

A system combining microbial fuel cell with photobioreactor for continuous domestic wastewater treatment and bioelectricity generation

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Abstract: A coupled system consisting of an upflow membrane-less microbial fuel cell (upflow ML-MFC) and a photobioreactor was developed, and its effectiveness for continuous wastewater treatment and electricity production was evaluated. Wastewater was fed to the upflow ML-MFC to remove chemical oxygen demand (COD), phosphorus and nitrogen with simultaneous electricity generation. The effluent from the cathode compartment of the upflow ML-MFC was then continuously fed to an external photobioreactor for removing the remaining phosphorus and nitrogen using microalgae. Alone, the upflow ML-MFC produces a maximum power density of 481 mW/m³, and obtains 77.9% COD, 23.5% total phosphorus (TP) and 97.6% NH₄⁺-N removals. When combined with the photobioreactor, the system achieves 99.3% TP and 99.0% NH₄⁺-N total removal. These results show both the effectiveness and the potential application of the coupled system to continuously treat domestic wastewater and simultaneously generate electricity and biomass.

Key words: wastewater treatment; microbial fuel cell; photobioreactor; microalgae; bioelectricity

1 Introduction

A microbial fuel cell (MFC) is a device that uses bacteria as catalysts to oxidize organic matters and generate current [1]. MFC is an emerging innovative technique for wastewater treatment and simultaneous power generation, which is especially important in offsetting the operational costs of wastewater treatment plants [2–3]. Besides energy recovery as electricity, MFCs can reduce as much as 50% of the electricity usage in the conventional biological treatment processes, and produce 50%–90% less solids than the conventional biological treatment processes [4]. In previous reports, various types of wastewater, including domestic and industrial wastewaters [5–6] have been successfully tested in MFCs with effective electricity generation. It has been reported that up to 80%–99% COD (chemical oxygen demand) removal could be achieved with MFCs [5–8].

High COD removal can be obtained using MFCs; however, this technology alone is not effective in

removing nitrogen and phosphorus. To improve nitrogen removal, systems combining a MFC with other nitrogen removal process have been developed [7–9]. Although these coupled systems have been proved to increase nitrogen removal, it is noteworthy that they have either not been able to completely remove the nitrogen or too complex to be used in practice. Besides, they did not resolve the removal of phosphorus. Up to now, it has not been reported that there are effective techniques for removing phosphorus from wastewater using MFCs.

Meanwhile, many researches have shown that microalgae are effective in removing nitrogen and phosphorus from wastewater [10–11]. Microalgae assimilate nitrogen and phosphorus in wastewater for synthesizing biomass in the process of photosynthesis as a result of removing nitrogen and phosphorus and purifying the wastewater.

Upflow mode membrane-less MFC (ML-MFC) inspired by the idea of in situ marine sediment MFC works in continuous upflow mode [12–14]. It omits ion exchange membrane and is simple in structure and easy in scaling, thus more cost-effective.

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The aim of this work was to develop an upflow ML-MFC and photobioreactor coupled system and evaluate the effectiveness of the coupled system for wastewater treatment, electricity generation and biomass production. Wastewater was first treated with the upflow ML-MFC to remove COD, phosphorus and nitrogen, and to produce electricity. Then, microalgae were cultivated using the MFC-treated wastewater to further remove the residual phosphorus and nitrogen and simultaneously produce biomass.

2 Materials and methods

2.1 Wastewater

Domestic wastewater was collected from the effluent of primary clarifier of Qingdao Tuandao Wastewater Treatment Plant (Qingdao, China), centrifuged ($8 \times 10^3 g$ for 10 min) to remove the particles with a centrifuge (CR22GII, Hitachi, Ltd., Japan), and stored in a refrigerator at $-20\text{ }^\circ\text{C}$ before use. The pH, COD, $\text{NH}_4^+\text{-N}$ and TP (total phosphorus) of the centrifuged wastewater were 7.37, (238.7 ± 7.6) mg/L, (52.56 ± 0.78) mg/L and (5.87 ± 0.02) mg/L (standard deviation, $n=3$), respectively. The centrifuged wastewater was used for all MFC tests without any additional treatment.

2.2 Configuration of upflow ML-MFC and photobio-reactor coupled system

Figure 1 shows the schematic diagram and photograph of the upflow ML-MFC and photobioreactor coupled system used in this work. The upflow ML-MFC was mainly consisted of a polymethylmethacrylate (PMMA) plastic cylinder (65 cm long and 8.0 cm in inner diameter) with one end sealed, a carbon fiber brush

anode, a carbon fiber brush cathode, and a glass wool layer and glass bead layer separator. The PMMA plastic cylinder was divided into anode compartment and cathode compartment by glass wool layer and glass bead layer. The anode compartment (24 cm in depth) was at the bottom, and the cathode compartment (31 cm in depth) was at the top of the PMMA plastic cylinder. Glass wool layer (4 cm in depth) and glass bead layer (4 cm in depth) were successively placed on the upper of the anode compartment, supported by a perforated PMMA plastic sheet (uniformly drilled with 3 mm holes and 2 mm hole spacing). An influent port was set at the bottom of the PMMA plastic cylinder for feeding substrate and an effluent port on the side near top of the PMMA plastic cylinder. Carbon fiber (Dalian Xingke Carbon Fiber Co., Ltd, China, with elastic modulus of 220 GPa) brush electrodes were constructed as described by LOGAN et al [15]. The brush anodes (22 cm long and 3.0 cm in diameter) were first acid treated with a solution of ammonium peroxydisulfate and sulfuric acid, then heat treated as previously described by FENG et al [16], and finally washed three times with deionized water before use in the MFC. The brush cathodes (24 cm long and 3.0 cm in diameter) were placed on a ceramic plate in a pre-heated furnace (SX₂-10-13, Longkou City Xianke Instrument Co., China) at $400\text{ }^\circ\text{C}$ for approximately 30 min to remove the organic binder from the surface of carbon fiber. Four brush anodes were uniformly fixed on the bottom of the anode compartment and four brush cathodes were placed on the upper of the glass bead layer. The distance between the top of the carbon brush anode electrode and the bottom of the carbon brush cathode electrode was approximately 10.5 cm. Copper wire was used to connect the circuit containing a $100\ \Omega$ load, unless stated otherwise. A round aeration tube was placed

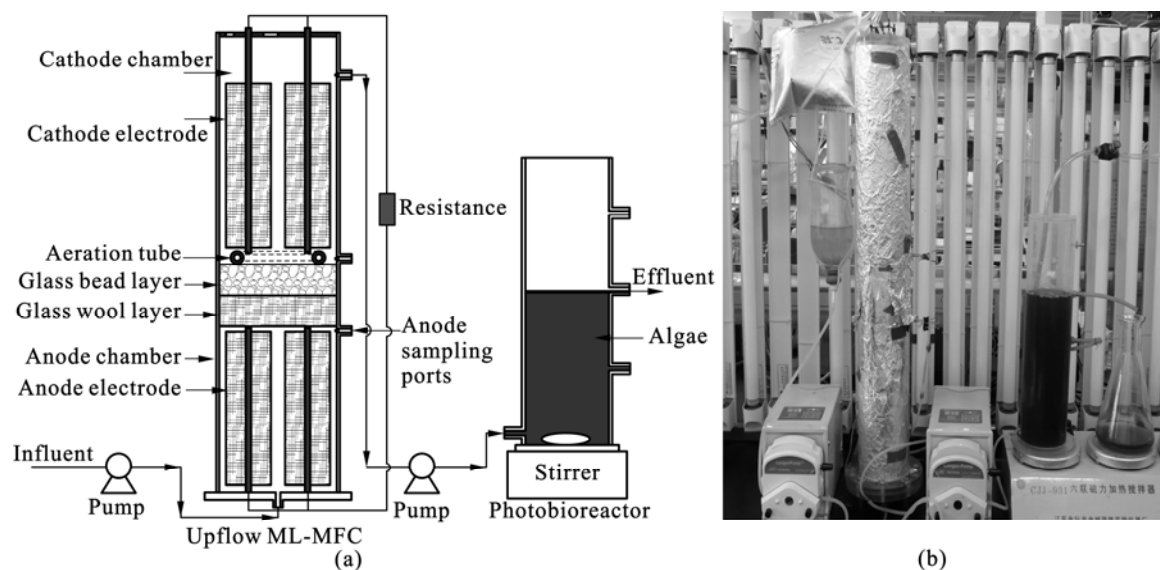


Fig. 1 Schematic diagram (a) and photograph (b) of upflow ML-MFC and photobioreactor coupled system

on the top of the glass bead layer. An external column photobioreactor (1.5 L) was coupled with the upflow ML-MFC.

2.3 Operation of upflow ML-MFC

The anode compartment was inoculated with a mixture of activated sludge and effluent of the primary clarifier (with a volume ratio of 1:7). The pH and volatile suspended solid concentration of the activated sludge were 6.39 and 1.6% (w/w), respectively. Besides, the pH and volatile suspended solid concentration of the inoculum were 7.02 and 0.3% (w/w), respectively. The centrifuged wastewater was purged with CO₂ and N₂ (with a volume ratio of 15:85) mixture gas to remove the dissolved oxygen, sealed and then sterilized (121 °C for 20 min) before use. The sterilized wastewater was continuously fed into the anode compartment from the bottom at a constant flow rate of 0.29 mL/min with a peristaltic pump (BT100-1J, Baoding Longer Precision Pump Co., Ltd., China), and the effluent exited through the cathode compartment on the side near the top. The cathode compartment was aerated with air at a rate of 120 mL/min using the aeration tube via an air pump (CX-0088, CHUANGXING Electrical appliances Co., Ltd., China). A polyester bag (1 L) filled with pure nitrogen gas was connected to the sterilized wastewater container through a silicon pipe and a disposable syringe filter (pore size 0.02 μm) to balance the pressure of the container. The voltage over the external resistance (R_{ext}) was recorded every 1 min to a desk personal computer via a data acquisition system (BC6040, Beijing BAOTRON Technology Co., Ltd., China). The upflow ML-MFC was covered with aluminum foil to exclude light. All experiments were operated at ambient temperature ((23±3) °C). When a maximum voltage was obtained, the anode was considered fully enriched with electro-active microbes. Then, the upflow ML-MFC was used for wastewater treatment. The voltage over the R_{ext} was recorded. Water samples (10 mL) were taken from the upper of anode chamber and effluent of cathode chamber every 24 h, respectively. The concentrations of COD, TP and NH₄⁺-N in the supernatant were measured after the water sample was centrifuged (8×10³g for 10 min).

2.4 Cultivation of microalgae

After the upflow ML-MFC voltage output stabilized, the effluent from the cathode compartment of the upflow ML-MFC was continuously pumped into a column photobioreactor (with a 7 cm inner diameter, a 40 cm column height and 850 mL of working volume) at a rate of 0.29 mL/min. A mixed culture of microalgae with an optical density of 0.331 (UV759S, Shanghai Precision & Scientific Instrument Co., Ltd., China), obtained from

domestic wastewater, was used as the inoculum. Mixture gas consisting of CO₂ and air (with a volume ratio of 2:98) was purged into the photobioreactor at a rate of 100 mL/min from the bottom of the photobioreactor with an aeration stone. The height of the aerated culture fluid was 24 cm. The culture, unless otherwise stated, was grown under a light intensity of 135.8 μmol/(m²·s) (LI-250A Light Meter, LI-COR, USA), with continuous illumination. After the OD₇₅₀ of the culture fluid stabilized, samples of the culture fluid (30 mL) were taken from the photobioreactor every 24 h during the cultivation, centrifuged (8×10³g for 10 min) with a centrifuge (Allegra X-22R, Beckman Coulter, Inc., USA), and the concentrations of nitrogen and phosphorus in the supernatant were measured.

2.5 Analysis

The power density, P (W/m³), was obtained according to the equation $P = IU/V$, where I (A) is the current, U (V) is the voltage over R_{ext} , and V (m³) is the volume of the anode compartment. The polarization curve was obtained by cutting off the circuit and establishing constant voltage, and then varying the R_{ext} over a range of 5 000–25 Ω, and the voltage over R_{ext} at 5 min intervals per resistor was recorded. The COD removal efficiency and Coulombic efficiency (CE) for a MFC running in continuous mode were calculated as described by LOGAN et al [1]. The polarization characteristics of anode and cathode were examined at different R_{ext} (5 000–25 Ω) by placing a reference electrode (Ag/AgCl) into the anode and cathode chambers, respectively.

The concentrations of COD, TP and ammonia nitrogen were measured by a multi-parameter COD Tachometer (5B-3(B), Lanzhou Environmental Protection Technology Co., Ltd., China). The concentrations of ammonia nitrogen and TP were determined by Nessler's reagent (HgCl₂-KI-KOH) colorimetric method at 420 nm wavelength [17] and ammonium molybdate spectrophotometric method at 710 nm wavelength [18], respectively. After the samples were digested at 120 °C for 30 min with potassium persulfate solution in sealed glass tubes, the TP concentrations of the samples were determined. COD was analyzed using fast digestion-spectrophotometric method [19].

3 Results and discussion

3.1 Enrichment of electro-active microbes

An up flow ML-MFC was used to enrich electrochemically active microbes using activated sludge and effluent of primary clarifier as inoculum and domestic wastewater as substrate. As shown in Fig. 2, during the electrochemically active microbe enrichment,

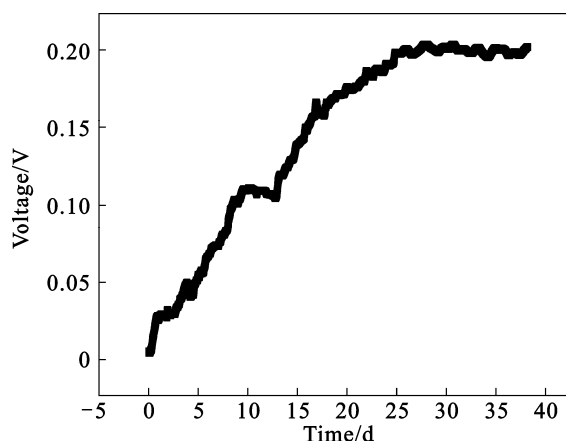


Fig. 2 Voltage output as function of time during start-up of upflow ML-MFC (Anode flow: 0.29 mL/min; Air flow: 120 mL/min; R_{ext} : 100 Ω ; Ambient temperature: (23 ± 3) $^{\circ}\text{C}$)

the voltage output of the upflow ML-MFC slowly increases. After approximately 40 d, the voltage output becomes stable and reaches an average value of 0.200 V, showing that the anode is fully enriched with electro-active microbe.

3.2 Performance of upflow ML-MFC

The performance of an MFC is often evaluated in terms of maximum power density, internal resistance (R_{int}), and CE. The maximum power density and R_{int} can be obtained from the power and polarization curves. As shown in Fig. 3, a maximum volumetric power density of 481 mW/m^3 (1 293.4 mA/m^3) with respect to the anode chamber volume is achieved at a cell potential of 0.372 V and optimal R_{ext} of 250 Ω . The maximum volumetric power density obtained is higher than the values (22.4–451 mW/m^3) reported in literatures for treating wastewater with similar MFC configuration [12–14], though the COD concentrations and ionic strengths of the wastewater used in the reported literatures are higher than those of the wastewater used in present work.

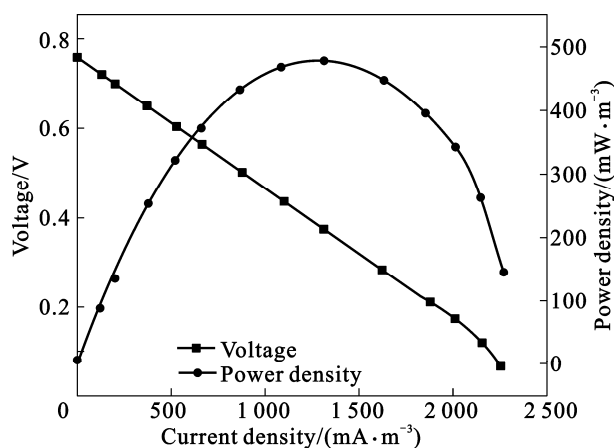


Fig. 3 Voltage and power density generated as function of current density of upflow ML-MFC

The high power density achieved in this work might be attributed to the use of carbon fiber brush electrodes, which have large surface areas compared to the electrodes used in the reported upflow ML-MFCs [12–13] and provide more sites for electrochemically active microbes adhesion and oxygen reduction, respectively. It has been proved that the power output of a MFC can be improved by increasing the surface area of electrode [15, 20]. LOGAN et al [15] achieved up to 2 300 mW/m^3 in an air-cathode MFC containing carbon brush anode electrode using wastewater, versus 965 mW/m^3 with a plain carbon paper anode electrode.

The R_{int} of an MFC is another important parameter to evaluate the MFC performance. The distribution and precise value of R_{int} are generally determined by impedance spectroscopy. For a linear polarization curve, the R_{int} of a MFC can also be obtained from the polarization curve as it is equal to the slope [1]. As shown in Fig. 2, the polarization curve of the SMFC is linear, so the R_{int} of the SMFC can be represented by the slope of polarization curve. The R_{int} of the MFC is approximately 256 Ω in this case.

CE demonstrates the ability of a MFC capturing electrons from the substrate as current and is an other important parameter in the evaluation of MFC performance. The CE of the upflow ML-MFC is 14%, a value that is consistent with the value (<20%) reported in the literatures for wastewater treatment also with upflow ML-MFCs [12, 14]. ML-MFCs simplify the reactor structure and reduce the manufacturing and operating costs, but at the expense of somewhat reduced CE. Low CE indicates that there is substantial COD that is not associated with power generation. COD removal in the absence of power generation may be as a result of oxygen transferring across the glass bead and glass wool layers on top of the anode, and a loss of COD using other electron acceptors (such as nitrate and sulfate) presented in the wastewater, and biomass production. The effect of oxygen diffusion from the cathode chamber into the anode chamber on CE is significant [21], leading to aerobic degradation of the substrate, resulting in less organic matter available for electricity production and consequently lowering the overall CE, as evidenced by low CE in previous study [3].

The changes in anode and cathode potential with the increase of current density are shown in Fig. 4. It can be seen that the cathode potential essentially is unchanged (from 0.362 V to 0.350 V) with the current density increasing. However, the anodic potential increases rapidly (from -0.351 V to 0.295 V) with the increase of current density, which indicates that severe polarization occurs in the anode. These results demonstrate that the upflow ML-MFC performance is mainly affected by the anode, which may include the anodic mass transfer, the

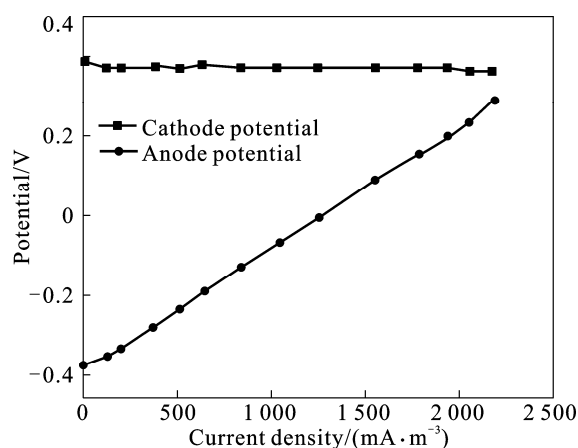


Fig. 4 Polarization characteristics of anode and cathode as a function of current density

rate of microbe oxidizing the substrate, and the rate of electrons transferring from microbes to anode electrode.

3.3 COD, phosphorus and nitrogen removals in upflow ML-MFC

The changes in the removal and concentration of COD, TP and $\text{NH}_4^+\text{-N}$ in the upflow ML-MFC are presented in Table 1. The COD of the influent is reduced from 238.7 mg/L to 91.3 mg/L in the anode chamber, and the COD in the effluent of anode chamber further decreases to less than 53 mg/L by feeding the anode effluent to the aerobic cathode. The average concentration of COD in MFC-treated wastewater meets the first level criteria (Class B) specified in discharge standard of pollutants for municipal wastewater treatment plant of China (GB18918—2002). A total COD removal of 77.9% is obtained with the upflow ML-MFC, and approximate 61.7% of influent COD is removed in the anodic compartment. These results demonstrate that the cathode chamber contributes to the total COD removal. High COD removal achieved in this work is due to the cathode process acting as an additional aerobic treatment step following the anodic treatment. LIU et al [5] demonstrated that MFC anodes did not have the ability to remove slowly biodegradable COD thoroughly. By feeding the effluent of anode to the aerobic cathode chamber directly, aerobic heterotrophic microorganisms growing in aerobic cathode chamber can use oxygen to hydrolyze and oxidize slowly biodegradable COD, thus enhancing COD removal.

As given in Table 1, the concentration of TP is reduced from 5.87 mg/L to 4.49 mg/L, with a total TP removal of 23.5%. Only 3.1% TP is removed in anode chamber, and the remaining TP is removed in the cathode chamber. These results demonstrate that the upflow ML-MFC anode alone is not effective in phosphorus removal, and the subsequent aerobic carbon-consumed phase following the anodic treatment promotes the TP removal. The removal of phosphorus is achieved in the upflow ML-MFC possibly in the following two ways: 1) conversion into biomass as part of microorganism growth and 2) excessive accumulation in microorganisms as polyphosphate. Microorganisms in the upflow ML-MFC successively go through an anaerobic process and an aerobic process, which is similar to the anaerobic/aerobic alternating enhanced biological phosphorus removal process (EBPRP), and is expected to favor phosphorus removal as EBPRP does. However, the upflow ML-MFC lacks the activated sludge that forms in the EBPRP and is rich in phosphorus-accumulating microorganisms, which inhibits the over-uptake of phosphorus by phosphorus-accumulating microorganisms. As a result, phosphorus can not be excessively accumulated in microorganisms as polyphosphate. Thus, phosphorus in the upflow ML-MFC is mainly removed by synthesizing new biomass in aerobic phase.

The concentration of $\text{NH}_4^+\text{-N}$ is reduced from 52.56 mg/L to 1.26 mg/L, and its total removal is as high as 97.6% (Table 1). Only 5.15% of influent $\text{NH}_4^+\text{-N}$ is removed by the anode chamber, and the cathode compartment accounts for approximately 92.45% $\text{NH}_4^+\text{-N}$ removal. These results show that most of the $\text{NH}_4^+\text{-N}$ is removed in the cathode chamber. A majority (68%–82%) of ammonia was oxidized and removed in the cathodic compartment through nitrification using a membrane-aerated MFC [8]. These results also show that conventional MFCs, only anode responsible for nitrogen removal, are not effective in removing $\text{NH}_4^+\text{-N}$. In the upflow ML-MFC, the effluent of anode is directly fed to the aerated cathode. $\text{NH}_4^+\text{-N}$ and other nutrients can then be utilized by aerobic heterotrophic microorganism growth in the cathode chamber to synthesize new biomass. Alternatively, $\text{NH}_4^+\text{-N}$ is converted to nitrate by nitrifying bacteria in the cathode chamber through aerobic nitrification, thereby reducing the $\text{NH}_4^+\text{-N}$

Table 1 Upflow ML-MFC performance for removals of COD, TP and $\text{NH}_4^+\text{-N}$

Item	Anode chamber			Cathode chamber	
	Influent	Effluent	Removal/%	Effluent	Removal/%
COD/(mg·L ⁻¹)	238.7±7.56	91.3±3.4	61.7±2.2	52.7±4.0	16.2±1.9
TP/(mg·L ⁻¹)	5.87±0.02	5.69±0.01	3.07±0.46	4.49±0.01	20.40±0.17
$\text{NH}_4^+\text{-N}$ /(mg·L ⁻¹)	52.56±0.78	49.84±1.47	5.15±3.59	1.26±0.11	92.45±3.78

COD—Chemical oxygen demand; TP—Total phosphorus; *—All “Influent” values are standard deviations ($n=3$)

concentration. Therefore, the upflow ML-MFC, having a sequential anode-cathode configuration, is more effective in removing nitrogen for wastewater treatment than conventional MFCs. The removal of $\text{NH}_4^+\text{-N}$ in MFC is a consequence of complex biological and physiochemical processes, which need deeper investigation in further work for better understanding of the mechanism for nitrogen removal and transformation in MFC.

3.4 Phosphorus and nitrogen removals in photobioreactor using microalgae

The effluent from the cathode compartment of the upflow ML-MFC was continuously pumped into a column photobioreactor to cultivate microalgae, which further removed the remaining phosphorus and nitrogen. The TP and $\text{NH}_4^+\text{-N}$ in the influent and effluent of the photobioreactor were measured and the results are presented in Table 2. The TP concentration is reduced from 4.49 to 0.03 mg/L, and 99% TP is removed at a hydraulic retention time (HRT) of 48.9 h. Meanwhile, $\text{NH}_4^+\text{-N}$ concentration is reduced from 1.26 to 0.52 mg/L, and a removal of 58.7% $\text{NH}_4^+\text{-N}$ is achieved. The majority of phosphorous and nitrogen present in the liquid medium is removed by the microalgae, which utilize these compounds to synthesize new biomass. Besides, a part of $\text{NH}_4^+\text{-N}$ might be removed by nitrification occurring in the reactor, as evidenced by concentration of $\text{NO}_x^-\text{-N}$ increased in a photobioreactor with algal-based immobilization process to treat the effluent from a secondary wastewater treatment plant [22]. A total removal of 99.3% TP and 99.0% $\text{NH}_4^+\text{-N}$ are achieved with the coupled system. After treated with the coupled system, the average concentrations of TP and $\text{NH}_4^+\text{-N}$ in treated wastewater meet the first level criteria (Class A) specified in discharge standard of pollutants for municipal wastewater treatment plant of China (GB18918—2002). To completely remove phosphorous and nitrogen, however, the algal biomass should be removed from the wastewater to prevent the microalgae from settling and subsequently releasing their stored phosphorus [23]. Simultaneously, the recovered biomass can be used as an energy source in the form of either biodiesel [24] or biogas [25–26].

Table 2 Micoalgae performance for removals of TP and $\text{NH}_4^+\text{-N}$

Item	Photobioreactor		
	Influent*	Effluent	Removal/%
TP/(mg·L ⁻¹)	4.49 ± 0.01	0.03 ± 0.04	99.12 ± 0.13
$\text{NH}_4^+\text{-N}$ /(mg·L ⁻¹)	1.26 ± 0.11	0.52 ± 0.03	58.73 ± 3.06

*—All “Influent” values are standard deviations ($n=3$)

4 Conclusions

1) A coupled system, consisting of an upflow ML-MFC and a photobioreactor, has been shown to efficiently achieve continuous domestic wastewater treatment and simultaneous electricity and biomass production. The coupled system is shown to improve phosphorus and nitrogen removal compared to wastewater treatment using only a MFC. It is also demonstrated that the coupled system can serve as a potential way to enhance pollutants removal and recover energy from wastewater treatment, providing a promising candidate for wastewater treatment and bioenergy generation. Furthermore, this novel coupled system for wastewater treatment and bioenergy generation has only been preliminarily explored and verified

2) The additional studies are needed to scale up and optimize the process in further, which includes the design of a more effective coupled system, the isolation of more effective microalgae, the effect of photobioreactor's and MFC's HRT on microalgae growth and pollutants removal and the recovery of biomass from the effluent.

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