

BIOELECTRICITY GENERATION AND REMEDIATION OF CONTAMINATED INTERTIDAL ZONE OF YAMAGUCHI BAY, JAPAN

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Abstract

In this study, to generate bioelectricity to power the monitoring sensors and to improve sediment simultaneously is discussed in Yamaguchi bay located in the south part of the Honshu island, Japan through sediment microbial fuel cell (SMFC). To supply energy to the geoenvironmental monitoring sensors is a serious problem, as traditional batteries and solar cells are often inconvenient due to the limitations of recharging and weather conditions, respectively. Four different sediments were used in the laboratory to generate bioelectricity and to improve sulfide contamination simultaneously by SMFCs. The acid volatile sulfide (AVS) was determined to check the improvement of the geo-environment of the sediments as it was the main source of contamination in the intertidal zone (tidal flat). Various factors which affected the voltage generation have been studied. Voltage values showed almost twice when two anodes were used instead of a single anode. Higher temperature showed the higher voltage due to increased activity of the bacteria in the higher temperature. AVS values reduced quickly when the closed circuit was used. All the marine sediments showed the decreasing trend of AVS value with time and reached at the 0.2 mg/g-dry mud indicating the geoenvironmental improvement of the marine sediments within 14 days. Field test showed the higher value of voltage comparing to the laboratory test by using the similar sized SMFCs.

Bioelectricity generation and remediation of contaminated intertidal zone of Yamaguchi Bay, Japan

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Keywords: AVS, bioelectricity, geoenvironment, marine sediment, SMFC

1. INTRODUCTION

We need green energy which could be a sustainable source for the power supply to the environmental monitoring sensors such as temperature sensors in the remote intertidal zone (tidal flats) in Japan and other places. Depletion of energy reserves, global warming and the concern of environmental pollution are inspiring the search for new environment friendly and sustainable energy production methods all over the world. Renewable bio-energy is viewed as one of the ways to alleviate fuel needs of the future and to overcome the crisis of global warming.

Microbial Fuel Cell (MFC) is the bio-electrochemical device that converts microbial reducing power into electrical energy which is a green and safe source of bioelectricity (Logan & Regan, 2006; Hong et al., 2009; Moqsud 2020). They use the existing substrates from renewable sources and convert them into harmless by-products with simultaneous production of bioelectricity (Habermann W. & Pommer, E.H., 1991). In this way, bioelectricity production employing MFC has generated considerable interest in both basic and applied research in recent years. Moqsud et al., (2014) tried to get bioelectricity from various organic wastes and also living plants by using MFC technology.

SMFCs are being considered for use as a potential power source for aquatic water quality sensors such as pH, temperature, and dissolved oxygen sensors (Moqsud 2020; Moqsud et al., 2017). SMFC can be a potential source of remediation of this problem. The geo-bacteria (which live in soil) break the organics in the soil and consequently generate the bioelectricity. Abbas et al., (2017) found that SMFCs could be a source of sustainable energy and heavy metal remediation. To change the batteries from the environmental monitoring sensors of this contaminated tidal flat is a serious problem and often the solar system is not working due to the bad weather or other reasons.

The major objective of this study is to generate bioelectricity from a contaminated sediment while improving its geoenvironmental condition in the laboratory experiment and to compare it through field experiment of SMFCs.

2. METHODS AND MATERIALS

2.1 SAMPLE COLLECTION

Four samples were collected for this experiment from Yamaguchi bay at depths of 0-20 cm from tidal flat and the river areas during the ebb tide. A ponar dredge sediment sampler was used to collect the samples. Sample 1, 2 and 3 were collected near the intertidal zone of Yamaguchi bay (33.93 N, 131.22 E), Ube city. Sample 4 was collected from the river near that tidal flat area (33.93 N, 131.22 E) to compare the marine sediments SMFCs and the river sediment SMFCs. The samples were collected middle of May, the average ambient temperature is around 25°C. The geoenvironmental condition of the Yamaguchi bay tidal flat area is not good due to the various human activities. The smell of the sediment was unpleasant and there is very small signs of benthos in the sediment. The AVS value is more than suitable range in different parts of the Yamaguchi bay and hence caused the declination of catch of shell and decrease of the horseshoe crab has been observed (Moqsud et al., 2017). This is a serious ecological problem and the researchers are trying to solve this problem.

Table 1 summerises the basic physico-chemical properties of the four sediments. From the grain size analysis of the four sediments, it was observed that all the three sea sediments showed the similar trend, however, the river sediment showed a different trend and the particle size is larger than the sea sediments. It was seen that the loss on ignition (LOI) which is the indicator of organic content was relatively lower in sea sediments (sample 1, 2, 3) 12% than in the river sediment 14% (sample 4). The AVS value was also higher in the case of marine sediments (more than safe limit , 0.2 mg/g-dry mud). Anaerobic bacteria (sulfate reducing bacteria) reliably to generate AVS by converting the sulfate to sulfide and is one of the major indicators for the geoenvironmental condition (Rickard & Morse, 2005). In this study, the AVS value will be taken as a key indicator of the geo-environmental condition improvement. According to the Japanese fisheries research association, the safe value for AVS is 0.2 mg/g dry mud for the benthos living in the mud.

2.2 Laboratory Experiments

Figure 1 illustrates the schematic diagram of SMFC in the laboratory experiment. One liter glass beaker was used in the laboratory as a single chamber SMFC without a separator. The sediment sample was placed in the cell. Same size (100 cm²) of carbon fiber (Toray Corp. Tokyo, Japan) was used for both the anode and cathode materials (Moqsud 2020; Moqsud et al., 2014). The anode was placed inside the sediment and the cathode was placed on the top of the surface (Moqsud et al., 2013; Omine, 2009). Both of the electrodes were connected to a fixed external resistance (100 Ω) and measured the data by using a multimeter (UNI LCD Multimeter, USA). The external resistance was fixed at 100 by assessing the polarization curve with different resistors and also by comparing open circuit voltage (OCV) (Moqsud et al., 2017). The polarization curve was drawn by following other literature (Logan nad Regan 2006). The laboratory investigation was conducted at a constant room temperature of 25°C. AVS was measured by following the GASTEC method.

2.3 Field Test

The field test was carried out in Ube city Yamaguchi bay tidal flat area (33.93 N, 131.22 E) (near the Sample 3) from where the sediment samples were collected for the laboratory experiments and it was considered that the basic physicochemical properties of the marin sediments were similar both in the laboratory and field experiments. The similar size of laboratory SMFC was set up in the tidal flat area in where the water inundates during the high tide and exposed to atmosphere during the low tide. The cathode was set up on the surface of the tidal flat sediment and the anode was set up inside the sediment (5 cm below). The same carbon fiber (Toray company, Japan) was used with the same surface area 100 cm² which was used during the laboratory experiment. The voltage was measured in the field and the AVS was calculated in each week by following the same method of the laboratory experiment. To check the AVS variation both from laboratory and field test, the samples were collected from the anode area.

Statistical analysis

For statistical significant testing, p (probability value) less than or equal to 0.05 was used.

4. RESULTS AND DISCUSSIONS

4.1 Variation of current with time

Figure 2 illustrates that the variation of voltage with duration in the laboratory experiment. It is seen that all the three marine sediments showed the higher value of voltage than the river sediments. Sample 3 showed the peak voltage and reached at around 300mV after 1 week. Sample 1 and sample 2 showed the similar trend of voltage generation with sample 3. The initial rate of voltage generation was higher as the bacteria got ample of food during the early stage of laboratory experiments. After reaching at the peak the voltage generation reduced and almost constant for another 2 weeks. However, voltage generation was always lower in the case of river sediment. Though the organic content of the river sediment was little bit higher than the marine sediment, the grain size of river sediment was larger and consequently affect the voltage generation. The peak voltage generation in river sediment was 140 mV which was 60% lower than the sea sediments. The high electrical conductive saline water of sea could be another reason for the higher value of voltage in the sea sediment (Song and Jiang, 2018). The external resistance used in this particular research was 100 both in the lab and the field. So, the peak power output from one cell is around 28 mW/m². Wang et al., 2014 showed that the voltage generation can be great influenced by the organic content present in the sediment. However, our results showed that the power generation was always higher in the sea sediment instead of river sediment. The particle size and the salinity affected the results more significantly than the organic content of the sediment.

4.2 Variation of anode number and generation of voltage

Figure 3 illustrates the effects of the anode number in the SMFC in the laboratory experiments. It was observed that the amount of voltage increased twice when the anode number was twice. The peak reached up to 250 mV when double anode (2 anodes) were used. The electron acceptor areas were increased by increasing the number of the anode, as a result, the peak value was also showed higher when double anode was used instead of a single anode. The number of anode has an influence on generation of bacteria-induced electricity which was revealed by this experiment. The bacteria also closely attached with the anode materials to transfer the electrons from the sediment by making biofilm on the surface of the anode materials. Again, the surface area of the anodes had a greater influence on the bioelectricity generation. Moqsud 2020 showed the variation of voltage generation was affected by the anode materials such as stainless steel, graphite materials or bamboo charcoals. However, he did not show how much power generation has been affected by the number of anode.

4.3 Effects of temperature on voltage generation

Figure 4 illustrates the effects of temperature on the voltage generation in the laboratory experiment. It was seen that when the temperature was 25⁰ C then the amount of voltage was also higher compared to 5⁰ C. The average winter and spring temperature in the Yamaguchi bay is around 5⁰ C and 25⁰ C, respectively and for this reason we fixed those temperature during the laboratory experiment. The bacterial activities increased in the increased temperature and consequently increased the voltage generation. At the 25⁰ C, the peak voltage reached around 85 mV and at the 5⁰ C the peak voltage reached around 48 mV. This proved that the temperature is one of the biggest affecting factors for the voltage output in the SMFCs. Lovley 2006 showed that the temperature is also a affecting factor for the waste water MFCs. The possible shortcomings of this particular research could be the effect of temperature in the field. The lower winter temperature could affect the power generation and consequently it could be affect the power supply to the sensors in the field. To minimize this problem, SMFCs could be developed and connec in a series connection and set in the tidal flat area.

4.4 Variation of AVS (AVS) in different kinds of sediment

Figure 5 illustrates that the AVS value varied with time in all types of sediment. The river sediment showed a lower value of AVS is probably due to the size of the sediment during the starting of the experiment. The larger particle size of the river sediment can be one of the reasons why the AVS is lower value. The another probable reason of the lower AVS in river sediment is due to the pollution level of the river water. The various human activities such as using the chemical products to grow the sea weed in the sea is one of the main reasons of the higher AVS in the sea sediment. However, AVS reduced for all the samples from the

1st week of the experiment and after 3 weeks, SMFC 1, 2 and 3 showed a dramatic reduction of the sulfide content. Nevertheless, all of the 4 samples showed that the sulfide content reduction trend after using the SMFCs compare to blank sediment (without using the SMFC). The probable reason of AVS reduction in the SMFC is that the electron movement is frequent in the SMFC and this condition is favorable for the geo-bacteria who work in the bioelectricity generation from the polluted sediments which was discussed in the earlier research (Moqsud 2020).

4.5 Variation of AVS with time in different circuit conditions

Figure 6 demonstrates the variation of AVS with time for the SMFC system in different circuit conditions. Three samples were prepared with different circuit conditions but the same amount of sediment to compare the effect of the circuit for AVS reduction. One MFC was designed with an open circuit, another one is used for 100 external resistance and another one is without electrode or circuit (blank) condition.

It was seen that the AVS reduction was faster when SMFC was used. The open circuit and the circuit with 100 were more effective to reduce the AVS than the sample without the circuit system. This figure confirms that by using the SMFC system, the AVS reduction will be faster than without using MFC for the sediment contaminated with higher AVS. This type of phenomena is mainly due to the enhanced flow of electron inside the MFCs (Rabaet et al. 2006; Zhang and Zhou 2009). The geo-environmental condition of the high AVS contaminated sediment can be improved by applying this method. The benefit of this method is that it could be used as a safe source of bioelectricity generation in the remote places and this electricity can be used for small applications such as to power the environmental monitoring sensors.

4.6 Variation of voltage generation and AVS reduction in the field

The field test in the marine sediment in the Yamaguchi bay was carried out in a similar size of the laboratory SMFCs. It was interesting that the AVS reduction trend was similar in the field, however, the voltage generation is not similar with the laboratory experiments. The voltage generation was higher in the field than the laboratory experiments for the similar size SMFCs. The probable reason of this trend is that the various nutrients supply in the intertidal region naturally and that might increase the activities of the bacteria and hence increase the voltage. The tidal effect and the other environmental factors such as solar radiation could also affect the power generation in the field. This was the first time to compare the voltage generation between the laboratory experiment and field experiment with the similar SMFC in the high sulfide content contaminated sediment. Kubota et al. 2019 showed that AVS value has reduced in the Tokyo bay tidal flat sediment during a field test in the high eutrophic water enclosed bay.

By utilizing the SMFC, the rate of flow of electrons can be probably increased and the process of AVS (mainly FeS and H₂S) reduction will be increased. The resulting electron flows promote remediation of sediment by enhancing physicochemical and microbial metabolic reactions. So the voltage generation and the AVS reduction were happened together in the field experiment (Erable et al., 2017). Though the voltage and power generation is higher in the field but the seasonal temperature variations could be a factor in the field to supply power to the environmental monitoring sensors. This present field test was carried out in relatively warm (ambient temperature varied from 20-25^o C). The field test during the winter might show some lower power generation due to the lower surrounding temperature. In this particular research, the similar size single anode SMFC was used both in the laboratory and the field experiments. The series connection of the SMFCs could increase the voltage and power generation and hence could be a potential source of power supply to the various geo-environmental monitoring sensors. The mechanisms of the AVS reduction was discussed in details in our previous paper (Moqsud 2020).

5. CONCLUSIONS

The bioelectricity generated by the SMFC can be used to supply the power to the environmental monitoring sensors. This power source can be a sustainable and green source of electricity. The performance of the system with two anodes was about twice higher than that of with one anode regarding bioelectricity generation. The variation of AVS value was observed and decreased about 0.2 mg/g-dry mud indicated the improvement

of the geoenvironment for the marine sediments after using SMFCs in the contaminated sediments both in the laboratory and in the field . By observing the different circuit conditions, it was found that the rate of AVS decreasing was the fastest and slowest when a close circuit and no circuit (blank) condition was set, respectively. It showed the similar trend as Abbas et al. 2019 have been observed. Higher temperature is better for enhancement of voltage generation because the bacterial activities enhanced with the enhancement of the temperature. It was observed that voltage at 25⁰ C was almost 8 times higher than that of at 5⁰ C. From the laboratory test and the field test result comparison, it was found that voltage generation is more durable in the tidal flat areas however, during the winter, the voltage generation might be decreased due to colder surrounding temperature in Japan. So, proposed SMFC system can both generate bioelectricity and improve the geoenvironmental condition of the marine sediment at the same time and extra measure should be taken at the winter season for sustainable power supply from SMFC. Economic analysis and the feasibility of the large scale SMFCs in the field should be carried out in the future.

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Statistical analysis

For statistical significant testing, p (probability value) less than or equal to 0.05 was used.

4. RESULTS AND DISCUSSIONS

4.1 Variation of current with time

Figure 2 illustrates that the variation of voltage with duration in the laboratory experiment. It is seen that all the three marine sediments showed the higher value of voltage than the river sediments. Sample 3 showed the peak voltage and reached at around 300mV after 1 week. Sample 1 and sample 2 showed the similar trend of voltage generation with sample 3. The initial rate of voltage generation was higher as the bacteria got ample of food during the early stage of laboratory experiments. After reaching at the peak the voltage generation reduced and almost constant for another 2 weeks. However, voltage generation was always lower in the case of river sediment. Though the organic content of the river sediment was little bit higher than the marine sediment, the grain size of river sediment was larger and consequently affect the voltage generation. The peak voltage generation in river sediment was 140 mV which was 60% lower than the sea sediments. The high electrical conductive saline water of sea could be another reason for the higher value of voltage in the sea sediment (Song and Jiang,

2018). The external resistance used in this particular research was $100\ \Omega$ both in the lab and the field. So, the peak power output from one cell is around $28\ \text{mW/m}^2$. Wang et al., 2014 showed that the voltage generation can be greatly influenced by the organic content present in the sediment. However, our results showed that the power generation was always higher in the sea sediment instead of river sediment. The particle size and the salinity affected the results more significantly than the organic content of the sediment.

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biofilm on the surface of the anode materials. Again, the surface area of the anodes had a greater influence on the bioelectricity generation. Moqsud 2020 showed the variation of voltage generation was affected by the anode materials such as stainless steel, graphite materials or bamboo charcoals. However, he did not show how much power generation has been affected by the number of anode.

4.3 Effects of temperature on voltage generation

Figure 4 illustrates the effects of temperature on the voltage generation in the laboratory experiment. It was seen that when the temperature was 25⁰ C then the amount of voltage was also higher compared to 5⁰ C. The average winter and spring temperature in the Yamaguchi bay is around 5⁰ C and 25⁰ C, respectively and for this reason we fixed those temperature during the laboratory experiment. The bacterial activities increased in the increased temperature and consequently increased the voltage generation. At the 25⁰ C, the peak voltage reached around 85 mV and at the 5⁰ C the peak voltage reached around 48 mV. This proved that the temperature is one of the biggest affecting factors for the voltage output in the SMFCs. Lovley 2006 showed that the temperature is also a affecting factor for the waste water MFCs. The possible shortcomings of this particular research could be the effect of temperature in the field. The lower winter temperature

could affect the power generation and consequently it could be affect the power supply to the sensors in the field. To minimize this problem, SMFCs could be developed and connec in a series connection and set in the tidal flat area.

4.4 Variation of AVS (AVS) in different kinds of sediment

Figure 5 illustrates that the AVS value varied with time in all types of sediment. The river sediment showed a lower value of AVS is probably due to the size of the sediment during the starting of the experiment. The larger particle size of the river sediment can be one of the reasons why the AVS is lower value. The another probable reason of the lower AVS in river sediment is due to the pollution level of the river water. The various human activities such as using the chemical products to grow the sea weed in the sea is one of the main reasons of the higher AVS in the sea sediment. However, AVS reduced for all the samples from the 1st week of the experiment and after 3 weeks, SMFC 1, 2 and 3 showed a dramatic reduction of the sulfide content. Nevertheless, all of the 4 samples showed that the sulfide content reduction trend after using the SMFCs compare to blank sediment (without using the SMFC). The probable reason of AVS reduction in the SMFC is that the electron movement is frequent in the SMFC and this condition is favorable for the geo-bacteria who work in the bioelectricity generation from the

polluted sediments which was discussed in the earlier research (Moqsud 2020).

4.5 Variation of AVS with time in different circuit conditions

Figure 6 demonstrates the variation of AVS with time for the SMFC system in different circuit conditions. Three samples were prepared with different circuit conditions but the same amount of sediment to compare the effect of the circuit for AVS reduction. One MFC was designed with an open circuit, another one is used for 100 Ω external resistance and another one is without electrode or circuit (blank) condition.

It was seen that the AVS reduction was faster when SMFC was used. The open circuit and the circuit with 100 Ω were more effective to reduce the AVS than the sample without the circuit system. This figure confirms that by using the SMFC system, the AVS reduction will be faster than without using MFC for the sediment contaminated with higher AVS. This type of phenomena is mainly due to the enhanced flow of electron inside the MFCs (Rabaet et al. 2006; Zhang and Zhou 2009). The geo-environmental condition of the high AVS contaminated sediment can be improved by applying this method. The benefit of this method is that it could be used as a safe source of bioelectricity generation in the remote places and this electricity can be used for small applications such as to power the environmental monitoring sensors.

4.6 Variation of voltage generation and AVS reduction in the field

The field test in the marine sediment in the Yamaguchi bay was carried out in a similar size of the laboratory SMFCs. It was interesting that the AVS reduction trend was similar in the field, however, the voltage generation is not similar with the laboratory experiments. The voltage generation was higher in the field than the laboratory experiments for the similar size SMFCs. The probable reason of this trend is that the various nutrients supply in the intertidal region naturally and that might increase the activities of the bacteria and hence increase the voltage. The tidal effect and the other environmental factors such as solar radiation could also affect the power generation in the field. This was the first time to compare the voltage generation between the laboratory experiment and field experiment with the similar SMFC in the high sulfide content contaminated sediment. Kubota et al. 2019 showed that AVS value has reduced in the Tokyo bay tidal flat sediment during a field test in the high eutrophic water enclosed bay.

By utilizing the SMFC, the rate of flow of electrons can be probably increased and the process of AVS (mainly FeS and H₂S) reduction will be increased. The resulting electron flows promote remediation of sediment by enhancing physicochemical and microbial metabolic reactions. So the voltage generation and the AVS reduction were

happened together in the field experiment (Erable et al., 2017). Though the voltage and power generation is higher in the field but the seasonal temperature variations could be a factor in the field to supply power to the environmental monitoring sensors. This present field test was carried out in relatively warm (ambient temperature varied from 20-25^o C). The field test during the winter might show some lower power generation due to the lower surrounding temperature. In this particular research, the similar size single anode SMFC was used both in the laboratory and the field experiments. The series connection of the SMFCs could increase the voltage and power generation and hence could be a potential source of power supply to the various geo-environmental monitoring sensors. The mechanisms of the AVS reduction was discussed in details in our previous paper (Moqsud 2020).

5. CONCLUSIONS

The bioelectricity generated by the SMFC can be used to supply the power to the environmental monitoring sensors. This power source can be a sustainable and green source of electricity. The performance of the system with two anodes was about twice higher than that of with one anode regarding bioelectricity generation. The variation of AVS value was observed and decreased about 0.2 mg/g-dry mud indicated the

improvement of the geoenvironment for the marine sediments after using SMFCs in the contaminated sediments both in the laboratory and in the field . By observing the different circuit conditions, it was found that the rate of AVS decreasing was the fastest and slowest when a close circuit and no circuit (blank) condition was set, respectively. It showed the similar trend as Abbas et al. 2019 have been observed. Higher temperature is better for enhancement of voltage generation because the bacterial activities enhanced with the enhancement of the temperature. It was observed that voltage at 25⁰ C was almost 8 times higher than that of at 5⁰ C. From the laboratory test and the field test result comparison, it was found that voltage generation is more durable in the tidal flat areas however, during the winter, the voltage generation might be decreased due to colder surrounding temperature in Japan. So, proposed SMFC system can both generate bioelectricity and improve the geoenvironmental condition of the marine sediment at the same time and extra measure should be taken at the winter season for sustainable power supply from SMFC. Economic analysis and the feasibility of the large scale SMFCs in the field should be carried out in the future.

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