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Bioremediation using microbial fuel cell for polluted marine sediment

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ABSTRACT. The pollutant from Karangsong port activities in Indramayu, West Java province, presumably has been affecting its coastal environment, which is reflected in the degradation of water quality and toxic accumulation in the sediment. Recovery of polluted area can be initiated using bioremediation agent such as bacteria. The aim of this study was to investigate indigenous bacteria from various sediment in Karangsong as bioremediation agent using Microbial Fuel Cell method to reduce pollutant and to investigate micro-energy thereafter. Sample of water and sediment from estuary, fishpond, and mangrove area had been collected and used as media for the MFC unit. In a 35-day experimental period, with 14 days of the peak, all MFC units were able to reduce the pollutant with best performance showed by the unit from mangrove area. The reduction of ammonia, nitrate, phosphate, Pb, and Total Petroleum Hydrocarbon showed 170 mg/kg, 4207 mg/kg, 1200 mg/kg, 12.19 ppm, and 58900 mg/kg, each respectively. Of all tested electricity parameters, the MFC unit taken from mangrove area showed better performance in comparison to the others. The voltage, current density, and power density showed as high as 327 mV, 211 mA.m^{-2}, and 69 W.m^{-2}, respectively, while other MFC units showed lower values. Further study on bacteria optimisation should be encouraged in order to reach the shorter duration of pollutant reduction and higher micro-energy result at the same time.

Keywords: bioremediation; mangrove restoration; microbial fuel cell; pollutant reduction; sediment surface

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INTRODUCTION

Eutrophication is a natural process that occurs in various fields, yet it can be dangerous if the process happens continuously in high intensity (Morand and Merceron, 2005), and followed by pollutant input from anthropogenic activity. The pollutant in eutrophic condition that is usually toxic comes from natural process (10%), industrial activity (7%), detergent (11%), fertiliser (17%), human litter (23%), and livestock (32%) (Morse *et al*., 1993). This proof showed that anthropogenic activities are severely polluting various environmental areas, including marine sediment. On the other hand, marine sediment potentially produces new chemical substances from biological activities (Mead *et al*., 2005). Sea sediment consists of organic compound with its dry weight ranging from 0.5% to 20% (Hong *et al*., 2010).

By looking at the broad impact of eutrophication and pollution on aquatic sediments, as well as the high content of organic matter in sediments, effort is required to reduce eutrophication by utilising the pollutants in natural mechanism that occur in the process of degradation triggered by microbes.

One of the methods that facilitates these needs is Microbial Fuel Cell (MFC) as it utilises a wide range of soluble and dissolved organic compounds present in wastewater and renewable biomass (Boghani *et al*., 2017). Other studies are dealing with wastewater from industrial process (Feng *et al*., 2008; Lu *et al*., 2009), wetland construction (Wang *et al*., 2016), fishery waste (Li *et al*., 2017), phytoplankton (Ma *et al*., 2017), constructed wetland (Lu *et al*., 2015), and sediment of eutrophic reservoir (Pribadi *et al*., 2019). However, only little information about MFC from marine sediment can be referred to, which utilises organic material including pollutants and also the reduction of the pollutant itself.

Indramayu is a district in northern West Java that becomes one of the most productive mariculture areas in Indonesia. The district of Indramayu is also located quite close to port, which potentially pollutes the surrounding areas, including the sediment in the coastal parts. Therefore, the marine sediment of Indramayu has a unique characteristic due to the rich organic compound from mariculture and pollutants resulting from ship traffic in the port. Considering this condition, it is necessary to find solution to reduce the pollutant and excessive organic compounds. MFC has been applied and therefore is considered as an alternative method to solve the problem. By using this method, not only the decrease of the pollutant is being reached, but micro-energy can also be developed as a potential by-product.

This study is aimed to observe the prospect of MFC method application in reducing the pollutant level in the sediment, and at the same time producing micro-electricity. The target is to make use of polluted material and turn them into something useful.

MATERIALS AND METHODS

Study area. Samples of sediment were taken from three locations of the study site due to different sediment characteristic: shrimp ponds (6°18’03.7”S; 108°22’02.0”E), estuary (6°18’17.9”S; 108°22’05.3”E), and mangrove restoration area (6°18’11.2”S; 108°22’06.6”E) (Fig. 1) in Indramayu, West Java, Indonesia.

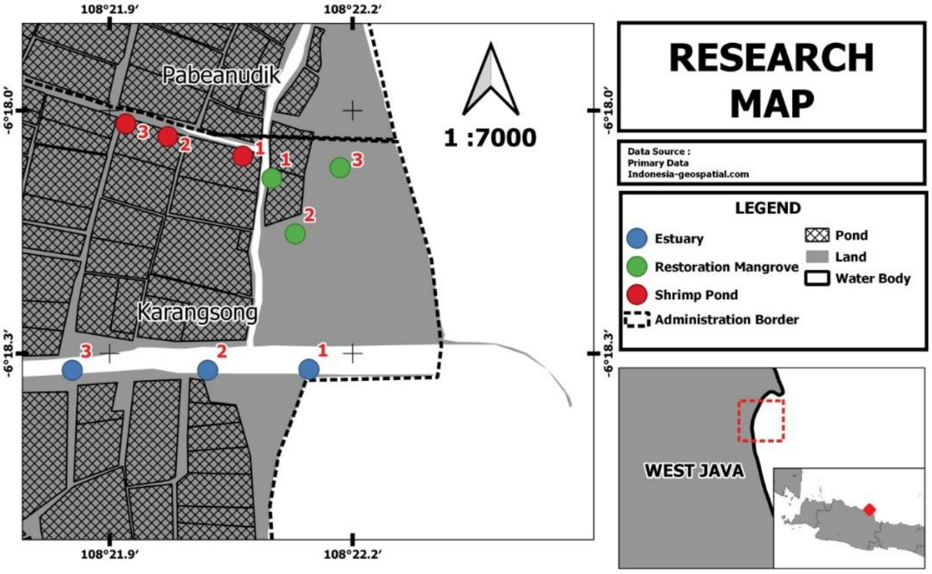


Fig. 1. Location of sediment sample of (red) shrimp ponds, (blue) estuary, and (green) mangrove restoration area in Karangsong, Indramayu, West Java, Indonesia

Water and sediment collection. The media used in this study were water and sediments from three sampling locations (Fig 1). Sediments were taken using Eickman grab as much as 5 litres of sediment in volume from each location with depth of approximately 10 cm below surface, and then stored in sterile plastic jar with airtight lid. Water was taken from the water column at the same location with sediment (Holmes *et al*., 2004). Water samples were taken in amount of 40 litres, which was collected in sterile jerry cans. Water and sediment samples were stored in a cool box during the trip, and kept in a refrigerator with the temperature of below 10°C to slow down the metabolism of microorganisms and avoid any unexpected chemical reactions prior to the experiment.

Microbial fuel cell experiment. The MFC unit used in this study was a closed-circuit single-chamber type, as referred to Holmes *et al*. (2004), Logan *et al*., (2006), and Pribadi *et al*., (2019), where the sediment samples were inserted into a vessel of 3 cm high. An electrode made of carbon (anode) is placed in the sediment and covered with the sediment for 2 cm high. The vessel is then filled with 400 ml of water sample and stored in room temperature for 24 hours to precipitate particles of the sediment. On the following day, a carbon electrode (cathode) was placed 1 cm above the sediment surface and kept emerge in the water column. MFC operated in dark condition (without lighting), without aeration, and at room temperature (27°C ± 1). Water loss due to evaporation during the measurement of electric current is replaced by sterilised distilled water demineralisation. The unit of sediment and water without MFC circuit were used as control. The experiments used three replicates for each source of the sediment.

Pollutant analyses. Pollutant analyses were done initially and finally after experimental period of 35-day MFC treatment for ammonium, nitrate, phosphate, Pb, and Total Petroleum Hydrocarbon (TPH). Pollutant analyses were measured as pollutant concentration decreased and removal efficiency. Analysis for N-total, nitrate, NH_4, phosphate, Pb, and TPH are referred to Indonesian National Standard for Seawater Quality (Agency for Standardization of Environmental and Forestry Instrument, 2003). Samples of 1 g dry weight sediment of each measurement were extracted and put into 200 ml Erlenmeyer flask. Samples were measured in spectrophotometer (Thermoscientific, Genesys 60, USA) for absorbance at a certain wavelength.

Electricity measurements. The electrical measurement parameters were expressed in voltage (mV), current density (mA. m^{-2}), and power density (W. m^{-2}). The electrical measurement had been taken once in a day in the same time frame, during the 35-day experimental period. The measurement was done by digital multi-function tester (ATN YX- 360TR$_{E-B}$,).

RESULTS AND DISCUSSION

Pollutant decrease. The content of initial pollutants in the sediment exceeded the quality standard of national content of pollutants in sediment and seawater as ruled in the Indonesian Government Regulation No.22/2021 for seawater quality standard. This proof showed that the study area has been highly polluted and eutrophicated. Almost all initial pollutants showed extremely high values for mangrove sediments, except in Pb with initial pollutant content of TPH from estuary sediment being the highest in comparison to sediments of pond and mangrove (Table 1).

Table 1. Initial pollutant content (N-Total, P-Total, ammonium, nitrate, Pb, and TPH) in the sediment of ponds, estuary, and mangrove, and Indonesian regulation on quality standard

Sediment	N-Total (mg.kg^{-1})	P-Total (mg.kg^{-1})	NH_4 (mg.kg^{-1})	NO_3 (mg.kg^{-1})	Pb (ppm)	TPH (mg.kg^{-1})
Ponds	1800	1800	270	530	20.83	80,300
Estuary	1800	1700	270	544	20.87	85,100
Mangrove	2000	2000	320	520	18.35	84,900
Quality standard	20 (mg.l^{-1})$^{*)}$	20 (mg.l^{-1})$^{*)}$	5 (mg.l^{-1})$^{*)}$	30 (mg.l^{-1})$^{*)}$	0.1$^{*)}$	10,000 µg.g$^{-1\,*)}$

Note: Indonesian Government Regulation No.22/2021 for seawater quality standard

In general, the pollutant contents detected in the sediment were reduced significantly due to the control with the MFC method. Samples with no MFC treatment also showed pollutant content decrease, though with a smaller range in comparison to the units with MFC (Fig. 2). The largest decrease of all parameters being measured in the MFC units was shown by MFC units of mangrove sediment, while the other two groups were slightly fluctuating (Fig. 2).

Removal efficiency of all pollutants being measured for MFC units of mangrove sediment showed greater than 60%, except for NH_4, 53% (Fig. 3). The MFC units of estuary sediment have low performance in removal efficiency compared to the other groups of ponds and mangrove sediment, with some of the values below 50%. The various result performances in pollutant decrease and electricity production of MFC units in this study were presumably due to the different bacteria consortia living in different sources of sediments. Several researches on mixed bacteria consortia in MFC units reported that considerable diversity in bacteria communities were able to be sustained in MFC systems, thus showing the stability and performance in terms of degradation of pollutant and electricity generation (Ki *et al*., 2008; Kim *et al*., 2006; Logan and Regan, 2006).

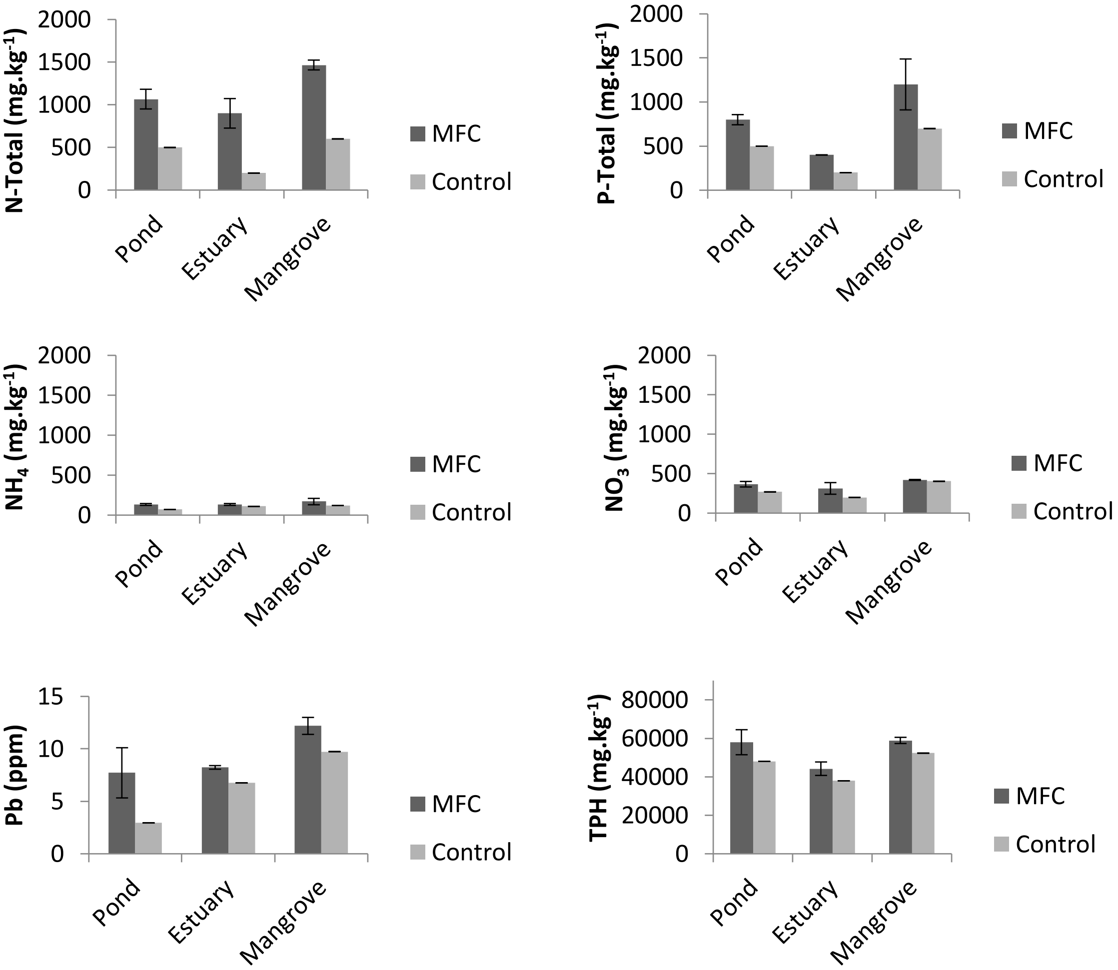
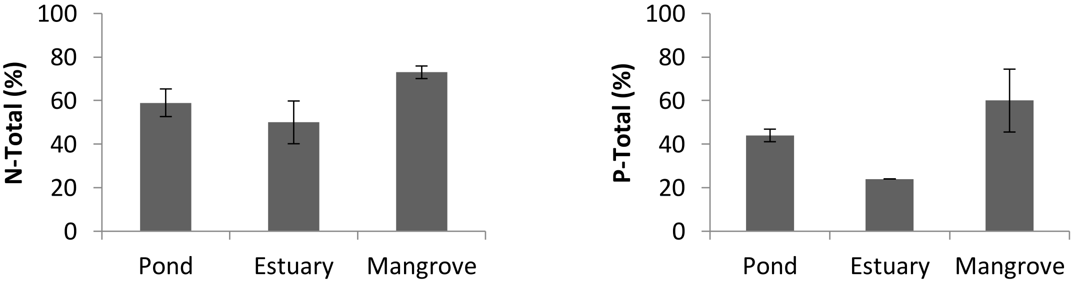


Fig. 2. Decrease of pollutants from sediments of ponds, estuary, and mangrove, after using MFC unit for 35 days of experimental period. (± SD)

The higher performance of pollutant decreases in MFC units using mangrove sediment can be related to the higher nutrient content (total-N and total-P) in comparison to the other groups of ponds and estuary sediments. The higher nutrient content of mangrove sediment supported more than ponds and estuary sediments to the metabolisms of microorganism, so that they can degrade faster with bigger capacity.



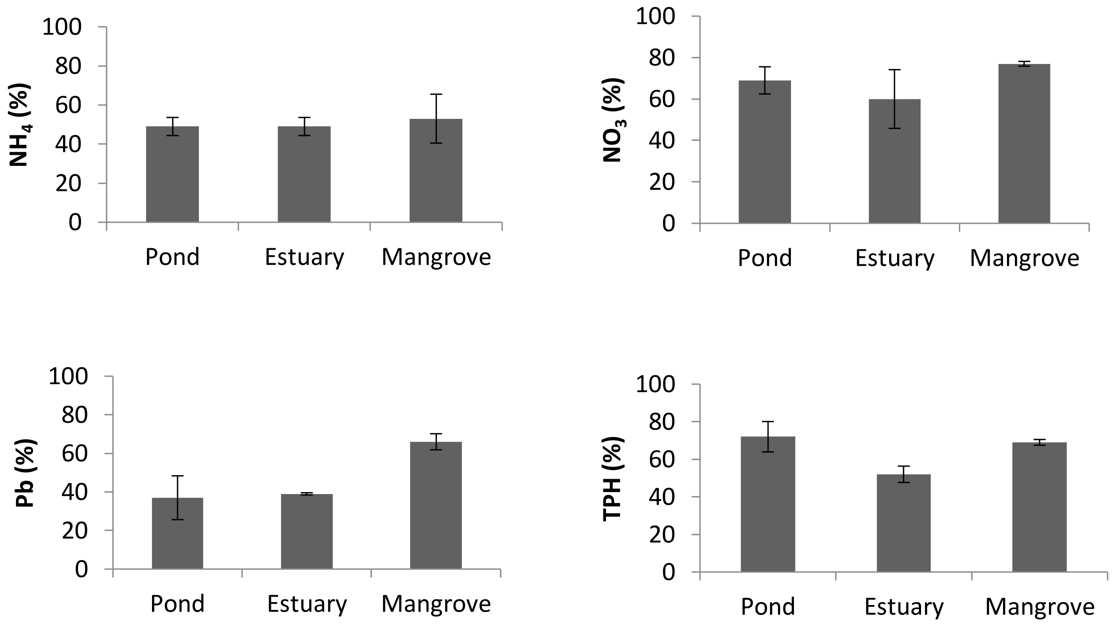


Fig. 3. Removal efficiency (%) of pollutants from sediments of ponds, estuary, and mangrove, after using MFC unit for 35 days of experimental period. (± SD)

Electricity production. After 35 days of experiment, results showed that MFC units generated different electricity productions for the groups of sediments from ponds, estuary, and mangrove. The MFC units with mangrove sediment showed higher electricity production in comparison to the MFC units with sediments from ponds and estuary (Fig. 4). When microorganism began to perform metabolism at the anode surface, high-energy molecules accumulated around the anode. Electrons were then accumulated around the anode while the cathode became more oxic. This condition has not only resulted a potential difference between the anode and the cathode, but also triggered an electric current (Holmes *et al*., 2004). The more effective performance of electricity production with mangrove sediment was presumably due to the higher nutrients of nitrogen and phosphate in comparison to the other units of ponds and estuary sediments. The high nutrient in mangrove area also reported in the study of nutrient profile in the same region (Frederika *et al*., 2021). The high nutrient in mangrove presumably come from tidal activities of the seawater, which then formed a tidal-borne sediment as being reported in other studies (Kamal *et al*., 2020). Higher nutrient content means higher micro-organic metabolisms, thus higher energy accumulation.

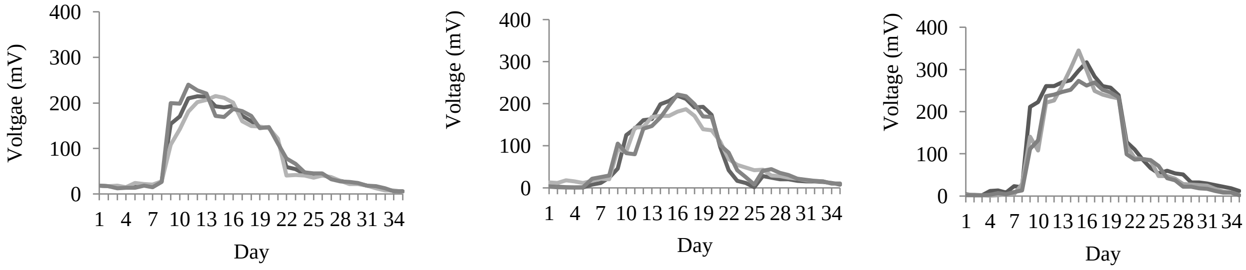


Fig. 4. Production of voltage (mV) from MFC units using sediment from ponds, estuary, and mangrove for a 35-day experimental period

All MFC units showed slight fluctuating pattern during experimental period. The significant electricity production started approximately on day 8, and it lasted for ±12 days. The pattern started to decline after day 22, and tended to decrease continually until the end of experimental period. The decline was presumably caused by the lack of nutrients for the bacteria to grow in the closed MFC units, as reported by other MFC researches (Gezginci and Uysal, 2016; Park *et al.*, 2008).

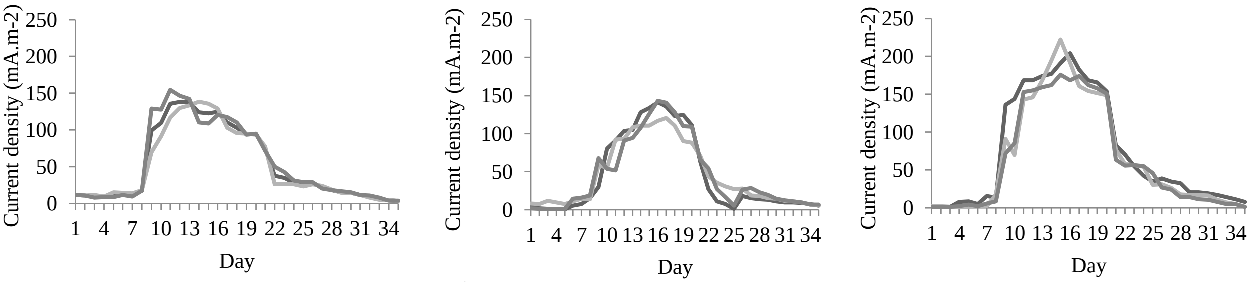


Fig. 5. Production of current density (mA.m^{-2}) from MFC units using sediments from ponds, estuary, and mangrove for a 35-day experimental period

All MFC units showed lag phase of approximately 5 days at the beginning of experimental period before they started to produce electricity. This lag phase was due to the adaptation phase of the bacteria to their new environment, especially to temperature, not to mention that the sediment had also been chilled during storage before experiments. The peak of the electricity production of MFC units from estuary and mangrove was reached at the same range between day 16 and 17, as shown by current density and power density (Fig. 5 and Fig. 6). The MFC units of pond showed peak electricity production earlier than the other two groups, somewhere between day 10 and 11 (Fig. 5 and Fig. 6). The peak electricity reached in this study were longer than the result from other study (Syafitri *et al.*, 2018), which presumably due to the additional molasses used in their study.

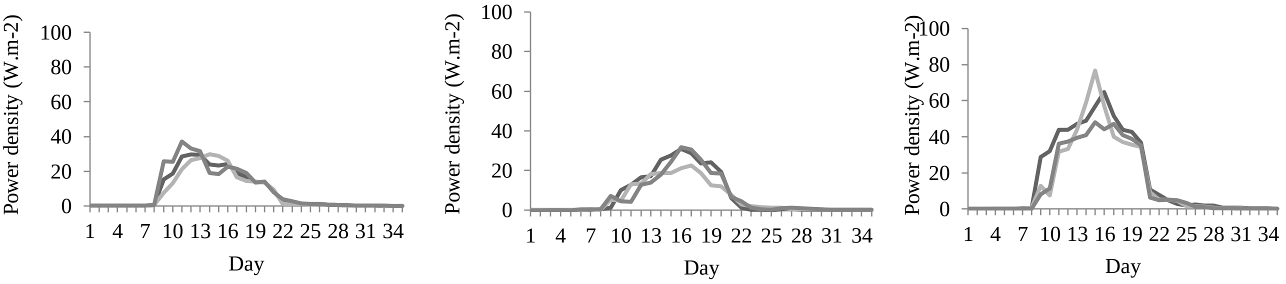


Fig. 6. Production of power density (W.m^{-2}) from MFC units using sediments from ponds, estuary, and mangrove for 35 days of experimental period

Sources of sediments that were used as substrate can also be the cause for low electricity production. Fresh water considered as having lower electrical conductivity than seawater would not be able to generate maximum level of electricity. MFC with marine sediments as a substrate experienced peak production within 20 days, while the MFC with freshwater sediments produced its electricity peak faster within 5 days (Holmes *et al.*, 2004). Voltage production showed minor fluctuating decrease during the 35-day experimental periods in all treatments, which consecutively had an impact on current density and power density (Fig. 4, Fig. 5 and Fig. 6). No additional oxygen in the sediments caused the cathode potential near 0, while the potential at the anode was negative.

CONCLUSION

Pollutant and excessive nutrients in the sediments in Karangsong area can be reduced by the process of degradation with the help of indigenous bacteria contained in the sediments, and significantly accelerated by MFC method. This method with a series of single-chamber type has definitely proven to accelerate the degradation process while generating electricity at the same time, that can be used as a source of alternative micro-energy. MFC units of mangrove sediment performs the best results in both pollutant reduction and electricity production, indicating that the method is very much potential and suitable to be applied in the field of bioremediation in terms of marine polluted sediment. Optimisation in degrading bacteria by engineering the environment with oxygen and other nutrients addition is a decisive and crucial factor for the continuation of this research. Likewise, the stability of the circuit in generating electricity.

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