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REMEDIATION TREATMENT

ENVIRONMENTAL PROGRESS & SUSTAINABLE ENERGY

The influence of organic pollutant load and external resistance on the performance of a solid phase microbial fuel cell fed orange peel wastes

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Abstract

The degradation of organic matter in marine sediments could be taken advantage of to produce electricity by using a sediment microbial fuel cell (SMFC) inspired system. A single solid phase microbial fuel cell (SPMFC) in which orange peel wastes were supplemented as a carbon source mixed to marine sediments produced a power of 0.33 mW and a voltage of 0.7 V. By stacking multiple SPMFCs powers of 2.08 mW were generated for a voltage of 4.6 V. The use of dewatered sludge to inoculate the marine sediment improved the SPMFCs' performance. The removal of organic matter in the SPMFC system under closed circuit conditions was very interesting, removal rates were 19%–40% from readily oxidizable organic matter,15 to 35% for loss on ignition and 22%–55% for total organic carbon, indicating the possibility of using these systems to treat solid organic wastes and produce electricity at the same time.

KEYWORDS
bioelectricity, dewatered sludge, orange peel waste (OPW), scale-up, sediment microbial fuel cells (SMFC), solid phase microbial fuel cell (SPMFC)

1 | INTRODUCTION

Microbial fuel cells (MFCs) have been recognized as an encouraging and challenging technology in saving energy and simultaneous wastewater treatment, overcoming environmental problems.[1] This technology is an excellent alternative for the generation of renewable and sustainable energy and has the potential to help alleviate the current global energy crisis.[2] He et al., [3] reported that applying MFCs to wastewater treatment is sustainable as a processes and has a good ability to directly convert substrate energy into electricity. The possible energy sustainability could be reached by the use of sedimentary MFCs, which help avoid energy expenditure necessary to produce the container for the system by introducing the electrodes directly in the environment. The sedimentary fuel cells (Benthic MFCs) seem sustainable from an energetic point of view, with a minimum functioning life of 2.7 years.[4] Facing the serious issues related to using chemical agents in the agro-industry which pollute soil and water sources, solid phase MFC (SPMFC) could be one solution, fermentative processes will allow the degradation of pollutants effectively while also producing energy.[5] In other words, the integration of SPMFCs with other conventional solid waste treatments could be used to produce sustainable green energy.[6]

SPMFCs are one of the developments in MFC technologies that can be applied to solid waste. These are claimed to accelerate the anaerobic waste degradation process, directly harvest electrical energy, and produce mature compost from organic compounds.[1] In SPMFCs the solid waste is used as a substrate to provide an environmentally friendly and sustainable source of electricity.[7,8] The bioelectrogenesis capability of microorganisms is employed to utilize organic compounds as electron donors to generate energy.[9] Bioelectricity was successfully generated utilizing organic wastes such as household food waste,[10] food waste hydrolysate,[11] vegetable and fruit residues,[9] mixtures of apples, lettuce, green beans, and soil,[12] kitchen and yard wastes,[13] vegetable and fruit wastes,[14] rice husks, soybean residue, coffee residue, and leaf mold.[15]

In this work we studied the performance of a solid phase MFC (SPMFC) inspired by sediment MFCs (SMFCs) and that would be used to treat a solid organic waste generated by the food industry: Orange peels.

Orange peel is one of the main wastes generated in large quantities following the consumption or the use of orange fruit. The Algerian Ministry of Agriculture and Rural Development shows that in 2011, the citrus culture covered a total area of 64,323 ha with a total production of around 1,100,000 tons. Orange contributes about 72% of the total production, a large part of which is used in the citrus industry where waste represents 50% of the product. Currently, these wastes are disposed of in landfills contributing to potential environmental issues related to solid wastes. It is known that the main constituents of orange peels are cellulose (9.1%), pectin (22%), and protein (6.6%),[16] therefore they could be used as a substrate, or carbon and energy source in MFCs. The main objective of this work is to see if a SPMFC could be used to treat orange peel wastes (OPWs), reducing the environmental impact of the juice industry while producing energy at the same time. The influence of the concentration of organic matter and the external load were studied. SPMFCs were also stacked to evaluate the possibility of a scale-up. Sediments inoculated with dewatered sludge served as a support for microorganisms and were mixed with the OPW, this allows a better repartition of the microorganisms through the solid organic substrate.

2 | METHODS

2.1 | Organic substrate

The organic substrate was prepared by grinding orange peel wastes until a homogenous paste was obtained. The paste was then fermented at room temperature for 20 days. The following quantities of OPW were added to the sediments: 15, 30, and 50 g. Control SPMFCs were made without any OPW addition and they were not inoculated. The composition of orange peel waste used in this study is presented in Table 1.

2.2 | Inocula, sediment, and seawater sampling

wDewatered sludge was collected from the anaerobic digester at the municipal wastewater treatment plant of Algiers (Algeria). The seawater and marine sediment were collected from the Mediterranean Sea in Algiers. The seawater was used as a cathodic medium.

TABLE 1 Composition of orange peel wastes

Component	Percentage (dry weight basis)
Moisture (% wet basis)	80
Protein	6.9
Sugars	45
Pectin	23
Cellulose	10.2

Marine sediments with volume of 300 ml were inoculated with 3 g of dewatered sludge after their collection.

2.3 | SMFCs' construction and operation

SMFCs used in this work consisted of a cubic Plexiglas container (10 × 10 × 10 cm) with a total volume of 800 ml and working volume of 600 ml (Figure 1). The bottom of the reactors were filled, up to a height of 4 cm with sediments inoculated with 3 g of dewatered sludge in which a certain quantity of OPW was mixed (as mentionned previously). The volume of seawater added was 300 ml.

The anode and cathode were cylindrical graphite rods with a diameter of 0.4 cm, length of 5 cm, and total projected surface area of 13.56 cm^2.

The anode was buried at a depth of 2 cm below the surface of the sediment and the cathode was placed 3 cm above the sediment–water interface. The SPMFCs' energy producing performance was monitored for 60 days and the degradation of the organic matter was evaluated by measuring readily oxidizable organic matter (ROOM) of the sediment-OPW mixture and its loss on ignition (LOI). The top cathodic medium level was maintained at a depth of 5 cm by routinely replenishing evaporated water with fresh medium. The anode and the cathode were connected with an external resistance (500 and 1000 Ω). All of the experiments were performed at room temperature (about 25 °C).

SPMFCs were stacked by connecting 3, 6, 9, and 12 SPMFCs in series.

2.4 | Analyses and calculations

LOI of the sediment was determined by weighing the sample before and after combustion at 550°C for 4 h.[17] The ROOM in the sediment was determined using the wet oxidation method.[18]

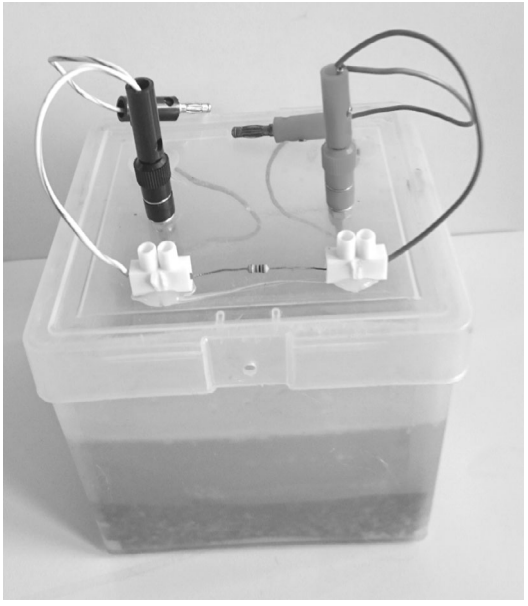

FIGURE 1 Solid phase microbial fuel cell (SPMFC) reactor

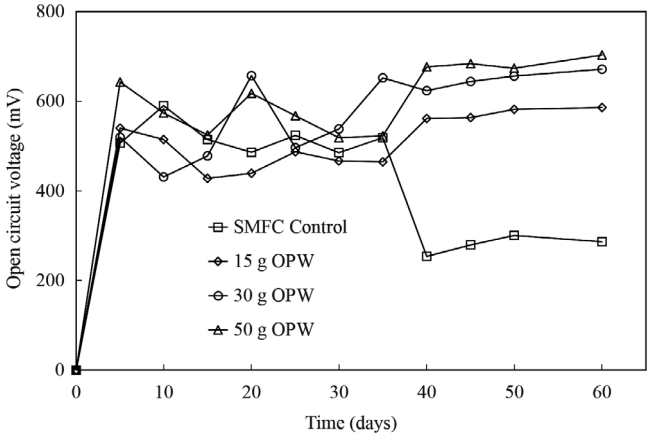

FIGURE 2 Voltage generation

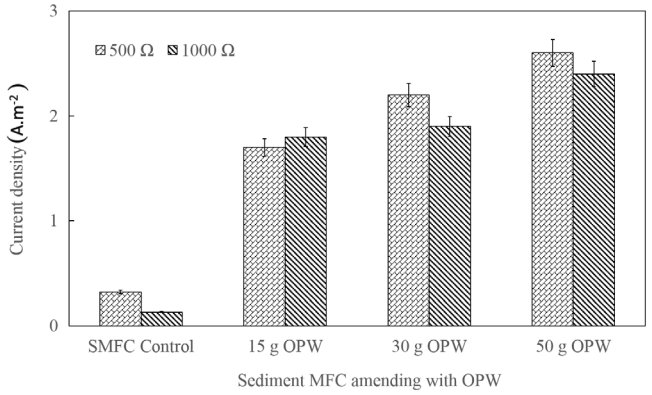

FIGURE 3 Current density generation

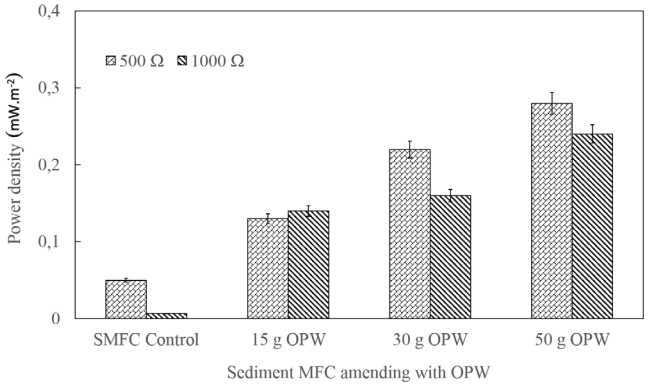

FIGURE 4 Volumetric power density generation

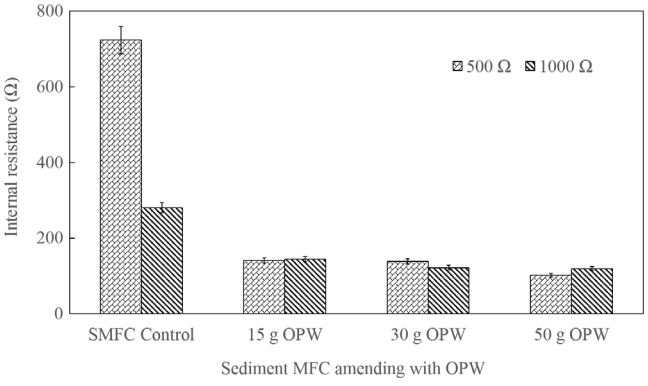

FIGURE 5 Internal resistance

The organic carbon content in the sediments was quantified as total organic carbon (TOC), which was measured using a TOC Analyzer (multi N/C 2100, Technology Quality Innovation analytikjena, Germany).

Voltage (V) generated by SMFCs during experiments was measured using a digital multimeter. Current (I) was calculated with Ohm's law ($I = V/R$), where R is the external circuit resistance. The power density was calculated with the formula $P = IV$ and normalized to the cathode projected surface area (A_{cath}). Polarization curves were obtained by varying the external resistance from 10 to 20,000 Ω and measuring the voltage after circuit stabilization at each resistance value (30 min). The internal resistance was calculated as the slope of the ohmic loss part of the polarization curves.

3 | RESULTS AND DISCUSSION

3.1 | Bioelectricity generation

The voltage produced by SPMFCs fluctuated during the first 5 days of operation; however, it increased rapidly afterwards in comparison to control SPMFCs (Figure 2). This phenomenon might be due to the formation of an electrochemically active biofilm on the anode surfaces.[19,20] After 35 days of operation, a maximum voltage of 670–700 mV was produced, in comparison, the control SPMFCs' voltage decreased to 250–280 mV. Later, the SMFCs reached a stable voltage and they continued operating for 25 more days (Figure 2). Previous studies[21] showed a decrease in the voltage after 25 days.

These results indicate that voltage generation with SPMFCs can reach a higher value that can be kept for longer when a carbon source is added to the sediments. The marine sediment inoculated with dewatered sludge allowed for a higher voltage, in comparison to control marine sediment. This is due to the formation of a mature, electro-active biofilm on the anode surface, which played a key role in producing a higher voltage.

The current density generation of SPMFCs as a function of OPW addition and external resistance is shown in Figure 3. The maximum current densities of 2.4 and 2.6 A m^{-2} were respectively obtained at resistances of 1000 and 500 Ω. The quantity of OPW had a significant influence on the SPMFCs' performance, a higher concentration of organic matter allowed for higher current density generation in comparison to control SPMFCs.

The current production was induced by the bacterial growth and their metabolic activity in the SPMFCs.[22] Additionally, a higher current production could be the result of an increased surface area for

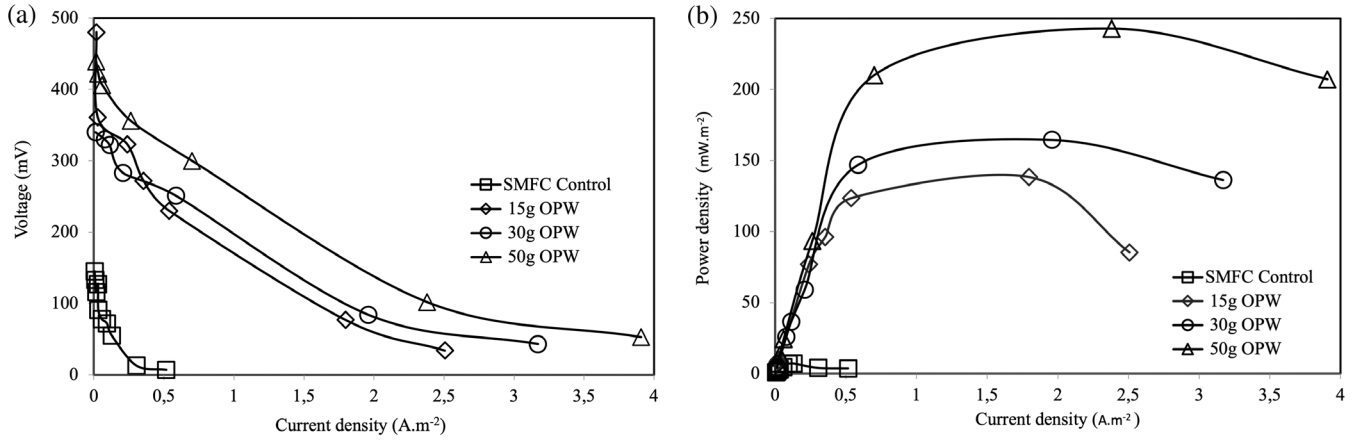

FIGURE 6 (a) Polarization curves of microbial fuel cell (SMFC) obtained with biofilm formed at external resistance 1000 Ω. (b) Power density curves of SMFC obtained with biofilm formed at external resistance 1000 Ω

the growth of bacteria, which is responsible for the generation of bioelectricity and a higher substrate availability.[23]

The experimental data indicate that the addition of OPW can significantly improve the SPMFCs' performance (Figure 4). A maximum power density of about 0.24 W m^{-2} was obtained with 50 g of OPW at an external resistance of 1000 Ω. Moreover, at the resistance of 500 Ω, a maximum power density of 0.28 W m^{-2} was achieved, in comparison to control SPMFCs. Indeed, the obtained power density was very low: 0.007 W m^{-2} at 1000 Ω and 0.05 W m^{-2} at 500 Ω. This process begins with a slow biodegradation of the organic matter and continues with a gradual oxidation caused by bacteria, therefore some organic matters in the sediment are degraded easily while the others may be oxidized slowly in a longer period of time.[24] Bhande et al.[25] showed that, compared to the unamended control, the addition of particulate substrates substantially improved and increased the power generated by SPMFCs.

The continuous current production without any inoculum supports the hypothesis that the microorganisms indigenous to the sediment inherently possess the biocatalytic activities responsible for the conversion of organic matter to electricity.[24]

Using sediments mixed with organic matter and seawater as a catholyte in the SPMFCs truly allowed a decrease of internal resistance (Figure 5). Also operating the SPMFC with a external resistance of 500 Ω significantly decreased the internal resistance (102 Ω) comparatively to a control SPMFC (724 Ω). At an external resistance of 1000 Ω the internal resistance decreases from a value of 280 Ω for the control to about 119 Ω from the SPMFCs amended with OPW.

Song et al.[17] show that the application of an external resistance of 100 Ω lead to a lower internal resistance than for 1000 Ω.

For the biofilms formed using an external resistance of 1000 Ω, the maximum power density with the highest (50 g) amendment of OPW reached a value of about 242.7 mW m^{-2} and generated the highest current density in the range of 2.3 A m^{-2} (Figure 6(a),(b)).

At 500 Ω, however, the SPMFCs were characterized by a higher power density of about 282.28 mW m^{-2} and a current density of 2.5 A m^{-2}, in comparison to control SPMFCs that only reached 50 mW m^{-2} and 0.5 A m^{-2} (Figure 7(a)).

The performance of these SPMFCs was more interesting than that obtained in previous studies (Table 2).

Rossi and Logan[30] explained that the power generation was mainly limited by internal resistance. In this study, the internal resistance was much lower than the ones stated in other works (Table 2).

When SMFCs are operated without replenishing the medium, the power significantly decreases through time due to the increase in ohmic and concentration losses, and finally reaches a point where no more power could be produced.[15] Majumder et al.[22] demonstrated that the internal resistance might restrict the transfer of protons through the electrolyte solution because of a greater overpotential at the anode, which was caused by mass transport loss or ohmic loss. This transfer resistance reduced the redox half-reaction at the electrodes, which in turn decreased power density.

The high external resistance can play an important role in reducing the power production. Aelterman et al.[31] demonstrated that the external resistance could affect the anode potential and the anode's resulting bioavailability for exoelectrogenic bacteria, which would further influence anode biofilm formation and MFC performance.

The results of this study suggest that the combination of OPW as a substrate amended in the sediment for the SPMFCs' conception with dewatered sludge as an inoculum could significantly enhance the power generation (Figure 4). The design developed for SPMFCs truly offers a solution for the internal resistance (Figure 5) and the improvement of the SPMFCs' performance in the course of 60 days (Figures 3 and 4). These SPMFCs are characterized by their great longevity and low internal resistance, in comparison to control SPMFCs. Gonzalez-gomboa et al.,[32] demonstrated that SMFCs could be operated for over 120 days..

In other words, these results suggest that the power generation could be sustainable and OPW could be valorized by using them as an energy source in SPMFCs.

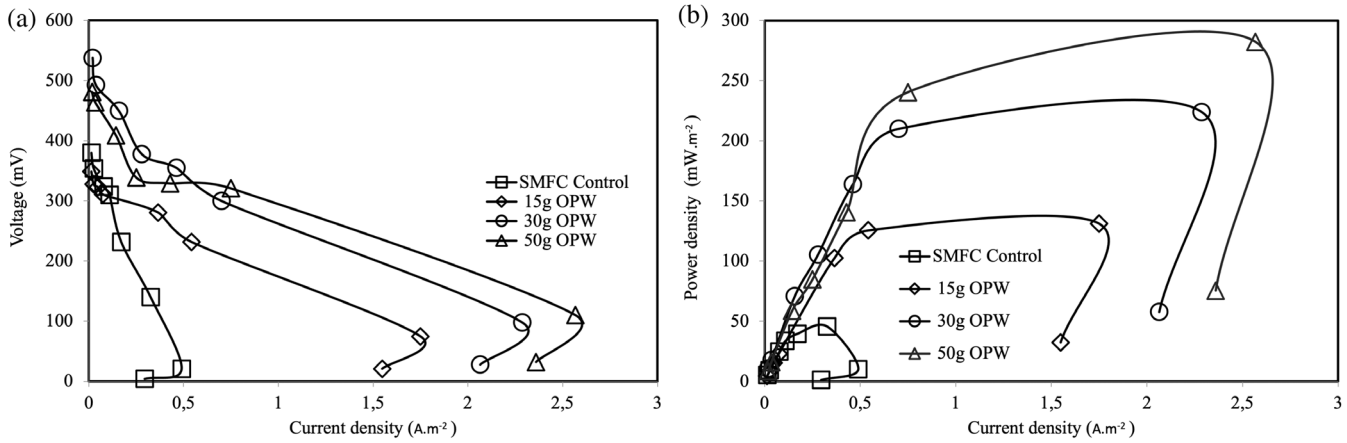

FIGURE 7 (a) Polarization curves of microbial fuel cell (SMFC) obtained with biofilm formed at external resistance 500 Ω. (b) Power density curves of SMFC obtained with biofilm formed at external resistance 500 Ω

TABLE 2 Comparison of solid phase microbial fuel cell performance

Operating phase	Particulate addition	Voltage (V)	Power density (mW m^{-2})	Internal resistance (Ω)	Reference
Solid (Marine sediment)	Orange peel waste (OPW)	0.7	282[a]	102	This study
			243[b]	119	
Solid (Lake sediment)	/	0.240	38[b]	489	26
Solid (Lake sediment)	colloidal iron oxyhydroxide	0.559	85.77[b]	138	27
Solid (Marine sediment)	/	0.680–0.720	62	/	28
Solid (Marine sediment)	Chitin 20	/	76[b]	646	29
	Chitin 80	/	80[b]	1297	
	cellulose	/	83[b]	1762	
Liquid	Food waste hydrolysate	0.57	173[a]	/	11
Solid (Soil + Compost)	Mix of apples, lettuce, green beans	/	5.29	/	12
Solid (Soil + Compost)	Kitchen and yard wastes	0.7	39.2	/	13
Solid (Soil + Compost)	Rice husks, soybean residue, coffee residue and leaf mold	/	4.6	/	15

[a]With 500 Ω resistor.
[b]With 1000 Ω resistor.

3.2 | Organic matter removal in sediment by SPMFCs

Previous studies proved the advantages of using SMFC for in situ sediment remediation and energy production.[33,34] In order to investigate the possibility of using SPMFCs for the depollution of organic wastes while generating electricity, the organic content of the sediment were analyzed after 60 days of operation for different external electric loads. The highest organic matter removal was observed with an external load of 500 Ω, which was in good agreement with the fact that the highest power output was achieved under the smallest external loading. As presented in (Figure 8), LOI removals were 25%, 26%, 34%, and 35% in Control SMFC, and the SPMFCs with organic contents of 15, 30, and 50 g respectively, at 500 Ω. The SPMFC with 50 g of OPW showed the highest LOI removal as well as the highest power density. At a 1000 Ω external loading, the same ascertainment of LOI removal was observed. The results reached 13%, 14%, 27%, and 29% at the end of the experiments from Control SMFC, and for 15, 30, and 50 g of OPW respectively.

As seen in (Figure 9), ROOM removal was significant. ROOM values decreased by about 12%, 19%, 24%, and 39% at 500 Ω for control and for initial OPW loadings of 15, 30, and 50 g respectively.

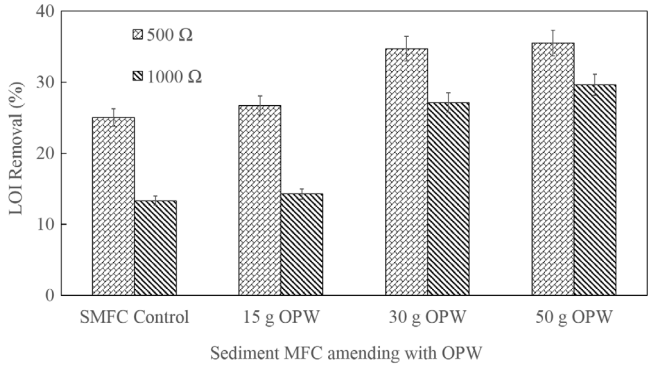

FIGURE 8 Loss on ignition removal (%)

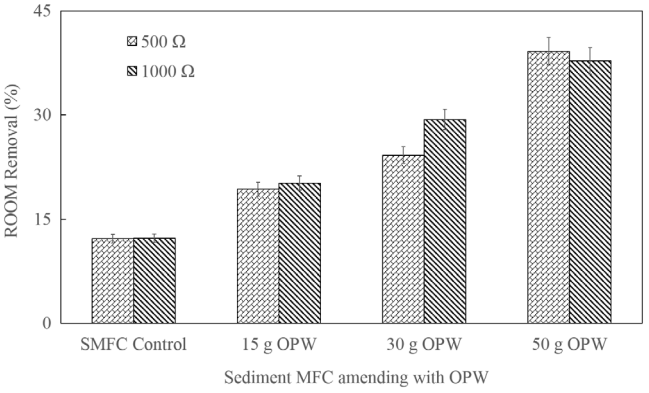

FIGURE 9 Readily oxidizable organic matter removal (%)

Under a 1000 Ω external loadings, the ROOM removal efficiency were 12%, 20%, 29%, and 37% from control and for initial OPW loadings of 15, 30, and 50 g respectively. These results indicated that the highest ROOM removal was observed for the highest initial OPW load of 50 g with a 500 Ω external loading.

Song et al.,[17] indicated that the highest LOI and ROOM removal efficiencies were achieved by SPMFCs with a lower external resistance of 100 Ω, they also noted that removal efficiencies of ROOM by SPMFCs at 100 Ω external resistance was significantly different from those at a 1000 Ω. Although the external resistance in an SPMFC plays an important role in the removal of ROOM rather than LOI in sediments.

Under closed-circuit conditions, organic matter content in terms of LOI and ROOM decreased significantly. These results suggest that the anode served as an alternative electron acceptor in the presence of biodegradable organic matter under electricity generating conditions.[35]

TOC removal efficiency was observed after 60 days of operation (Figure 10). In the control, TOC removal reached 16.73%. For cells containing 15, 30, and 50 g OPW TOC removal was 22.09%, 32.09%, and 52.82% respectively at a 1000 Ω external resistance. With a 500 Ω external resistance, the control showed TOC removal efficiency of 18.56% and the SPMFCs supplemented with 15, 30, and

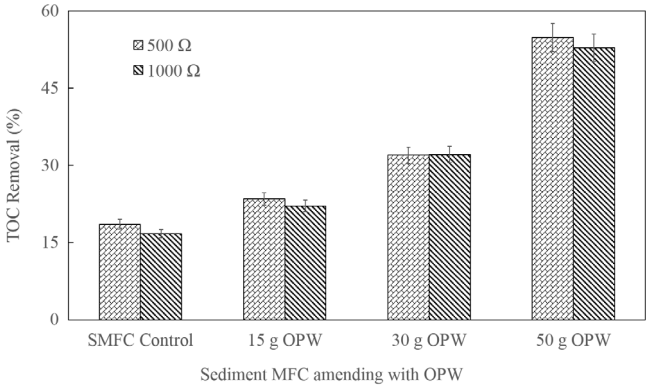

FIGURE 10 Total organic carbon removal (%)

50 g OPW presented TOC removal efficiencies of 23.44%, 31.94%, and 54.8% respectively. Our results are comparable to the ones obtained by Gonzalez-gamboa et al.[32] who observed promising performance of SMFC for TOC removal efficiency after 30, 60, 90, and 120 days of operation from a various sediments enrichment of organic matter.

The highest organics matter removal and the highest power generation were both observed in the SPMFC with the highest OPW content. Unsurprisingly, the control SPMFC which was not inoculated and in which no OPW was added showed minimum removal efficiency and lowest power generation (Table 3). It is known that the growth of microorganisms is proportional to the utilization of substrates. This experiment showed that the increase of microbial biomass in SPMFC compared to the control SPMFC increased the growth of EAB in the total biomass. The sewage sludge serves mostly as an inoculum or microorganism source but it could also account for an additional source of organic matter that can be used as substrate at the anode.[36]

The ROOM in sediments mostly decreased after SPMFC operation due to the degradation of organic matters by microbes.[37] The higher decrease of organic matter in the sediment was achieved by larger specific electrode surface areas for the growth of EAB. Consequently, a higher current generation reflected a larger decrease of organic matter in the sediment.[38] The ability of EAB to oxidize a substrate and subsequently transfer electrons to the anode is a possible reason for current generation in SMFCs.[38] Zhu et al.,[39] demonstrated that the cellulose-fermenting microorganisms in anaerobic environments degrade cellulose, hence, OPW could have been utilized by the microorganisms and was converted to electricity. The end products of fermentation (such as acetic, propionic, butyric, isobutyric, valeric, isovaleric, and lactic acids with acetate as the major end-product) can be used by the exoelectrogenic bacteria on the anode of MFC to produce current.[40]

Yang et al.[41] tested the long-term applicability of SMFCs in the bioremediation of toxic-contaminated sediments and obtained a TOC degradation efficiency of 22.1% after 2 years. Xu et al.,[21] obtained a higher TOC degradation efficiency of about 57% within a shorter period of 60 days.

TABLE 3 The role of dewatered sludge to enhance organic matter removal and power generation

SPMFCs	Dewatered sludge Inoculum	LOI removal (%)	ROOM removal (%)	COT removal (%)	Power density (W m^{-3})	Current density (A m^{-3})
Control SPMFC	Non-inoculated	13–25	11.5–12	16–19	0.01–0.2	0.5–1.5
SPMFC	Inoculated	14–35	19–40	22–55	1–1.3	10–11

Abbreviations: LOI, loss on ignition; ROOM, readily oxidizable organic matter; SPMFC, solid phase microbial fuel cell.

TABLE 4 Scale up of SMFC in single mode and series mode

SMFCs	OPW (g)	Volume (L)	Power (mW)	Voltage (V)
Single SMFC	50	0.3	0.33	0.7
3 SMFC	150	0.9	0.45	1.2
6 SMFC	300	1.8	0.54	1.9
9 SMFC	450	2.7	0.72	3.95
12 SMFC	600	3.6	2.08	4.6

Abbreviations: OPW, orange peel waste; SMFC, microbial fuel cell.

3.3 | Stacking of SPMFCs

The scale-up of SPMFCs was carried out by stacking them. Series of 3–12 SPMFCs could generate voltages ranging from 0.7 to 4.5 V and powers from 0.33 to 2.08 mW for total reactional volumes of 0.3 and 3.6 L, respectively (Table 4). These results are in agreement with the ones obtained by Ewing et al.,[28] who recorded a power of 2.33 mW with an electrode surface area of 0.36 m^2. In this experiment, we obtained a power of 2.08 mW with an electrode with a lower surface area of 0.016 m^2.

These results suggest that an upscaling of the process can be carried out by simply adding more SPMFCs making the process particularly convenient.

4 | CONCLUSION

SPMFCs can be used to treat solid organic wastes produced by the citrus industry while producing energy at the same time. It was shown that power outputs were interesting and were dependent on substrate concentration or organic load as well as the resistance of the external load. A maximum of 0.28 W m^{-2} was obtained for an initial OPW content of 50 g and an external resistance of 500 Ω. Under these conditions, LOI, ROOM, and TOC removals of 35%, 39%, and 55%, respectively, were recorded.

It was also noted that designing the SPMFC as we did allowed for a reduction of the internal resistance which clearly contributed to the improved performances that were recorded.

In conclusion, this study showed that it is possible to use SPMFC to decompose OPW while simultaneously producing electricity .

The SPMFC can improve environmental quality due to the reduction of agro-industrial waste pollutants; contribute to a better urban development and providing renewable energy. Also, the results obtained after scaling-up suggest that this process may be integrated into a solid waste treatment plan in the citrus industry to treat the wastes and to produce renewable energy in situ.

DATA AVAILABILITY STATEMENT

Research data are not shared.

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