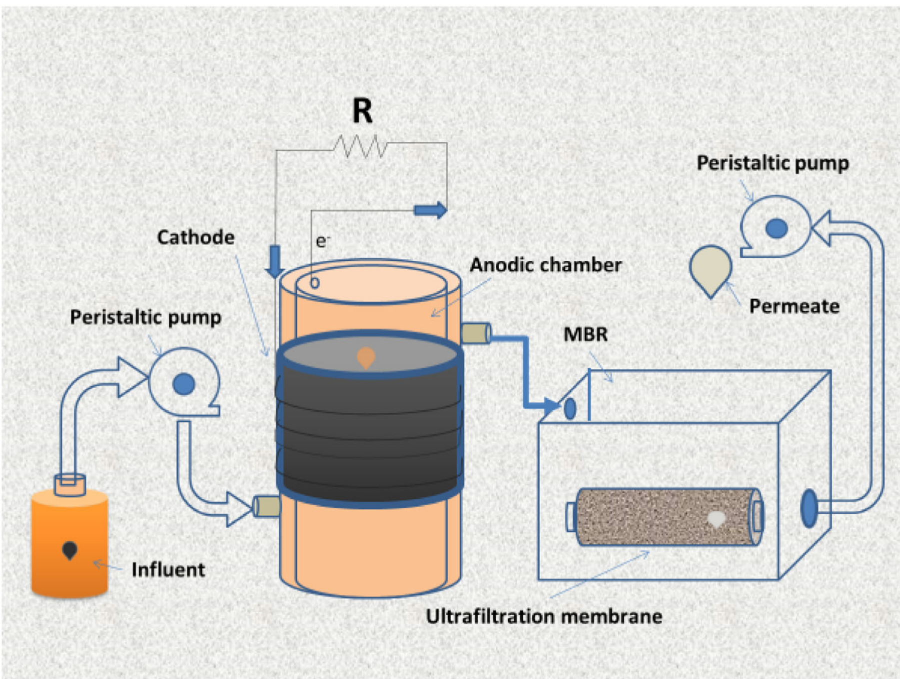


FIGURE 1 The schematic representation of bench-scale prototype of standard combined MFC-MBR system

complete removal of total suspended solids (TSS) by this unit. Moreover, the harvested energy by the MFC (20 Wh/m^3) was considered to be sufficient to compensate for the energy demand for operating the system self-sufficiently.[13] However, there is little literature on effective integration of both the technologies, with the possibility of further developments, especially in terms of reduced footprint and the higher energy recovery from the combined system.

Furthermore, there are few cases where the MBR has not been placed as pre- or post-treatment units but instead as a combination with intermediate units.[8] In one such investigation, the hollow-fiber (HF)–ultrafiltration (UF) membrane was positioned in an anodic chamber of tubular-MFC.[14] The system attained 90% of the COD removal efficiency and production of effluent with a turbidity of <1 NTU (Nephelometric turbidity unit) when it was treating synthetic wastewater while generating 3 to 25 Wh/m^3 of normalized energy. The polyvinylidene difluoride (PVDF) based HF membranes have been located along with the electrodes made with carbon brush and functioned sporadically to lessen the fouling of the membrane.[14] If we dig down other technologies tackling the wastewater treatment issues like Spirulina cultivating in wastewater to solve the purpose of nutrient removal as well as feedstock for biorefining, these technologies are also on the verge of further developments for their possible sustainable applications.[15] Therefore, additional opportunities need to be identified to improve the treatment efficiency of this integrated MFC-MBR system while introducing natural wastewater by enhancing the bioenergy recovery by modulating the energy harvesting system and mitigating the biofouling of filtration membranes.

Moreover, industries like fish processing produce enormous amounts of organic-rich wastewater either in soluble, colloidal, or particulate form with varying pollutants to the receiving water bodies. Therefore, stringent treatment facilities were implemented to meet the discharge norms, including activated sludge process, moving-bed biofilm reactor, etc., leading to an overall energy-intensive process.[16] Hence, there is research going on worldwide to find effective treatment systems to eliminate these issues with a reduced overall footprint.

Given the above, an anaerobic ultrafiltration membrane bio-fuel cell (UFBFC) was developed in this investigation. The aim was to reduce the footprint and cost associated with wastewater treatment drastically. Because fouling of membranes in the anaerobic conditions tends to be more intense, the longitudinal vibration technique was employed to mitigate the biofouling of the UF membrane module placed inside the anodic chamber. The capacitor-based circuit was acquainted with the UFBFC from 75 to 90 days of the operational period to understand the feasibility of circuit-amplified energy generation from UFBFC compared to when it was connected with an external resistance of 100 Ω for the rest of the operational period. For the first 90 days, synthetic wastewater was introduced to the system, followed by introducing fish processing wastewater for the next 30 days to validate the system's performance while treating a more complex real-life wastewater. The UFBFC was further examined for its performance in terms of bio-electricity generation, trans-membrane pressure (TMP) variation, and effluent quality for its energy-economic and sustainable real-life applications.

2 MATERIALS AND METHODS

2.1 Fabrication and operations of UFBFC

The UFBFC was constructed using 8 mm thick poly-(methyl methacrylate) fiber sheets with multiple (12) air-cathode combinations (Figure 2). Ceramic plates, 10 mm

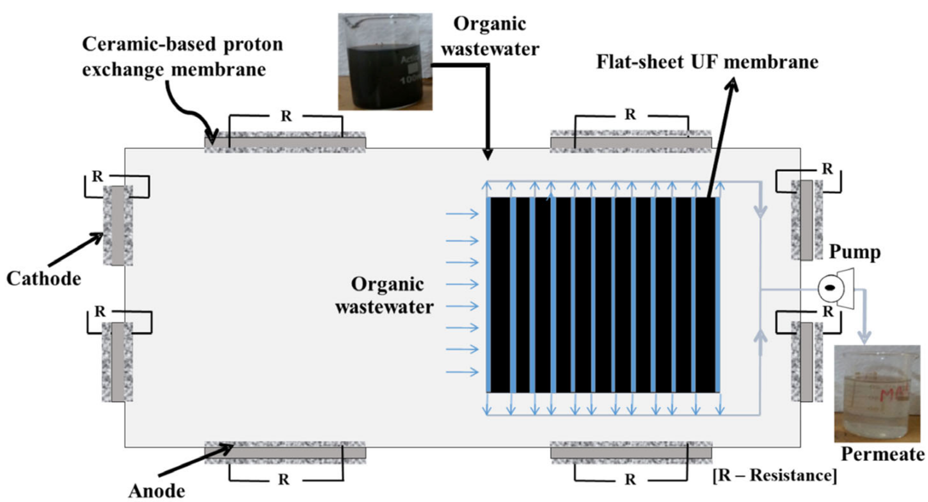


FIGURE 2 The schematic view of the UFBFC with a flat-sheet UF membrane assembly

wide, made of red-soil with 20% montmorillonite, were used as proton exchange membrane (PEM) as reported elsewhere.[17] The anodic chamber had a working volume of 40 L. The cathode was exposed to the atmosphere to evade further the need for active aeration to reduce the system footprint (Figure 2).[18] Twelve numbers of ceramic-based PEMs (four placed lengthwise with two at the upper side and the other two at the lower side and two width-wise on each side) with an effective exposed surface area of 47 cm^2 (each) were cut and placed like windows at the parapets of UFBFC. The pre-treated carbon felts were used as electrode (anode and cathode) materials for the UFBFC with a projected surface area of 47 cm^2 each (Figure 3). Iron phthalocyanine-modified carbide-derived carbon (FePc/CDC) was used as a cathode catalyst material due to its improved oxygen reduction reaction (ORR) characteristics in MFC, as discussed in our earlier research.[19]

The UFBFC was inoculated with the anaerobic mixed consortium collected from the septic tank of IIT Kharagpur, India, with an average volatile suspended solid concentration (VSS) of 19.9 g/L and TSS of 30.2 g/L. This inoculum was pre-treated with the 0.25% (v/v) chloroform to suppress the methanogenic consortia for the better exo-electrogenic activity of the biofilm.[20] The microbial dynamic study confirmed that the inoculum majorly consisted of *Pseudomonas* (13.79%), *Cloacibactrerium* (7.03%), *Peptostreptococcaceae* (6.73%), *Thiobacillus* (6.77%), etc. as reported in our earlier paper.[21]

The synthetic wastewater with sucrose as a carbon source was supplemented as a feed with trace nutrients

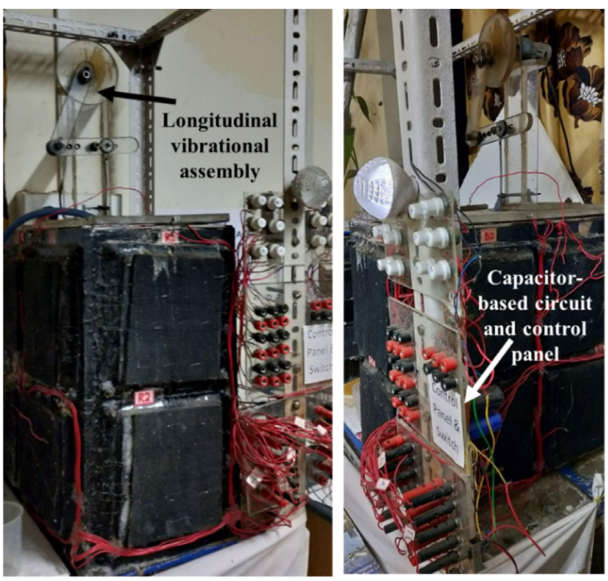


FIGURE 3 The UFBFC set-up with longitudinal vibration assembly for biofouling mitigation of UF membrane and capacitor-based circuit system

with an average COD concentration of 2026 ± 61 mg /L.[22] Over an operational period of 90 days, the synthetic sucrose-based wastewater was fed to the UFBFC followed by fish processing wastewater for another 30 days to validate the system's performance for the natural wastewater treatment with an average initial COD concentration of 1915 ± 63 mg/L. The fish processing wastewater was collected from the fish market present in the IIT Kharagpur complex and stored in the freezer for further use. The UFBFC was operated in the open environment under the temperature range of 26 ± 2°C.

The permeate flux rate (LMH) (L/m^3h) directly relates to the driving force, viz. the transmembrane pressure (TMP) and the total hydraulic resistance of the filtration membrane and its interfacial regions. The pressure drop across the membrane ΔP was referred to as TMP (bar or kPa), and the permeability was estimated as the flux ratio to the TMP (LMH/kPa). For clean water, the UF membrane used in this investigation showed a TMP of 2 kPa for a permeate flux rate of 40 LMH with a permeability value of 20 LMH/kPa, which was consistent over time. During the operational period of UFBFC, the flux due to permeation was retained constantly. A pressure gauge measured the TMP across the filtration membrane. Moreover, as per the instructions of the flat-sheet membrane manufacturer (Tech Inc., Pune, India), the preferred operating TMP was maintained below 25 kPa.

The material used for the construction of the filtration UF membrane was PVDF with a dimension of 465 mm (in height) × 340 mm (in width) × 7.5 mm (in thickness). The pore size of the filtration membrane was varied from 0.2 to 0.3 μm. The filtration membrane was periodically taken out of the UFBFC for off-line chemical washing and soaked into a solution of 500 ppm NaOH with a pH of 12 for 30 min. Whereas it was backwashed for 30 min during physical cleaning using tap water. The longitudinal vibration technique mechanism was also employed to avoid membrane biofouling as filtration membrane located within the anodic chamber of UFBFC, where the chances of bio-fouling were much higher (Figure 3). An additional longitudinal vibrating mechanism with 0.25 W motor arrangement was thus used with a membrane vibrating frequency of 0.5 Hz and at an amplitude of 8 mm to mitigate the biofouling as reported elsewhere.[23]

2.2 Performance monitoring of the UFBFC

The operating voltages (OVs) and the open-circuit voltages (OCVs) were measured at a regular interval using the data-acquisition set-up (Agilent Technologies, Penang

Malaysia). Furthermore, the polarization data were generated by altering the external resistance values from 20 kΩ to 10 Ω using the variable resistor (Decade Resistance Box, Bengaluru, India). The internal resistance of UFBFC was assessed from the slope of the current vs voltage graph. The power density (W/m^2) was estimated by the produced power (in W) per m^2 of the electrode surface area. The anode potential was measured simultaneously using Ag/AgCl based reference electrodes (CH Instruments Inc., RE-5B; +0.197 V vs SHE, Texas, USA) by placing it in the close vicinity of the anode. The electrical energy from the UFBFC was harvested for a duration of 15 days (from 75th to 90th days of the operational period) using a capacitor-based circuit, which consisted of 2 supercapacitors (100 F, DNATECHINDIA, India) and a microcontroller unit (XD-J16H, Xunda Corporation, China). The UFBFC was used to charge the supercapacitors in the parallel mode. The charged supercapacitors were then connected to a 0.1 W LED bulb in series to dissipate the stored energy while disconnected to the UFBFC.

When the UFBFC was connected to a capacitor-based circuit, it was charged and discharged regularly from the discharging potential (V_d) to the charging potential (V_c). The amount of charge (Q) being obtained in any charging cycle can be estimated by Equation (1).[24]

$$Q = nC(V_c - V_d) \tag{1}$$

where n is the no. of capacitors and C is the capacitance in F.

The coulombic efficiency (CE) was estimated based on COD removal in UFBFC when connected to a capacitor-based circuit according to Equation (2)[25]:

$$CE = Q/C_{th} \times 100\% \tag{2}$$

where C_{th} represents the quantity of charge available from the amount of substrate utilized.

Further, the CE was estimated at the steady-state mode of operation (when the current output remained relatively stable, that is, when UFBFC was connected to an external resistance of 100 Ω) by estimating the coulombs discharged compared to the maximum available coulombs present in the substrate consumed in the continuous-mode of operation as per Equation (3)[26]:

$$CE = \frac{M_s I}{F b_{es} Q \Delta COD} \tag{3}$$

where M_s is molecular weight of the substrate in g/mol, I is the current drawn from the MFC, ΔCOD is the changes in the substrate concentration in g/L, Q is the influent flow rate in L/d, F is the Faraday's constant, that is, 96 485 C/mol, and b_{es} is the number of generated electrons for each mol of substrate oxidation (mol of e^-/mol of the substrate).

Similarly, the energy (W_c) harvested from the UFBFC connected to the capacitor-based circuit was calculated for a single charging cycle using Equation (4)[24]:

$$Wc = \frac{1}{2}C\left(V_c{}^2 - V_d{}^2\right) \tag{4}$$

The normalized energy recovery (NER) was estimated by the energy harvested (W_c) to the effective anodic chamber volume, that is, kWh/m^3. The NER from the UFBFC at the steady-state mode of operation (connected to an external resistance of 100 Ω) was estimated in terms of the effective anodic chamber volume according to Equation (5)[27,28]:

$$NER = \frac{P \times t}{V_w} \tag{5}$$

where P is the power output estimated by $V_{cell}{}^2/R$, V_{cell} is the voltage in V obtained against the external resistance R in Ω, t is the retention time in h, and V_w is the effective anodic chamber volume in m^3.

The power requirement (in kW) for the suction pump was calculated according to Equation (6), considering pump shaft efficiency (η_s) and motor efficiency (η_m) of 75% and 65%, respectively[29]:

$$P = \frac{Q\rho gh}{3.6 \times 10^6 \times \eta_s \times \eta_m} \tag{6}$$

where Q is the flow rate in m^3/h, ρ is the liquid density in kg/m^3, h is the head in m, and g is the acceleration due to gravity (9.81 m/s^2).

The organic matter concentration in influent and effluent of UFBFC was analyzed in terms of reduction in COD values. The estimation of COD removal was done by the closed reflux method.[30]

3 RESULTS AND DISCUSSION

3.1 Electrical performance

The operating, open circuit, and electrode potentials were supervised during the whole experimental period to understand the effect of different substrate consumption and application of capacitor-based circuit on the performance of the UFBFC. The UFBFC could generate continuous bio-energy with an average OV of 320 ± 27 mV while treating synthetic wastewater for the first 90 days. However, while treating fish processing wastewater against 100 Ω of external resistance, an average OV of 299 ± 33 mV was observed for the last 30 days of the operational period. The corresponding current output is shown in Figure 4A, where a slight disturbance occurred when the filtration membrane was taken out for physical or chemical cleaning, evidenced on the 37th and 69th day of operation. The disturbance caused a slightly compromised electrical output for a couple of days because of slight disruption in the bacterioplankton population while lifting and placing back the membrane sheet, which is considered to affect the anodic microenvironment.[31,32]

Another disturbance occurred on the 91st day when fish processing wastewater was introduced to the UFBFC, and physical cleaning of the membrane was done (Figure 4A). Though the anodic biofilm was mature enough, the substrate complexity played a crucial role in achieving unstable current output values.[33] After a few days of operation, it eventually came back to slightly compromising average values compared to the synthetic wastewater treatment period because of complex wastewater characteristics.[34]

A transient state was achieved by the UFBFC when it was connected to a capacitor-based circuit as the current values changed abruptly due to cyclic charging and discharging phenomena causing varying resistance.[35] The current remained comparatively stable in steady-state conditions, that is, when UFBFC was connected to the fixed external resistor. The anode and cathode potentials were measured over the whole operational period at steady-state conditions except for operation days from 75 to 90 when a capacitor-based circuit was incorporated to understand the behavior of UFBFC under transient-state conditions.

At steady-state conditions, almost a constant cathode potential (208 ± 5 mV) was witnessed, indicating the indifferent contribution of FePc/CDC catalyzed air-cathodes for the whole operational period with and without natural wastewater. The efficient electrochemical performance and related ORR can be accredited to a higher amount of pyridinic-N, porous structure, high specific surface area, and metal-nitrogen-carbon active sites in the FePc/CDC material.[19] The variation in anode potential at the start-up phase and after introducing fish processing wastewater showed the development phase and acclimatization phase of anodic biofilm to the different substrates, respectively (Figure 4B). Anodic potential values improved until the first 30 days of operation and reached relatively stable values. After introducing fish processing wastewater, the anode potential got affected in the initial days due to complex substrate behavior. However, the average anode potential value remained almost the same as the synthetic wastewater treatment period.

The typical characteristics curve for the voltage output at three representative charging and discharging cycles for the capacitor-based circuit on the 80th day of the operational period is illustrated in Figure 5. Throughout the three charging cycles, the UFBFC was connected to the capacitors, and the voltage of UFBFC gradually increased to around 520 mV after 30 min of charging (Figure 5). In a complete charge condition, the UFBFC was functioned in open circuit mode, generating a higher voltage of around 550 mV (whereas at steady state, the

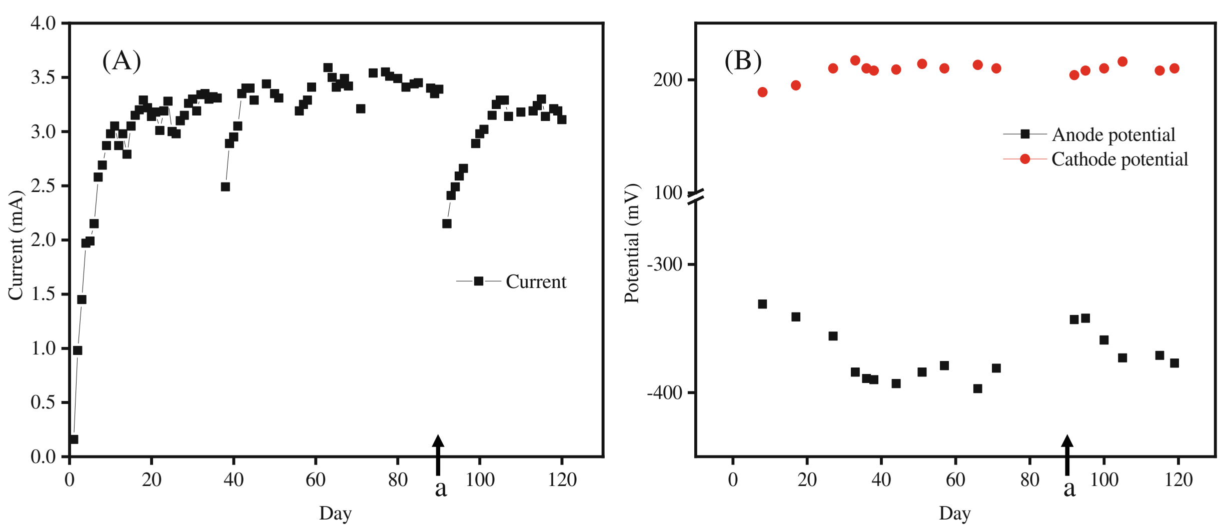


FIGURE 4 A, Variations in the current generation and B, anode and cathode potential values at the steady-state condition for UFBFC. The arrows A, indicate the introduction of fish processing wastewater to the system

OCV was 580 ± 10 mV). With the gradual increment of voltage output during the charging cycle, the anode potential drastically increased from - 102 ± 7 mV to −318 ± 7 mV after 30 min of charging. However, the cathode potential values remained almost the same (+205 ± 5 mV) (Figure 5).

The persistent nature of cathode potential values was because of the highly stable electrocatalytic behavior of FePc/CDC-based ORR catalyst applied in the UFBFC. The anode potential thus played a crucial role by attributing to the change in output voltage values when connected to the capacitor-based circuit. Moreover, the anodic microbiota showed acclimatized nature to the periodical changes in anode potential and a radical change in current values. The acclimatization and higher energy output were witnessed (as discussed in subsequent sections) because of these periodic changes in voltage values, which compelled the anodic biofilm to introduce a more negative charge to the anode surface due to less mass-transfer limitation as described elsewhere.[36] In addition, higher values of anode potential (more positive) were expected soon after the discharge process because the anode was connected to the capacitor directly at the start of the charging state, thus helping exoelectrogens to exchange more negative charges in the process.[37]

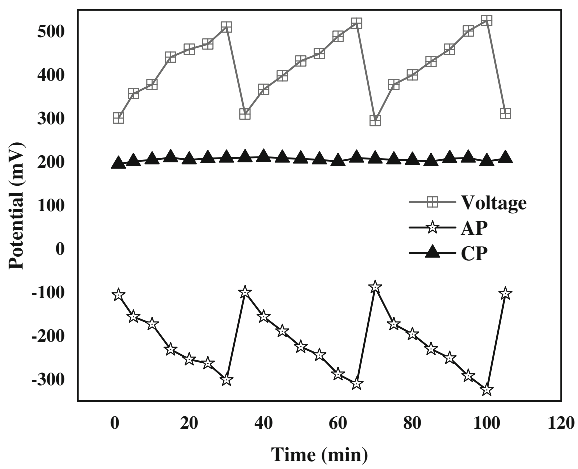
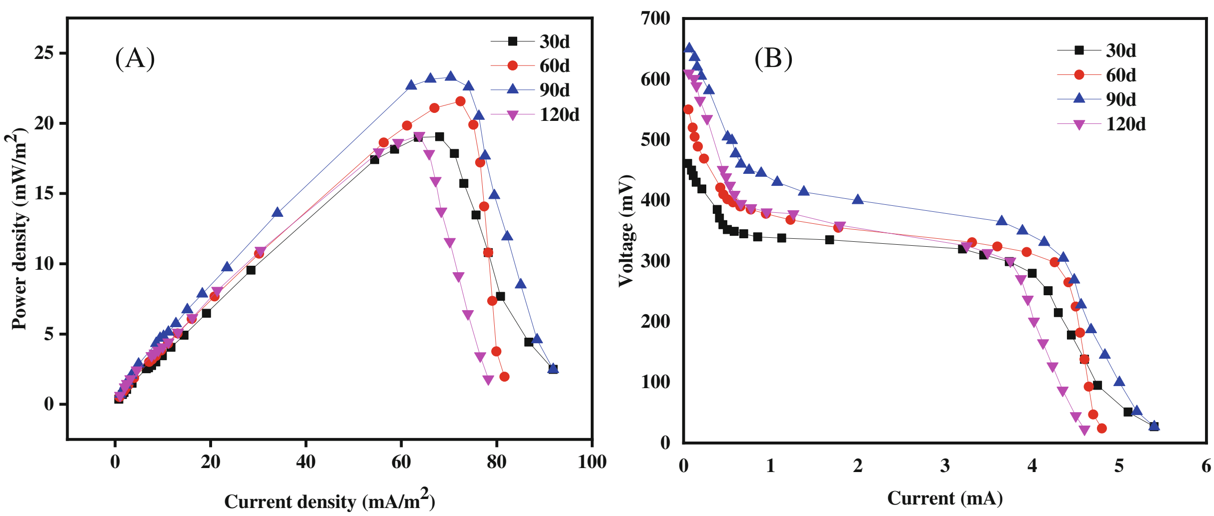


FIGURE 5 Charging and discharging cycle of the capacitor-based circuit in UFBFC. (AP – Anode potential; CP – Cathode potential)

Polarization was done after every 30 days of the interval to observe the performance chronology of the UFBFC (Figure 6A). For synthetic wastewater treatment, the maximum power density of UFBFC increased from 19.0 mW/m^2 (day 30) to 23.3 mW/m^2 (day 90), demonstrating the steady-state performance of UFBFC with a slight improvement in power performance over time (Figure 6A). During this treatment phase, a better adaptation of anodic biofilm and excellent robustness or chemical stability of FePc/CDC as cathode catalyst was confirmed.[19,38] However, after introducing the fish processing wastewater to UFBFC, a slight decrease in the power density value (19.1 mW/m^2) was witnessed. An increase in internal resistance value was witnessed over that time for UFBFC because of the anodic limitations due to the complex nature of the substrate for the anodic biofilm to utilize (Figure 6B). The same has been witnessed in earlier investigations with the MFC-based wastewater treatment systems upon introducing natural wastewater, which is still a matter of further scientific explorations.[4,39]



3.2 Wastewater treatment and membrane fouling mitigation

The anodic biofilm and planktonic microorganisms in the anodic chamber of UFBFC removed most of the organic matter present in the introduced wastewater with the organic loading rate (OLR) of 1.92 to 2.03 kg COD/m^{3}d. The average COD removal efficiency was 97.4 ± 0.3% and 97.4 ± 0.2%, respectively, for synthetic and fish processing wastewater. The final effluent with an avg. COD values of 52 ± 5 mg/L and 49 ± 4 mg/L were witnessed while treating the synthetic and fish processing wastewater. The results showed consistent performance almost throughout the operational period of 120 days (Figure 7A). This system's organic matter removal capacity was thus much higher than for any of its operational components, that is, MFC or MBR alone, as per previous investigations.[40] It evidently shows the applicability of this system for long-term industrial applications as well.

The conventional wastewater treatment processes, like the activated sludge process, are considered energy-intensive, where the sludge treatment and disposal become a significant obstacle as well.[41] A remarkable advantage of the UFBFC has been its higher organic matter removal efficiency compared to conventional activated sludge processes. The COD removal efficiency in UFBFC could reach an efficiency above 97%, whereas, for the traditional activated sludge process, the COD removal efficiency has been relatively lower (~70%).[42] A previous investigation on an integrated MFC-MBR system achieved 90% of COD removal efficiency with the HF-UF membranes being positioned inside the anode chamber of a tubular-MFC. However, it caused membrane fouling at a much higher rate (TMP rose from 5 to 26 kPa within 7 days)[14] as compared to the current investigation as discussed later (TMP rose from 5 to 21 kPa in around 30 days). Furthermore, the performance assessment of a two-stage MFC-anaerobic fluidized bed MBR demonstrated an average COD removal efficiency of 92.5% with an average energy recovery value of 20 Wh/m^3.[13] It was much lower than the present investigation (58 and 24 Wh/m^3, respectively, when connected to a capacitor-based circuit and external resistance of 100 Ω). Furthermore, the organic removal efficiency was higher than the investigations reported earlier with voluminous systems (>10 L) for the treatment of wastewater using MFC-based systems.[43,44]

Therefore, higher organic matter removal was witnessed for the UFBFC with much higher energy recovery and biofouling mitigation ability than previous research with segmental approaches similar to this investigation.[8,45]

Due to the anaerobic nature of the process, another advantage of the UFBFC is its much lower sludge yield than that for the conventional activated sludge processes[46] or the aerobic MBR technology.[39] Furthermore, any conventional MFC could not confine the solid concentration in its effluent; whereas, through the integration of a flat sheet UF membrane, the UFBFC improved the effluent quality with a much lower concentration of suspended solids (<10 mg TSS/L), while treating the fish processing wastewater.[14] The treatment efficiency could be improved further by optimizing the design and operating conditions of UFBFC, such as the anodic surface area to volume ratio, modulating the HRT in UFBFC, etc.

To alleviate the TMP increase and filtration membrane biofouling, the longitudinal vibrational mechanism was imposed with a frequency of 0.5 Hz and an amplitude of 8 mm to mitigate the membrane biofouling as

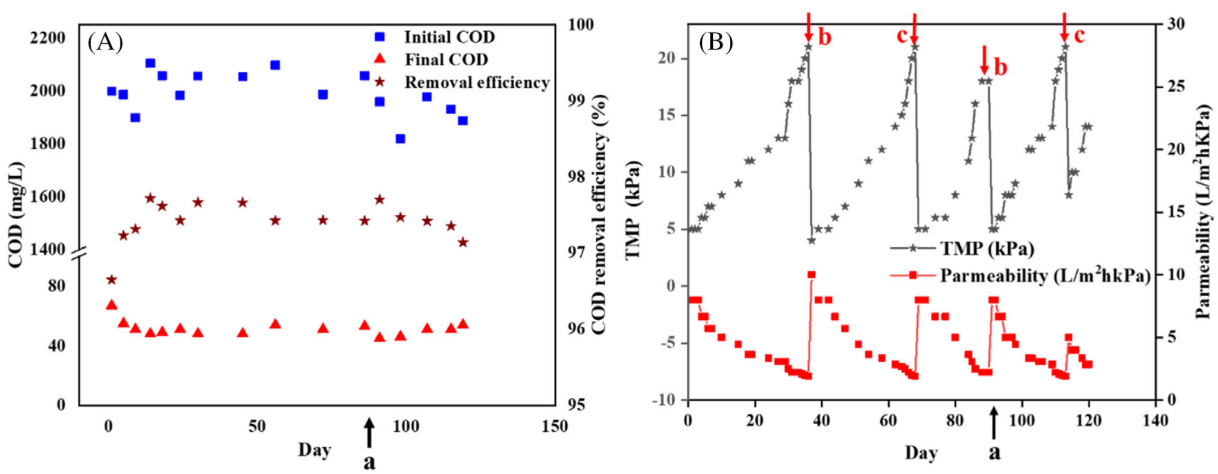


FIGURE 7 A, Initial and final COD, and COD removal efficiency of UFBFC, B, transmembrane pressure (TMP) and permeability variation over the operational period [A, introduction of fish processing wastewater, B, physical and C, chemical cleaning of membrane]

investigated for the flat-sheet UF membrane application in an anaerobic MBR elsewhere.[23] The pace of the TMP increment decelerated because of incorporating the longitudinal vibration technique, and it required about 30 days to upsurge the TMP from 5 to 21 kPa (Figure 7B). Whereas in a previous investigation on a membrane bioelectrochemical reactor (MBER) installed with hollow-fiber UF membranes inside a tubular MFC, it took only 7 days to increase the TMP from 5 to 26 kPa.[14] It clearly shows the benefits of the longitudinal vibrational mechanism in mitigating the biofouling of a flat-sheet UF membrane.

Following the instructions of the flat-sheet membrane manufacturer (Tech Inc., Pune, India), the preferred operating TMP should be maintained below 25 kPa. Therefore, after every 30 days, the filtration membrane was taken out and cleaned either physically or in a chemical way. The off-line cleaning of the UF membrane was soaked in a 500 mg/L NaOH solution followed by clean water as described earlier. Whereas it was backwashed for 30 min using clean water during physical cleaning. The aeration method has been generally adopted to mitigate biofouling in most MBR modules worldwide. However, the antifouling ability of the longitudinal vibration technique is far more efficient than the pure aeration method, as reviewed elsewhere.[23] Furthermore, the aeration cost can reach up to 117 \$/kLD in conventional MBR with aeration to mitigate biofouling of membrane compared to only 0.25 \$/kLD in the present investigation.[47,48] The intermittent vibration could further help conserve energy for vibrating the membrane of the UFBFC. In this case, a 15 min of vibration followed by a 45-min stationary period could result in a 75% drop in energy demand to keep the UF membrane from biofouling through this technique. The need for energy or nutrient recovery from the wastewater to achieve energy-neutrality and sustainability has been proved as demonstrated elsewhere as well.[49,50]

3.3 Energy recovery

The CE and NER values were estimated differently for different power harvesting mechanisms used in this investigation, that is, for capacitor-based circuits during 75th to 90th day of the operational period and with external resistance of 100 Ω for the rest of the operational period. The CE values were in the range of 2.9 ± 0.1% using the capacitor-based circuit and 1.0 ± 0.2% when UFBFC was connected with a stable external resistance of 100 Ω. Higher CE values can be witnessed when UFBFC was connected to the capacitor compared with when connecting it with an external resistor, which revealed that a higher amount of coulombs could be recovered in the transient operational mode of the UFBFC.[51] However, the CE values are much lower than the CE achievable by any chemical fuel cells which are because in this kind of bio-electrochemical system, substrates with high-molecular-weight converts to products with low-molecular-weight along with the production of some metabolites, especially in the case of real-life wastewaters which can reduce the performance of the system.[52] The same was witnessed and elaborated by Torres et al. in their investigation on glucose-fed MFCs, which showed much lower CE values than the simple acetate and propionate-based synthetic wastewaters.[53]

The mass-transfer limitations occurred in a steady-state condition with stable external resistance because of steady current flow from the biofilm to the anode surface.[54] Whereas, in the transient state with the capacitor-based circuit, the mass-transfer limitation can be overcome by the changes in the current output due to varying potential differences, which ultimately increases the electron production rate and, thus, improves the overall energy harvesting phenomenon of UFBFC as discussed elsewhere.[36] In addition, higher values of anode potential (more positive) were expected soon after the discharge process because the anode is connected to the capacitor directly in the transient state, thus helping exoelectrogens exchange more negative charges in the process.[37] This enhances the electron transfer rate from the biofilm to the anode surface to achieve a higher energy recovery.[36] The effect of the capacitor-based circuit on the increment in CE values seemed to be evident from this investigation as reported elsewhere.[37,54]

When attached to the capacitor-based circuit, the maximum NER achieved by the UFBFC was estimated to be 58 Wh/m^3 by Equation (4). On the other hand, the maximum NER was assessed to be 24 Wh/m^3 when it was connected to external resistance of 100 Ω at a steady-state mode of operation, as shown in Equation (5). This result further ascertained the efficacy of the capacitor-based circuit modeled UFBFC over mere external resistance attachment for higher energy recovery. Furthermore, the performance evaluation of a two-stage MFC-anaerobic fluidized bed MBR has shown an average NER value of 20 $Wh/m^{3.13}$ Another investigation has achieved a NER in the range of 3 to 25 Wh/m^3 with a similar system in treating synthetic wastewater.[14] These reports have shown much lower NER values than the current investigation because of incorporating a capacitor-based circuit in the present one.

Furthermore, a more significant fraction of the total energy (37 Wh/m^3) was consumed in the current investigation by the motor used in the assembly for longitudinal vibration of the UF membrane. Though, it is negligible compared to the energy requirements for the aeration (500-700 Wh/m^3) in the conventional aerobic MBRs.[55] Hence, the electrical energy produced by UFBFC at a transient mode (58 Wh/m^3) was more than the power requirement for the vibrating assembly (37 Wh/m^3), making the UFBFC an energy-economic wastewater treatment system. This is in accordance with the investigation by Ren et al., where the energy harvested by the integrated MFC-MBR system (20 Wh/m^3) was illustrated to be just enough to meet the energy demand to operate the system itself.[13] Another work reported that the membrane bio-electrochemical reactor (MBER) with the HF-UF membranes assembled into a tubular MFC produced 36 to 38 Wh/m^3 of viable energy to support the pumping system (28 Wh/m^3) while treating the synthetic acetate solution.[14] However, the MBER was not self-sufficient in energy demand when treating the natural wastewater, producing 3 to 25 Wh/m^3, which was much lesser than the energy demand for the pumping system itself.[14] However, the electrical energy produced by UFMFC at transient mode (58 Wh/m^3) could be successfully utilized to power the pumping system (17 Wh/m^3) and vibrator assembly (37 Wh/m^3). These results thus demonstrate that UFBFC can harvest the energy present in the wastewater to make the system with almost zero external energy input, thus making it energy-economic.

The outcomes of this investigation demonstrated that the UFBFC could harvest sufficient energy to suppress the overall energy demand to treat the synthetic and fish processing wastewater when connected to a capacitor-based circuit in the transient mode of operation. Better treatment efficacy was witnessed for the integrated MFC-MBR system or UFBFC compared to the MFC or MBR technology alone for treating natural wastewater. Although the feasibility of UFBFC was ascertained, a few different approaches are required, such as nutrient recovery, xenobiotic removal, etc., from the UFBFC to term the technology to be more sustainable for wastewater treatment along with bioenergy recovery.

4 CONCLUSIONS

The UFBFC was developed to treat real-life wastewater as a membrane-based energy-economic bio-electrochemical system. It used a longitudinal vibration technique with a frequency of 0.5 Hz with an amplitude of 8 mm to mitigate the biofouling and elongate the operational life of a flat-sheet UF membrane. The maximum power density of UFBFC increased from 19.0 to 23.3 mW/m^2 during three months of operation, demonstrating the system's robustness in harvesting electricity while treating synthetic wastewater. However, a slight decrease in power density was witnessed after introducing the fish processing wastewater because of substrate complexity. The UFBFC also achieved high COD removal efficiencies (>97%) for synthetic and fish processing wastewater. The results further ascertained the efficacy of a capacitor-based circuit modeled UFBFC over mere external resistance attachment for the higher energy recovery. However, further research is required to improve the coulombic recovery of the system upon optimizing the reactor constituents, majorly the electrode kinetics. Hence, UFBFC drastically lessens the overall footprint for the treatment of wastewater and enhances the treatment efficacy along with the bioenergy recovery to use it in energy-economic, sustainable real-life applications.

AUTHOR CONTRIBUTUIONS

G. D. Bhowmick, M. M. Ghangrekar, K. Tammeveski, and M. Wilhelm designed the investigation. G. D. Bhowmick, M. M. Ghangrekar, I. Zekker, and E. Kibena-Põldsepp performed the research. G. D. Bhowmick, M. M. Ghangrekar, I. Zekker, K. Tammeveski, R. Banerjee, and M. Wilhelm prepared the article. All authors have approved the final article.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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