Ammonia removal via microbial fuel cell (MFC) dynamic reactor

I Alabiad, U F M Ali*, I A Zakarya, N Ibrahim, R W Radzi, N Z Zulkurnai and N H Azmi

School of Environmental Engineering, Universiti Malaysia Perlis, Kompleks Pusat Pengajian Jejawi 3, 02600 Arau, Perlis, Malaysia

E-mail: umifazara@unimap.edu.my

Abstract. Landfill leachate is generally known as high-strength wastewater that is difficult to handle and contains dissolved extracts and suspended matter. Microbial fuel cells (MFCs) were designed to treat landfill leachate while continuously producing power (voltage output). Three different anodes were tested in MFC reactors: carbon black, activated carbon, and zinc electrodes. Movements in the MFC reactor during treatment were also a key factor for testing. Results showed a difference in ammonia levels in the three anodes used. The study compared the efficiency of static and dynamic modes of MFC in removing ammonia. Continual leachate movement in the reactor could increase the rate of removal of the ammonia components. The setup provided a viable condition for maximum removal because the reactor movement caused the sludge to disintegrate, which allowed ammonia to separate easily from the parent leachate. Ammonia removal also resulted from the transfer of ammonium through the membrane or from ammonia loss. Constant exchange of ionic content benefited the MFC performance by increasing power production and decreasing internal electrode material resistance. This paper presents the results of the analyses of leachate treatment from the solid waste landfill located in Padang Siding Landfill, Perlis. The performance of ammonia removal was enhanced using different types of electrodes. In both modes, activated carbon performed better than black carbon and zinc. The respective percentages of ammonia removal for activated carbon of dynamic over static were 96.6%, 66.6%, and 92.8% for activated carbon, zinc, and black carbon. The results provide further information on the possibility of using MFCs in landfill leachate treatment systems.

1. Introduction

Leachate is a major challenge that threatens environmental quality in Malaysia. Leachate results from the production of large amounts of solid waste, which comprises unwanted matter produced by human activity [1, 2, 4, 5]. Population growth in urban areas in Malaysia has increased the generation of solid waste. Malaysia is one of the most economically successful countries owing to its stable
political conditions (Solid Waste Management and Public Management Cleansing Corporation, Composition Solid Waste Management and Public Management Cleansing Corporation 2011[2, 4, 5-7]. Rapid economic transition and urban population increase reflect the escalation of solid waste generation [1, 8-11]. The Solid Waste Management and Public Management Cleansing Corporation, a general policy for solid waste management, was implemented in 2011. The urban population in Malaysia has increased by over 50% in the last few decades [10-15, 17]. This population growth has caused serious concerns in solid waste production (Composition Solid Waste Management and Public Management Cleansing Corporation 2011 [2, 4, 5-7]. The number of cities in peninsular Malaysia increased by 400% from 1957 to 2000 [14, 18-22]. Urbanization and industrialization in Malaysia have changed the characteristics of solid waste [22-24]. Thus, effective solid waste management practices must be updated to suit the current waste quantity. Landfilling is the main disposal method in Malaysia. Malaysia, however, is facing serious landfill problems, such as, insufficient capacity, overflow of landfill sites, serious odour and lack of ventilation, and lack of leachate treatment facilities. Therefore, ammonia removal using the proposed method will improve the treatment of landfill waste and reduce the total cost of handling waste in Malaysia.

Several approaches of leachate treatment have been reported such as aerobic biological treatment, anaerobic biological treatment, physiochemical treatment, coagulation and advanced techniques including carbon adsorption and ion exchange. However, most of the approaches have one or more disadvantages, until recently, micro fuel cell (MFC) received significant attention as they enable the use of microorganism as promoting agent or catalyst to convert chemical energy of the electro donors into electricity. In addition to that, MFC have shown capabilities in various treatment of ammonia and ammonia related chemicals [22-26]. Therefore, the present work focused on the ammonia removal from the leachate existing in a landfill located in Padang Siding Landfill, Perlis. The performance of ammonia removal using different types of electrodes at static and dynamic modes of MFC will be analysed.

![Figure 1. Schematic diagram of The MFC](image)

2. Methods

A two-chamber microbial fuel cell (MFC) as shown in figure 1 is required to perform the bio-electrochemical reactions. Square, circular, and large-scale MFC designs are required for characterization, and a pH meter is required to determine pH levels. The sample was collected from Padang Siding Landfill by determining the physical and chemical parameters of the target ammonia. It was taken with proper precautions to prevent contamination and protected from rainfall. The leachate was collected from the landfill and sun-dried prior to laboratory testing. The temperature during sampling was 35 °C. The leachate sample was collected from the solid waste landfill pond at the back of the landfill. A total 20 L of leachate was manually collected and placed in plastic containers. After collecting the sample, it was placed in a controlled environment. The sample was immediately transported to the laboratory, characterized, and cooled at 4 °C to minimize biological and chemical reactions [22-25]
The next step is the MFC construction. The MFC is constructed from Perspex, with valves, wire, three material types and electrodes, a proton exchange membrane, and a silicon sealant that can produce electricity and harbor bacteria. The electrode and proton exchange membrane are encased in a Plexiglas compartment that is designed with valves and wires connected to the electrode and rheostat (R). A sealant is then used to ensure the compartment is airtight. A flask (1 L capacity) is used as the anode chamber, and a cylinder of Perspex (125 mL capacity) with two end pieces, one of them with a hole, is fixed to the anode chamber. Both chambers are physically separated by a proton exchange membrane with an interchange surface of 7 cm². Each chamber contains a zinc, activated carbon, or carbon black electrode. The total accessible geometrical surface of the electrodes is intended to be 6.5 cm². The electrodes are attached to the external system by a copper wire with all exposed surfaces sealed with non-conducting silicone. The MFC are operated in two different modes of electrode; static and dynamic. In the dynamic mode, the leachate is continuously stirred whilst the leachate is in quiescent condition at static mode.

3. Results and Discussion

The oxygen required for the initial starting of the device is obtained from the leachate itself through the nitrification process and the cathodic electrode oxygen reduction reaction, which relies on ion diffusion through the membrane cathode. However, the dynamic feeding medium may bring some dissolved oxygen to the system in addition to the oxygen already present in the leachate. The MFC is constructed with three configurations based on the anode electrode [5, 7]. Table 1 shows the results obtained using different anodes at a dynamic system which have almost linear removal over 4 weeks. The maximum amount of ammonia removal was by activated carbon (AC) electrode at 96.6% at week 4.

| Anode Material / Day | Week 1 % of Removal | Week 2 % of Removal | Week 3 % of Removal | Week 4 % of Removal |
|----------------------|----------------------|----------------------|----------------------|----------------------|
| Zinc                 | 20.0                 | 53.3                 | 56.3                 | 66.3                 |
| AC                   | 33.3                 | 55.5                 | 83.3                 | 96.6                 |
| CB                   | 32.3                 | 54.9                 | 80.3                 | 92.8                 |

Three MFC electrodes were evaluated in terms of their efficiency in removing ammonia. The process was dynamic for liquor sample cycling from the two tanks. The waste was recycled again by pump. All three operations were conducted under dynamic mode conditions. After the process, the MFC required approximately 30 days to achieve an optimal removal rate. This process was possible when the bacterial community found the necessary conditions to enrich and allow the MFC to operate for this long period. Figure 2 shows the ammonia removal by the zinc electrode dynamic system [2, 16, 20-24].

A linear reduction in ammonia occurred with the passage of time. The maximum amount of reduction after one week was approximately 20%. The amount of reduction varied with different types of materials. The removal rate of activated carbon was higher than those of zinc and carbon black. The dynamic process of the activated carbon sample produced a higher removal potential than any other anode material. This result may be due to the fact that activated carbon has low internal resistance resulting from the porosity of the materials that supports mass transfer. Generally, the rate of ammonia removal depends on factors such as mass transfer and internal resistance. The low internal resistance of activated carbon allows for increased mass transfer.
Figure 2. Ammonia removal by zinc electrode dynamic system.

Ammonia removal increased linearly throughout the experiment in the case of activated carbon (figure 3), which also produced a stable voltage ranging from 5 mV to 10 mV. However, the zinc electrode produced a slightly higher voltage than the two carbon materials, because electrochemically active bacteria were more dominant in zinc than in carbon materials (Table 2). Zinc produces low voltage output and low ammonia removal rate because zinc materials possess higher resistance than do carbon materials.

Figure 3. Ammonia removal by activated carbon dynamic system.

Figure 4 shows the results of carbon black anode materials. The data for this process were also monitored for 30 days before and after the MFC process. The ammonia removal percentages were 32.3\%, 55.6\%, 80.3\%, and 92.8\% for the first, second, third, and fourth weeks, respectively. Although zinc can generate high power density (voltage), its ability to remove ammonia was lower than that of carbon materials. The removal rates of both activated carbon and black carbon were high. However, activated carbon had removal rates of 33.3\%, 55.5\%, 83.3\%, and 96.3\%. The effluent ammonia concentration was 600 mg/L in all cases.
As stated earlier, the MFC can simultaneously remove ammonia and generate electricity. This ability has made MFCs a viable approach to treat water and leachate. The highest rate of voltage generation in this study was achieved when the MFC was operated with zinc anode material (0.3 V), followed by carbon materials (5–10 mV). However, figure 5 shows that activated carbon provides the highest and most consistent rates of ammonia removal. The highest efficiency of ammonia removal was achieved by activated carbon (96.3%), followed by carbon black (92.6%) and zinc (66.6%). Activated carbon is the most efficient among the three types of anode materials.

Table 2 shows the power generated by different materials. Zinc electrode achieved the highest power generation (3.60 mW), followed by activated carbon electrode (0.75 mW). The lowest power generated for activated carbon (0.4 mW) resulted from the passive nature of activated carbon in supporting electron transfer.

### Table 2. Comparison of power generation by three anode materials

| Electrode     | Voltage (mV) | Current (A) | Power (mW) |
|---------------|--------------|-------------|------------|
| Activated carbon | 5.00         | 0.15        | 0.75       |
| Zinc          | 9.00         | 0.40        | 3.60       |
As mentioned earlier, the use of MFCs can produce voltage. Figure 6 and 7 show the voltage values for the dynamic and static mode, respectively from the start until the end of treatment. The highest voltage obtained from zinc electrode was 0.761 mV, and the lowest was 0.492 mV. The highest voltage obtained from activated carbon electrode was 0.260 mV, and the lowest was 0.038 mV. The highest voltage obtained from black carbon electrode was 0.749 mV, and the lowest was 0.372 mV.

|                     | Carbon black | 4.00 | 0.10 | 0.40 |
|---------------------|--------------|------|------|------|

Figure 6. Comparison of voltage generation by three anode materials for the dynamic mode.

Figure 7. Comparison of voltage generation by three anode materials for the static mode.

4. Conclusion
This study demonstrated the process of ammonia removal by using an MFC with three different electrodes: zinc, activated carbon, and carbon black. The MFC shows the potential to use the bacterial community in mixed cultures of leachate and thus treat waste and produce electricity. The efficiency of anode materials to remove ammonia was successfully compared. Carbon materials were more efficient in removing ammonia than zinc. Over a period of 30 days, the removal rates of ammonia
were 96.3% for activated carbon and 66.6% for zinc. However, zinc is a more appropriate choice for power generation. Therefore, MFCs are a promising tool in waste treatment, wastewater treatment, and electricity generation.

Acknowledgement
This project was supported by the School of Environmental Studies, University Malaysia Perlis (UniMAP)

References

[1] Adhikari B, Dahal K R and Khanal S N 2014 A Review of Factors Affecting the Composition of Municipal Solid Waste Landfill Leachate Int. J. Eng. Sci. Innov. Technol. 3(5) 273–281
[2] Ghasimi S M D, Idris A, Ahmadun F R, Tey B T and Chuah T G 2008 Batch Anaerobic Treatment of Fresh Leachate from Transfer Station J. Eng. Sci. Technol. 3(3) 256–264
[3] Diamadopoulos E 1994 Characterization and Treatment of Recirculation Stabilized Leachate Wat. Res. 28(12) 2439-2445
[4] Evren A T, Pelin C and Baris C 2013 Bio-Electrochemical Post-Treatment of Anaerobically Treated Landfill Leachate Bioresource Technol. 128 266–272
[5] Ganesh K and Jambeck J R, 2013 Treatment of Landfill Leachate Using Microbial Fuel Cells: Alternative Anodes and Semi-Continuous Operation Bioresour. Technol. 139 383–387
[6] Karmakar S, Kundu K and Kundu S 2010 Design and Development of Microbial Fuel Cell Current Research Technology and Education Topics in Applied Microbiology and Microbial Biotechnology 1029-1034
[7] Puig S 2011 Microbial Fuel Cell Application in Landfill Leachate Treatment J. Hazard. Mater. 185(2-3) 763–767
[8] Qixing W L , Zhou and Tao H 2010 Removal of Organic Matter from Landfill Leachate by Advanced Oxidation Processes: A Review Int. J Chem.Eng. 1-10
[9] Raghab S M, Abd El Meguid A M and Hegazi H A 2013 Treatment of leachate from municipal solid waste landfill HBRC J. 9(2) 187–192
[10] Rahimnejad M, Adhami A, Darvari S, Zirepour A and Oh S E 2015 Microbial fuel cell as new technology for bioelectricity generation: A review Alexandria Eng. J. 54(3) 745–756
[11] Kettunen R H, Holiijoki T H and Rintala J A 2009 Anaerobic and sequential anaerobic-aerobic treatments of industrial and municipal landfill leachate at low temperatures Bioresour. Technol. 58 40-41
[12] Ringeisen B R, Henderson E, Wu P K, Pietron J, Ray R, Little B and Jones-Meehan J M. 2006. High power density from a miniature microbial fuel cell using Shewanella oneidensis DSP10 Environ. Sci Techno. 40(80) 2629-2634
[13] Sani A 2014 The influence of pH on the removal of ammonia from a scheduled waste landfill leachate J. Tech. (Sci. Eng.) 68(5) 25–28
[14] Seow-Wee 2012 New Perspective of Integrated Solid Waste Management in Malaysia Proceeding 3rd International Conference on Human Habitat & Environment in the Malay World 19-20 Jun 2012 UKM Bangi
[15] Solid Waste Management and Public Management Cleansing Corporation 2011 Implementation of Solid Waste and Public Cleansing Management Act (Act 672), Annual Report 2011 Chapter 4 Kuala Lumpur: Solid Waste and Public Cleansing Management Corporation
[16] Tong, Zhag B, Liu Y, S., Zheng M, Zhao Y, Tian C and Feng C 2014 Enhancement of bacterial denitrification for nitrate removal in groundwater with electrical stimulation from microbial fuel cells J. Power Sour. 268 423-429
[17] Watanabe K and Biosci J 2006 Recent developments in microbial fuel cell technologies for sustainable bioenergy J. Biosci. Bioeng. 106 528-536
[18] Woo J H, Im H J M W, Choi K B, Han C W and Kim 2001 Simultaneous organic and nitrogen removal from municipal landfill leachate using an anaerobic-aerobic system Water Res. 35 2403-2410
[19] Wu Y, Zhou S, Qin F, Ye X and Zheng K 2010 Modelling physical and oxidative removal properties of Fenton process for treatment of landfill leachate using response surface methodology (RSM) *J. Hazard. Mat.* **180**(1-3) 456–465

[20] Zhang Q Q 2013 Investigation on characteristics of leachate and concentrated leachate in the landfill leachate treatment plants *Waste Manage.* **33**(11) 2277–2286

[21] Zhou W, Qin Y S, Zheng F K and Ye X 2010 Modelling the oxidation kinetics of Fenton’s process on the degradation of humid acid *J. Hazard. Mat.* **179**(1-3), 533–539

[22] Jayesh M S, Enrico M and Prakash C G 2014 Treatment of domestic and distillery wastewater in high surface microbial fuel cells *Int. J. Hydrogen Energ.* **39** 21819-21827

[23] Guodong Z, Yan J and Duu-Jong L 2015 A lab-scale anoxic/oxic-bioelectrochemical reactor for leachate Treatments *Bioresour. Technol.* **186** 97–105

[24] Zhao G D, Zhang Q L, Jiao Y, Wang K, Lee D J and Ren N Q 2012 Biocathode microbial fuel cell for efficient electricity recovery from dairy manure *Biosens. Bioelectron.* **31** 537–543

[25] Jiao Z G D, Zhao Q L Y, Zhang J N, Jiang J Q, Ren N Q and Kim B H 2011 Improved performance of microbial fuel cell using combination biocathode of graphite fiber brush and graphite granules *J. Power Sour.* **196** 6036–6041

[26] Wang Z F W, Zhang X and Tao G, 2011 Electricity generation in a membraneless microbial fuel cell with down-flow feeding onto the cathode *Bioresour. Technol.* **102** 7324–7328