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Further Investigation of the Power Generation and Pollution Removal Capacity of Microbial Fuel Cells for the Treatment of High Nickel Content Wastewater

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Abstract: *This study investigates microbial fuel cells (MFCs) for the treatment of high nickel-contaminated wastewater and their ability to generate electricity at different nickel levels. As well as exploring trends in treatment capacity, optimal conditions for achieving functionality were identified and the effects of time and nickel dose were assessed. Nickel is commonly found in industrial wastewater, and this study measured removal efficiency by comparing influent and effluent concentrations. The results indicate that increasing nickel levels reduce the power generation capacity of the MFC, which may affect long-term efficiency. However, the ability of MFC to remove COD and nickel was unaffected by increasing nickel levels, highlighting the potential of MFC to treat industrial wastewater without increasing resources.*

Keywords: MFC, Nickel, Heavy metal, Biofilm, COD Removal

1. INTRODUCTION

There are many types of wastewaters, with industrial wastewater being one of the most difficult and harmful to treat. [1] Before it can be discharged, it needs to be cleaned as thoroughly as possible [2]. Current treatment methods for industrial wastewater are costly [3], inefficient, technologically supported, and sometimes do not provide a clear picture of the contaminants [4]. In today's world, the adoption of new technologies to eliminate water pollution are critical [5][6][7]. Microbial Fuel Cells (MFCs) represent an anaerobic technology capable of addressing many of these challenges. They are highly resource-efficient, requiring no electricity or additional resources to operate. MFCs are also effective in treating organic compounds and heavy metals and are adaptable to extremely high-temperature environments. [8]. Anaerobic environments allow microorganisms to degrade organic matter as well as produce additional resources (e.g., methane, carbon dioxide) [9]. Within the MFC chamber, the main objective is to cultivate the growth of microorganisms that play a crucial role in eliminating harmful chemicals as well as organic matter in the substrate.[10]. While a portion of the heavy metals will be adsorbed by the biofilm, adsorption plays an important role in the water treatment process [11].

The heavy metals like nickel (Ni^{2+}) are widely used in many industrial processes, such as metal polishing, electroplating, coinage, mineral processing, and the production of stainless steel and batteries [12]. However, heavy metal pollution has become a global environmental concern due to its high toxicity, difficult degradation, and biological enrichment. Therefore, treatment of wastewater contaminated with heavy metals is necessary for improving our ecosystems. Many methods have been developed to address this challenge, such as ion exchange, chemical precipitation, reverse osmosis, electrolysis and electrodialysis. However, most of these methods have high operation and maintenance costs and produce harmful sludge [13][14]. As Figure 1 demonstrates, the main mechanism of MFC is the

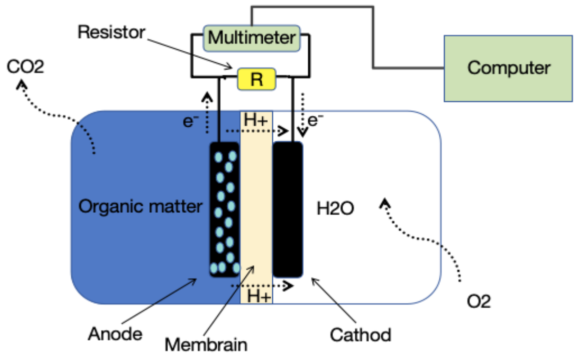

Figure 1. Mechanism of MFC

degradation of organic matter in the substrate by microorganisms at the anode, generating electrons as well as protons, which move through a cation exchange membrane (CEM) to the cathode chamber. Additionally, the electrons in the anode chamber are transferred to the cathode chamber through an external circuit. Once in the cathode chamber, protons and electrons then combine to form water molecules [15]. The electrons that pass through the external circuit generate
an electric potential, which means that an electric current is generated.

When treating industrial wastewater containing large quantities of heavy metals, the ability of the MFC system to simultaneously generate electricity and remove pollutants is critical. On the surface of the anode electrode, biofilms proliferate and utilize their ability to carry out redox reactions on contaminants. In this study, different concentrations of nickel solution will be added to the MFC anode chamber, and several parameters will be recorded like voltage, COD removal and nickel removal.

In today's industrialized world with increasing human discharge of heavy metal pollutants, it is essential to prioritize understanding the pollution removal rates and power generation efficiency of novel wastewater treatment methods, such as MFCs. This is because MFCs can use microorganisms to reduce pollutants while generate electricity. The results of this research could have far-reaching implications for building a better and more sustainable society.

2.METHODOLOGY

The study was divided into two phases, the inoculation phase and the metal phase. During the inoculation phase, cell membranes were cultured in the MFC, and in the metal phase, data was collected and analyzed for substrates with varying nickel content. Figure 2 shows the MFC used in this experiment. Temperature was checked throughout and was at room temperature, averaging about 21.5 degrees Celsius. For the statistics of the data, for COD measurements, HACH's LCI 400 COD tester was used as well as a spectrophotometer. For heavy metal measurements, a Spectroscopy Autosampler was used, and samples were analyzed for heavy metals using ICP-MS and ICP-OES. Finally for current or voltage, a Pico data logger ADC-20 & TERM | Pico Technology Voltage Data Logger, USB 1.1, USB 2.0, USB-Powered | RS (rs-online.com) was used to detect real-time voltage changes.

Figure 2. MFC used during the experiment

2.1 MFC structure

The structure of the MFC and the materials used during the experiment are shown in Figure 3. The MFC had an acrylic body, which was fixed using fixing pegs also made of acrylic material. In the anode chamber, a rubber tube was connecting the inside and outside of the MFC to facilitate substrate replacement and sampling. It is worth mentioning that solid adhesive was used to seal the rubber tube and ensure an anaerobic environment is achieved Furthermore, a carbon veil was set up in both the anode and cathode chambers as electrodes, which were connected by a Stainless-steel wire. Ion exchange membranes were used in both the anode and cathode chambers to ensure that ions travelled from the anode chamber to the cathode chamber. The cathode chamber was set up to be in contact with the outside air, and water was sprayed on a daily basis during the test period to ensure sufficient contact between the electrodes and the ion exchange membrane.

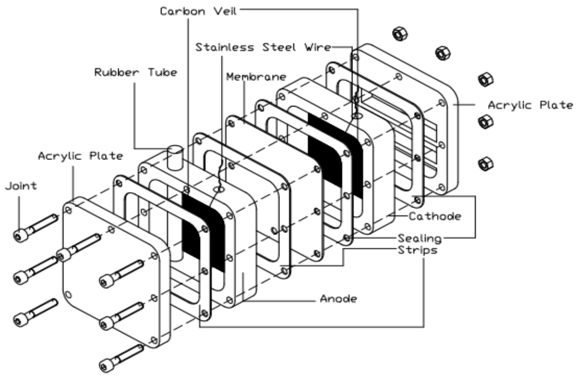

Figure 3. Structure of MFC used in the experiment.

2.2 Inoculation period

In this period, the primary objective is to culture the biofilm attached to the electrodes in the new MFC. After four rounds of incubation with a substrate containing wastewater that was obtained from Northumberland wastewater treatment plant, and 100% acetate, the biofilm in the MFC was left to stabilize. During this time the pH, conductivity, dissolved oxygen and current output were recorded to determine the stability of the process. During the experiment, the solution was flushed with nitrogen to reduce the oxygen content of the solution in order to create an anaerobic environment.

In the first round of inoculation, a substrate solution consisting of Sodium Acetate (CH_3COONa), Sodium phosphate monobasic monohydrate ($NaH_2PO_4.H_2O$) and Disodium phosphate (Na_2HPO_4) were used to prepare a 1000 mg/L COD substrate solution. The doses were 1.95g of CH_3COONa, 0.792g of $NaH_2PO_4.H_2O$ and 1.476g of Na_2HPO_4 to 1.5L of deionized water. Sodium phosphate monobasic monohydrate and disodium the substrate in the anode chamber at a steady pH of 7 [16][17]. Maintaining pH 7 mitigates the reduced power generation efficiency of the MFC and supports normal growth of microbial membranes in the anode which can be adversely affected by a low pH [18], therefore, controlling the pH inside the system is critical to its efficiency.

Afterwards, this solution was mixed with the effluent at a ratio of 7:3 and an anaerobic environment was ensured through nitrogen flushing for a period of 5 days. During this period, the MFC voltage was measured continuously, and it was found that the power generation capacity was slowly decreasing. Following this realization, the COD content was adjusted to 250mg/L to ensure the progress of the experiment.

When this experiment was repeated to confirm accuracy, the same method was used to prepare a mixed substrate of acetate and sewage water, although the COD was reduced to 250 mg/L and the ratio of acetate to sewage water was adjusted to 80:20. After three rounds of incubation, it was found that the power generation capacity was close to the same as the beginning of each round and remained stable prior to entering the metal period.

2.3 Metal period

A total of six rounds of testing were conducted during the Metal period. MFC with nickel acetate was used for the first 3 rounds at levels of 10mg/L, 20mg/L, and 30mg/L, and the last three using MFC with nickel levels of 1mg/L, 10mg/L, and 100mg/L. During this period, COD, nickel, voltage, pH, and conductivity were measurement at the influent and effluent locations of the MFC.

For solution formulation, NiCl2 was used to make nickel containing solution. 2.208g of nickel chloride (NiCl2) was added to 100ml of deionized water in a volumetric flask. The nickel acetate containing 1mg/l, 10mg/L, 20mg/l, 30mg/l, and 100mg/L was prepared by different dosages of delusion.
During this period, for the accuracy of the results, two MFCs were used for each dose, and the influent and effluent were tested twice.
In the first round of the metal period, the COD level was 250mg/L and in the second round of the final test, the COD level was 1000mg/L to see if the treatment capacity of the high COD effluent condition was affected in the long term.
Finally, the performance of the MFC in removing wastewater at different nickel and COD concentration was tested to obtain and analyze the data of COD removal, power generation capacity and nickel removal capacity in each round.

3.RESULT AND DISCUSSION

A total of six rounds of tests were conducted on the samples during the experimental period. Firstly, at nickel levels of 10 mg/L, 20 mg/L and 30 mg/L, and then the last three rounds at nickel levels of 1 mg/L, 10 mg/L and 100 mg/L. The MFCs were tested for COD, voltage and conductivity. In this experiment, COD, nickel content, voltage, pH and conductivity were measured in the influent and effluent of the MFC. COD removal capacity, metal removal capacity and power generation were considered to be the most important measures to evaluate the performance of the MFC. However, the pH and conductivity tests were designed to understand whether the MFC was operating properly and to control the variables, so these results do not provide details of the experiment.

3.1 COD removal

For COD testing, the LCI400 was used, and the results are shown below. From table.1, it can be seen that the MFC has a long-term ability to remove COD in this case. This means that the ability of MFC to remove COD will not be reduced or eliminated even at high concentrations of heavy metals. As shown in Figure 3, two groups of MFC with 10 mg/L nickel substrate had the highest COD removal capacity, and surprisingly, even its removal percentage had an increasing trend with time. On the other hand, the COD removal ratio of the 1 mg/L sample was lower than 10 mg/L, which may be due to the growth difference of microorganisms inside the MFC. The COD removal capacity of MFC tends to follow a decreasing trend when the nickel content increases over time. In this experiment, the MFC of the sample with 20 mg/L nickel content had the highest nickel removal rate. Unlike what was hypothesized before the experiment, the results do not show a common linear relationship, but rather 20mg/L has peak COD removal capacity, and lower or higher nickel content samples have lower COD removal capacity. In contrast, the COD removal capacity of the higher nickel-containing matrices were decreasing significantly as the nickel content continued to increase. This may be due to two reasons, one of which is due to the individual disparity between MFCs, which can be observed at the initial stage to present different power generation efficiencies under the same conditions.

Table.1 COD value in metal period

Concentration mg/L	COD Inf mg/L	COD Eff mg/L	COD Removal Value(mg/L)	COD Removal Rate
10mg/L	232.00	140.00	92.00	39.66
20mg/L	237.00	96.40	140.60	59.32
30mg/L	234.00	178.50	55.50	23.72
10	242.00	154.00	88.00	36.36
20	223.00	117.15	105.85	47.47
30	220.00	174.50	45.50	20.68
10	244.00	118.65	125.35	51.37
20	233.00	111.00	122.00	52.36
30	240.00	132.50	107.50	44.79
1mg/L	244.00	185.00	59.00	24.18
10mg/L	236.00	159.00	77.00	32.63
100mg/L	202.00	151.50	50.50	25.00
1	242.00	145.00	97.00	40.08
10	240.00	118.50	121.50	50.63
100	223.00	218.00	5.00	2.24
1	1131.00	542.00	589.00	52.08
10	1171.00	411.00	760.00	64.90
100	1163.00	612.00	551.00	47.38

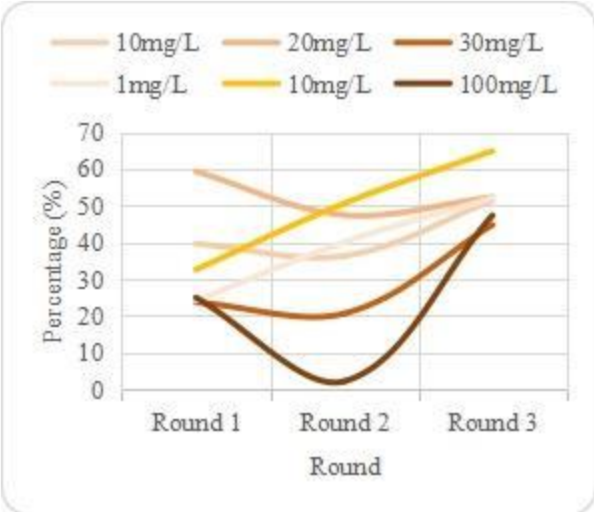

Figure 4. COD Removal Rate Trend

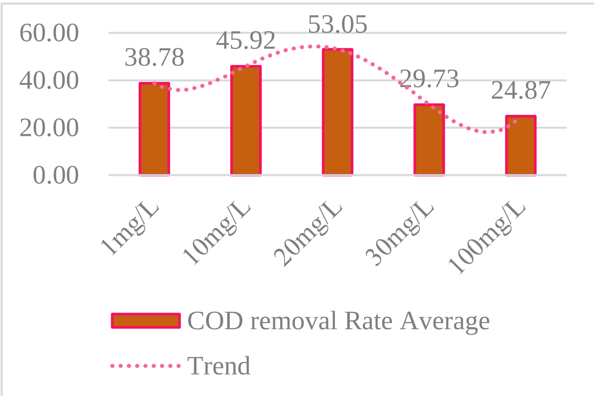

Figure 5. COD Removal Rate Average

second may be since the nickel metal adsorbed by the biofilm in the anode of the MFC was not completely cleared with the change of substrate in each round of testing. Therefore, verifying these two points in subsequent experiments may allow more reasonable linear results to be observed.

3.2 Current production

For the current measurements, the voltage of all MFCs were recorded for the full period of this experiment and the current of the MFCs was finally calculated using

Ohm's law. For voltage measurements, a Pico data logger (ADC-20 & TERM Pico Technology Voltage Data Logger, USB 2.0, USB-Powered RS) was used. If the conditions were not favorable, the MFC voltage was also manually recorded using a handheld multimeter.

Figure. 6 shows the voltage of the MFC and the temperature of the holding tank for the full period of this test in the software to detect the normal operation of the MFC as well as the normal operation of the holding tank. The software also provides an excel output of the voltages for each 10-minute period, which is used in subsequent configurations.

What can be seen in the first three rounds of MFC with 10mg/L, 20mg/L, and 30mg/L nickel content effluent matrices is that the higher the nickel content matrices the higher the voltages produced, which is different from that shown by COD removal. Mechanistically, this may be due to the higher conductivity of the higher nickel content substrates, as the higher nickel content has not yet inhibited the biofilm in the MFC due to its toxicity at this stage. The higher conductivity leads to higher performance of MFC [19], which may be the reason for these results.

The results of the last three rounds show that, MFCs with lower nickel content have higher MFC generation capacity as the nickel content increases, as can be seen in figure 8. Considering the mechanism, although higher conductivity can improve the performance of MFC, the high concentration of heavy metals will have an inhibitory effect on the biofilm in MFC and may kill the bacteria in it.[20] In the later stage, when a longer period of time has passed, the inhibitory effect outweighed the effects of the increased power production efficiency due to conductivity. Therefore, the results show that the samples with lower nickel content have a better capacity to produce electricity.

Taken together, both in the early and late stages, the MFC's generating capacity produced an overall irreversible decline. Even, later on when the MFCs in the 10 mg/L nickel content samples were replaced with 1 mg/L nickel content solution substrate, the power generation capacity of their MFCs was still lower than that of the early results, which implies that high concentration of nickel-containing matrices harms the power generation capacity of the MFCs, and the results are not long-lasting. Similarly, the rest of the MFCs tested with different dosages of substrates will have basically 0 power generation capacity at the end of the test, which again proves that high dosages of the heavy metal, nickel, in the long-term, will completely nullify the power generation capabilities of the MFCs, which especially in terms of biofilm stability as well as power generation capacity. This may be due to the toxicity of the biofilm after adsorption of heavy metals which leads to the destruction of the biofilm in order to reduce its stability as well as power generation capacity.

3.3 Nickel removal

For the removal of nickel in this experiment, it is worth mentioning that the results were consistent with the hypothesis, but what was not expected was that the removal rate of nickel was able to reach nearly 95% at the beginning, which proves that the MFC can be used for the treatment of industrial wastewater to carry out the next step of the large-scale study.

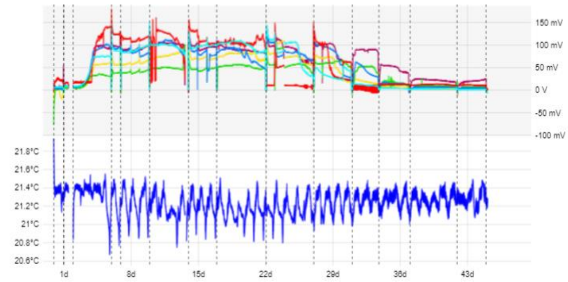

Figure 6. Voltage reading during the experiment

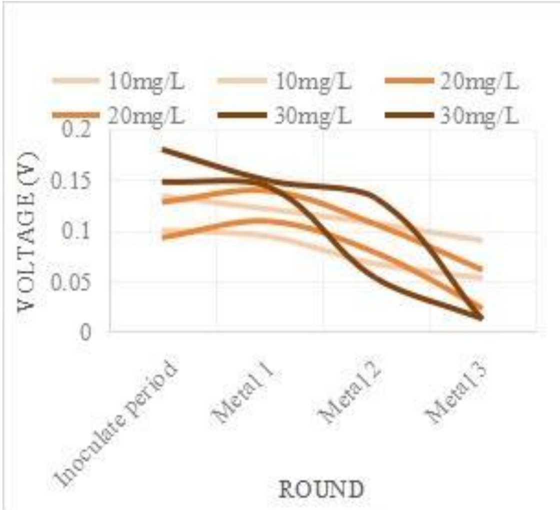

Figure 7. Voltage peak in first three rounds

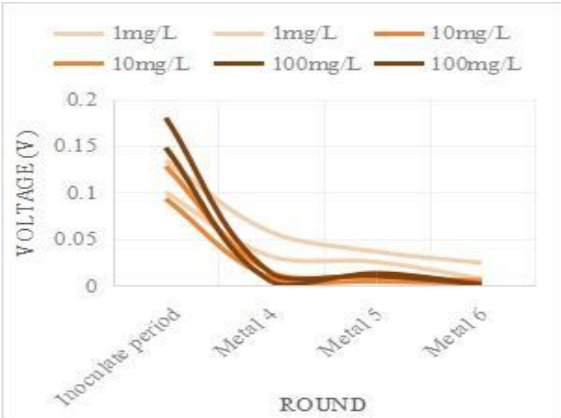

Figure 8. voltage peak in last three rounds

Table 2. Nickel Content at the inf and eff locations

Designed Concentration (mg/L)	Inf Ni Concentration (mg/L)	Eff Ni Concentration (mg/L)	Ni Removal Value (mg/L)	Ni Removal Rate (mg/L)
10mg/L	10.38	0.47	9.91	95.51
20mg/L	20.96	1.04	19.92	95.04
30mg/L	32.85	1.51	31.34	95.41
10mg/L	10.49	1.01	9.48	90.40
20mg/L	20.93	1.84	19.09	91.19
30mg/L	32.95	3.82	29.13	88.40
10mg/L	10.32	2.04	8.29	80.27
20mg/L	19.27	3.36	15.91	82.58
30mg/L	29.33	6.37	22.96	78.28
1mg/L	0.77	0.97	−0.20	−26.38
10mg/L	8.83	2.56	6.26	70.95
100mg/L	100.55	29.82	70.74	70.35
1mg/L	0.84	0.56	0.27	32.60
20mg/L	11.05	3.74	7.31	66.15
30mg/L	105.54	20.79	84.76	80.30
1mg/L	4.27	0.84	2.90	77.62
20mg/L	11.43	6.49	14.30	68.77
30mg/L	80.74	22.86	57.88	71.69

As shown in Table 2, the removal rate of nickel by MFC reached more than 95% for all three doses at the beginning, and although it decreased later, it still reached more than 80%. After the last three rounds of replacing the dosage, it can be seen that the nickel removal rate for the 1mg/L nickel content sample was negative, which is caused by the fact that even though the substrate was replaced, the nickel remaining on the inner walls of the MFC and on the electrodes were not well treated because the previous dosage was 10mg/L. Therefore, for the 1mg/L sample, the data is not precise enough nor is it incorrect. In the subsequent study, different MFCs to measure the same dose from start to finish were used, but there was no way to do this in this experiment because of the limitations of the number of MFCs.

The nickel removal efficiency of MFC with 10mg/L, 20mg/L, and 30mg/L nickel content substrate did not differ much and performs well, as can be seen in Figure 9. However, it can be seen from the details that the lower the nickel content substrate, the MFC had a higher nickel removal efficiency. And it can be seen that with time, the nickel removal efficiency of the MFC was gradually decreasing, which may be due to the fact that the removal mechanism of the MFC for heavy metals was mainly adsorption and precipitation, and as more nickel is adsorbed on the biofilm, the subsequent adsorption capacity also becomes worse.

And in the last three rounds, as shown in Figure 10, the nickel removal efficiency of the samples with 1 mg/L nickel content substrate was negative in the first round, while its removal ability recovered again with time and was higher than other samples with higher nickel content at the end. This is due to the nickel residue from the prelude experiments and the recovery of its removal capacity at a later time. From the other results, the 100 mg/L nickel samples had higher removal rates than predicted by the first three trends, and even had higher rates than the lower nickel samples for reasons that are not clear. What is certain is that MFC has a strong potential for treating industrial wastewater containing heavy metals, as mentioned earlier.

Finally, in the overall analysis, as shown in Figure 11, MFC at 20 mg/L nickel content substrate has the best removal efficiency, but it is undeniable that MFC at other dosages also has a good nickel removal effect.

From the above results and discussion, it can be determined that MFC has a high potential for nickel removal for nickel-containing wastewater, and in the long term, also has good treatment potential and recovery capability. In the case of samples with low nickel content, the nickel removal capacity is high, while in the case of samples with high nickel content, the results are acceptable and can be maintained.

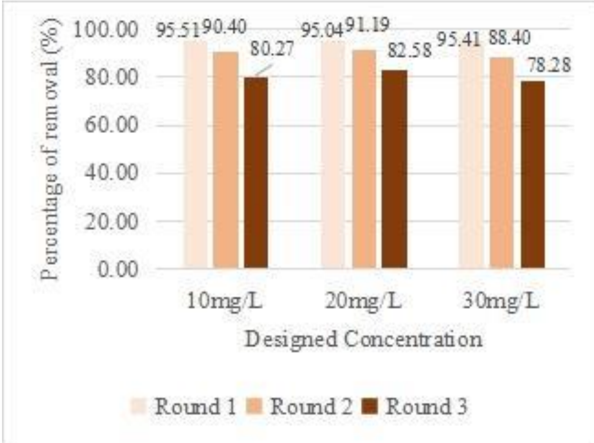

Figure 9. Nickel removal rate in the first 3 rounds

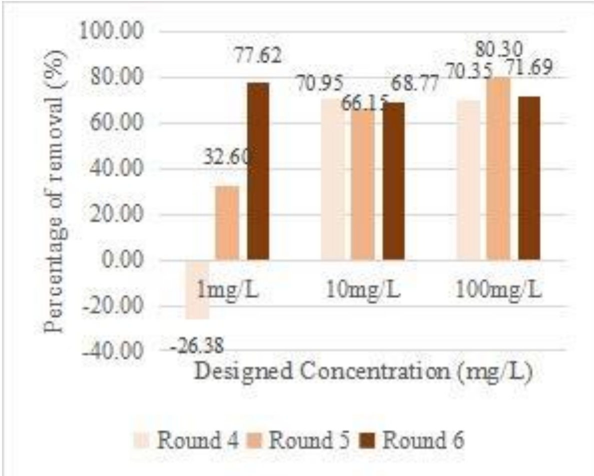

Figure 10. Nickel removal rate in the last 3 rounds

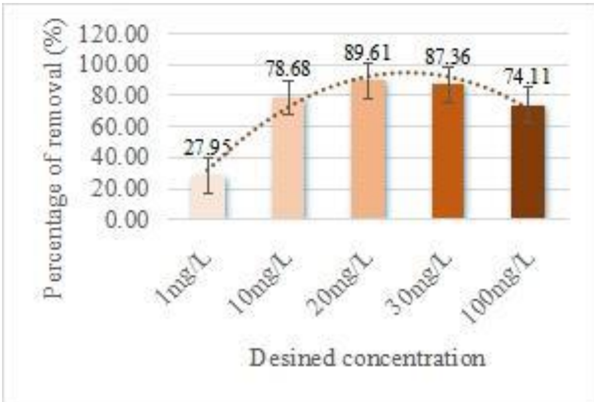

Figure 11. Average nickel removal rates

4.CONCLUSION

This study further explores the power generation and pollutant removal capabilities of MFC for nickel-containing wastewater, briefly analyzes the mechanisms involved, and acknowledges the potential of MFC for treating this type of pollutant. Firstly, for the COD removal ability of MFC in treating nickel-containing wastewater, MFC containing 20 mg/L nickel-containing substrate performed the best in this experiment. As the nickel content increases, its COD removal capacity decreases. Due to experimental limitations, the effects at lower concentrations require further investigation. In this experiment, lower nickel content, specifically at 20 mg/L, showed better performance compared to higher concentrations. Secondly, the MFC had a very good nickel removal capacity, which may be due to its adsorption effectiveness of the heavy metal, degradation and precipitation capacity. Moreover, for heavy metal removal, MFC also showed a very good long-term removal ability, which is also surprising. Due to issues with the experimental design, the sample with lower nickel content (1 mg/L) did not yield the expected results. Future research should focus more on lower nickel content to avoid this issue.

5. FUTURE & IMPROVEMENT

There are many parts of this experiment that need to be improved and a few research gaps have been created which need to be discussed.

5.1 Dosage

In this experiment only a few doses were used which includes 1mg/L, 10mg/L, 20mg/L, 30mg/L, 100mg/L were tested with high Nickel content. Although, in many cases of industrial wastewater or general sewage do not have such high nickel concentrations. From the results of this experiment, maybe the lower concentrations of heavy metal substrate will lead to higher removal efficiency of MFC, or it can also be said that the ability of MFC for treating heavy metal wastewater has more potential.

5.2 Experiment setting

As mentioned above, the 1mg/L sample did not turn out well, mainly because the effluent residue from the previous round of 10mg/L samples was still in the MFC anode chamber and had not been cleaned sufficiently, but it was not possible to clean it up by filling with water repeatedly, as this could have harmed the structure of the biofilm itself, affecting the accuracy of the experiment. Therefore, in future studies, each content should correspond to the same MFC until the end of the experiment to overcome this error.

5.3 Conductivity

Also in the present study, it was found that in the samples with lower nickel content, the conductivity had a significant effect on the power generation capacity of the MFC anode chamber. At lower nickel contents, the effect of conductivity may even be more important than the toxicity of the heavy metal to the MFC and the effect of the heavy metal. Therefore, conductivity should be set as an important research index in future studies.

5.4 Metal combination

Industrial wastewater in real-world scenarios do not consist of only one heavy metal, but rather a cocktail of contaminants including many different heavy metals as well as other organics [21]. While it is important to study the effect of a single chemical on the MFC mechanism, the biproducts formed when different chemicals are combined is also crucial. Therefore, in future experiments, it is recommended to consider the design of experiments incorporating different chemicals with greater potential for chain reactions, along with analyzing their effects on the MFC mechanism.

Overall, the current research on MFC is still far from reaching its full potential. Future studies will continue to explore the ability of MFC to treat wastewater in real-world applications.

Thirdly, for the power generation capacity of MFC on nickel-containing wastewater matrices, it shows weakness for higher levels of nickel-containing wastewater. The experimental results indicate that the power generation capacity of the MFC decreases as the nickel content in the substrate increases, demonstrating the long-term impact of this effect. Whereas, in the short term, the sample with 20mg/L nickel content substrate demonstrated a higher power generation capacity than the sample with 10mg/L nickel content substrate. This represents the importance of the conductivity associated with higher metal content in increasing the power generation capacity of MFC anodes in the short term when heavy metals do not deteriorate the biofilm due to their toxicity.

5.5 nZVI

The use of nZVI for the removal of pollutants from water is now an affordable [22], efficient and potentially promising new technology [23][24][25]. Existing studies have shown that nZVI can effectively improve the capacity of MFCs, and subsequent work can be carried out by adding nano-zero-valent iron (nZVI) to MFCs in order to improve the heavy metal removal efficiency as well as the power generation capacity of MFCs [26].

In summary, the ability of MFC for treating nickel-containing wastewater is evident, and its potential capability is worth further exploration in future studies.

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