RESEARCH ARTICLE

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Tetracycline Antibiotic Removal from Wastewater via Air-Cathode Microbial Fuel Cells

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Background & Aims of the Study: Tetracyclines are the second most used group of antibiotics in the world. This type of antibiotic has a weak attraction in the body and enters wastewater through urine and feces. This study investigated the effectiveness of tetracycline removal from wastewater by air-cathode microbial fuel cells.

Materials and methods: The current study was bench-scale experimental research as a batch mode. The anode was made of flat graphite and the air cathode was a carbon cloth with four PTFE diffusion layers with platinum cover (0.3 mg/cm^2). Two similar reactors were used. The influent wastewater (500 mg/L) was injected into two reactors (one with tetracycline and the other without tetracycline). Both reactors were used in a batch mode with 1000 Ohm external impedance in 25±2 C^o via artificial wastewater.

Results: The results of the study showed that the voltage production time in the tetracycline reactor was considerably longer than the tetracycline-free reactor. The amount of COD reduction was almost similar in both reactors. Although the effectiveness of COD reduction was similar in both reactors, because the operation time for the tetracycline reactor was longer, the rate of COD removal was considerably higher in the tetracycline-free reactor.

Conclusion: The air-cathode microbial fuel cell reactor could remove about 50% of tetracycline antibiotic from the wastewater.

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Background

Over the past few years, drug remaining has been regarded as one of the most controversial issues in environmental studies. The overuse of antibiotics around the world (100,000 to 200,000 tons per year) and their influent into the environment have raised concerns around the globe (1-2). According to the research, Iran is the leading country in the world in using antibiotics because of over prescription and overuse (3-4).

Tetracyclines are the second most used group of antibiotics in the world. The molecular structure of tetracycline is shown in Figure 1 (5-6).

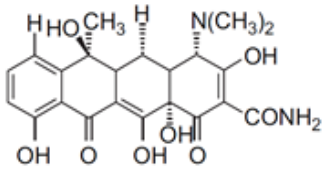

Figure 1) The chemical formula of tetracycline antibiotic

Many antibiotics have a weak attraction in the body when they are consumed by human beings or animals. After consumption, about 25% to 75% of these compounds are disposed from the body by urine or feces (7).

These compounds not only enter the environment via human wastes but also through agricultural manure, landfill site leachate, and slaughterhouses (8-10). Recent studies have shown that incomplete metabolism in the human body and inappropriate discharge of antibiotics into the wastewater treatment plants are the main sources of the release of antibiotics to the environment. Although their detectable concentration level is low in the environment, even such a low level could have poisonous effects on humans and aquatic organisms (2). In fact, their presence in the environment can bring about some problems in the future (11). Antibiotics have irreparable negative effects on human beings and the ecosystem. The influent of these compounds into aquatic environments and finally into the human body causes disorders in the function of endocrine glands and exerts teratogenic effects in pregnant women (12). The most important issue about the presence of antibiotics in the environment is the increasing resistance toward these compounds in the body that might lower their effectiveness in future (13-16). Studies conducted in Iran have observed that drug resistance in some bacteria is considerable and reaches 90% in some of these bacteria (17-19). Actually, this phenomenon can be attributed to the release of antibiotics in the aquatic environments via municipal wastewater. Whereas huge amounts of antibiotics enter the wastewater treatment plants, these treatment plants are not able to remove these compounds completely (20). Thus, antibiotics still exist in the effluents of the wastewater treatment plants Based on the results of some studies focusing on the effectiveness of drug removal in wastewater treatment plants in Europe, 80% of influent drugs entering the wastewater treatment plants are discharged into the surface waters without any removal.

Various methods have been experimented for reducing drug compounds in aquatic resources. For example, it has been shown that oxidation processes, such as ozonation and Fenton, can remove 90% of oxi-tetracycline from aquatic solution (21).

The removal of tetracycline from aquatic solutions by natural and modified zeolite has also been studied. Research findings in this regard have shown that the amount of tetracycline removal was 78.8% by natural zeolite and 77.4% by modified zeolite in artificial aquatic environments and 60% by natural zeolite and 65% by modified zeolite in natural environments.

In microbial fuel cells, exoelectrogenic bacteria transfer the electrochemical energy of organic materials as electrons (22-23). Electrons move from anode to cathode and produce electricity. One of the main mechanisms for transferring electrons by bacteria is Pyocianin that is made by the bacteria themselves and has antibiotic characteristics (24). Most of the processes employed to remove tetracycline include physical and chemical processes. Biological processes, however, have been used limitedly. Therefore, the current study used microbial fuel cells for this process.

Aims of the study:

The aim of this study was to evaluate the efficiency of microbial fuel cell for tetracycline removal and its voltage production from wastewater.

Materials & Methods

This study has a descriptive-analytic design focusing on the changes made by microbial fuel cells in the concentration level of tetracycline antibiotic.

In the current study, two reactors were simultaneously used so that the results could be compared: one of these reactors used synthetic wastewater without tetracycline and the other used with tetracycline .

First, the reactors required for the experiment were built and relevant equipment such as power supply, electrodes, and membranes was attached to them. To build the body of the

pilots, 0.5 and 2 cm Plexiglass sheets were used. The anode electrode used in the experiment was flat graphite carbon heated to 450 ºC temperature and the cathode electrode was carbon cloth. One side of cathode that was exposed to the air was covered by 4 layers of Polytetrafluoroethylene (PTFE) (25). By so doing, there was no need to aeration in the catholyte. Given the fact that recent studies have shown that using platinum as a catalyst increases the effectiveness of fuel cells, to increase the effectiveness of the system carbon-platinum cover were used in the air-exposed side of the cathode in this study (0.3 mg/cm^2 of cathode area) (26-27). The cation exchange membrane used in ACMFC was a CMI7000 model from Membrane International INC. Given the importance of preparing membranes before using, they were put in NaCl 1M solution for 72 hours. Meanwhile, the solution containing the membranes was changed several times (28). The wastewater chamber volume, after the equipment and relevant tools were added, was 80 ml and the catholyte chamber volume was 20 ml .

Moreover, 2 g/L of sodium acetate trihydrate was used to produce synthetic wastewater. In order to avoid rapid decrease of pH level in the anolyte chamber, 25 mmol (pH 7) of phosphate buffer solution (PBS) was used. After the phosphate buffer was prepared, 2 g of sodium acetate, 1.5 g of NH_4Cl, 0.1 g of $MgCl_2.4H_2O$, 0.1 g of $CaCl_2.2H_2O$, and 0.1 g of yeast extract were added. Afterwards, 10 ml of trace mineral was also added (29). In addition, a few multivitamins were finally added to the solution to have the needed vitamins in the solution. In this study, 500 µg/L concentration of tetracycline antibiotic was prepared in the synthetic wastewater. Finally, 25 mM PBS was also used as catholyte .

At the startup phase, the prepared synthetic wastewater (80% of the volume) with the influent wastewater into the aeration pond of the municipal wastewater treatment plant (20% of the volume) was injected into the wastewater chamber. In this stage, all the existing solutions including wastewater and catholyte were replaced every 5 days. After several runs of startup phase, the produced voltage reached the maximum degree and stayed constant for a while. The final stage of the startup was the time when in two successive periods, the maximum voltage was reached. Before attaching the reactors to the external power supply, both reactors were able to reach the maximum OCV by connecting electrodes to a multimeter via a wire. During 24 hours, the maximum OCV produced by both reactors was equal to 810 mV. In the end, by connecting the wires to a resistance box, recording the produced voltages in different situations, and drawing the polarization curve, the optimal external resistance was finally determined as 1000 Ω .

The operation of the reactors was conducted based on the buffer capacity of 25 mM PBS in a batch mode in 25±2 ºC ambient temperature. When the output voltage reached less than 80 mV, the wastewater and catholyte were removed from the chamber and new solutions were replaced. Different parameters such as pH, EC, COD, and tetracycline were tested before the wastewater entered the anolyte chamber. After the wastewater exited the chamber, wastewater experiments were also conducted as standard methods.

Before conducting the COD experiment, wastewater samples were filtered by the paper filter. They were later filtered by 0.45-micron membrane filter for removing suspended particles. Next, the COD of influent and effluent samples were measured by spectrophotometry method (DR4000 device).

In order to measure the amount of tetracycline in the effluent wastewater, the samples were filtered through the paper filter and 0.45-micron membrane filter. The tetracycline was then extracted by Sep-Pak cartridges from Waters

Company. Before the extraction, the cartridge was prepared by 5 ml of methanol and 5 ml of water and its pH level was adjusted to pH 4 with hydrochloric acid. Later on, 10 ml sample was injected into the cartridge and the solid phase extraction (SPE) was done by 4 ml per second. After condensation, the extracted tetracycline from the cartridge was released by 1 ml of methanol and injected into the HPLC device .

The employed HPLC in this study was an HPLC-DAD model equipped with diode array detector, had a reverse-phase column (C18 4-5 micron mm*125mm particles), and had a mobile phase of oxalic acid and acetonitrile.

By using $P = \frac{V^2}{RA}$ and $I = \frac{V}{RA}$ formulas, the voltages produced by the reactor were turned into mW/m^2 electric power density and electric power density based on mA/m^2 of the cathode area. Likewise, the amount of produced power by the reactor was reported based on m3 of wastewater volume. The rate of tetracycline reduction and COD in the wastewater were calculated by this formula:

$$Q_c = \frac{C_i - C_t}{t}$$

C_i and C_t: the influent and effluent COD or tetracycline in the reactor, respectively .

t: the hydraulic retention time of wastewater

After using the reactor, producing the electric current, and doing the COD experiment on the influent and effluent of wastewater of the reactor, the Coulombic effectiveness was measured by the following formula:

$$c_E = \frac{8\int_0^{tb} Idt}{FV_{An}\Delta COD}$$

In this formula, *I* is the current density, t is the time, F is Faraday constant, and V_{An} is the volume of wastewater chamber.

Results

During 510 hours of operation of the tetracycline-free reactor, 3 operation runs were done separately with the average time of the operation equaling to 165.8±8.2 hours per cycle. On the contrary, during 450 hours of operation of the tetracycline reactor, 6 operation runs were performed with the average time of 72.2±5.1 hours per cycle.

Figure 2 shows the average of the maximum produced voltage by the two reactors. Accordingly, in 25 mMPBS, the average voltage in the tetracycline-free reactor in 3 consecutive phases was 420±15 mV and the average voltage in the tetracycline reactor in 6 consecutive phases was 530±25 mV.

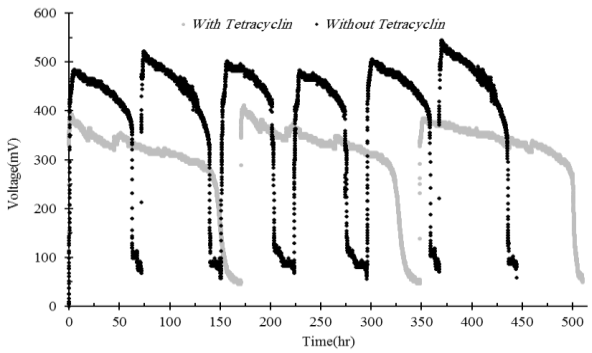

Figure 2) The diagram of changes in the produced voltage (mV)

Therefore, the average of the maximum power density, current density and volumetric power was 249.5±0.3 and 397.3±0.9 mW/m^2 , 294.21±1.2 and 749.5±35.8 mA/m^2 , 2.2±0.01 and 3.5±0.01 w/m^3 in the tetracycline reactor and in the tetracycline-free reactor respectively.

Analyzing the pH changes in wastewater and catholyte

The changes in the pH level of wastewater and catholyte in both reactors are shown in Figure 3.

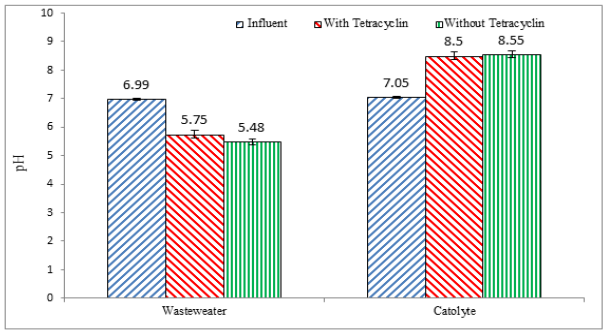

Figure 3) The comparison of pH changes in wastewater and catholyte

As shown in Figure 3, the amount of pH reduction in wastewater in both reactors is roughly the same. In the tetracycline reactor, the pH level of the wastewater decreased from 6.99±0.01 to 5.75±0.1. The pH level also decreased in the tetracycline-free reactor from 6.99±0.01 to 5.48±0.1. In the catholyte, however, the pH increased from 7.05±0.01 to 8.5±0.1 in the tetracycline reactor and to 8.0±55.1 in the tetracycline-free reactor.

Analyzing EC changes in wastewater and catholyte

Figure 4 shows the EC changes in wastewater and catholyte in the two reactors.

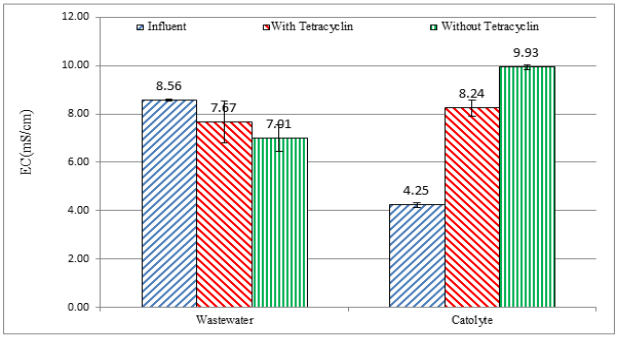

Figure 4) The comparison of EC changes in the two reactors

As shown in Figure 4, the EC of artificial wastewater was about 8.56±0.04 mS/cm due to the addition of bacteria's needed compounds. This amount, however, decreased after the operation runs to 7.0±67.9 in the tetracycline reactor and to 7.01±0.6 in the tetracycline-free reactor. The EC of the catholyte also lowered from 4.25±0.1 mS/cm down to 7.67±0.9 mS/cm in the tetracycline reactor and to 9.93±0.1 mS/cm in the tetracycline-free reactor.

- Analyzing COD changes in wastewater

Because 2 g/L of sodium acetate was used in this study and other bacteria's needed compounds were added, the influent COD of the reactors was 930±4 mg/L. After the operation run was over, the COD decreased to 168±9 mg/L in the tetracycline reactor and to 112±16 mg/L in the tetracycline-free reactor. Therefore, the removal percentage of the COD was 81.9% in the tetracycline reactor and 88% in the tetracycline-free reactor. Meanwhile, the average amount of COD removal in the 25mMPBS was 4.0±50.0.5 mg/L.hr in the tetracycline reactor and 11.0±33.22 mg/L.hr in the tetracycline-free reactor. Since the COD removal effectiveness was roughly similar in both reactors while the voltage production time was longer in the tetracycline-free reactor, it could be concluded that the COD removal rate was lower in the tetracycline-free reactor. Thus, the Coulombic effectiveness of the tetracycline reactor was 0.34 whereas it was 0.17 for the tetracycline-free reactor.

Analyzing tetracycline concentration changes in wastewater

The amount of tetracycline removal by the reactor in different operation phases was 54±18%. The concentration level decreased from 500±26 mg/L to 229±12 mg/L. Accordingly, the rate of tetracycline removal by the air-cathode microbial fuel cell was 1.63 mg/L in 165.8 hours.

Discussion

Given the similarity in the effectiveness level of COD removal in both reactors, the operation time for the tetracycline reactor was considerably longer than the time for the tetracycline-free reactor. This finding reveals that this amount of tetracycline concentration can have significant effects on exoelectrogenic bacteria in a way that it can increase the

necessary COD removal retention time by 200%. Therefore, considering large-scale industrial systems for COD removal in wastewaters including drug compounds, such as tetracycline, this result shows that longer retention times need larger chamber volumes and hence drastically increase the costs of designing wastewater treatment processes.

The ACMFC reactor could decrease the influent tetracycline about 54% in the specified operation time. Given the complexity of tetracycline chemical formula, long time is needed for bacteria to be able to break down and degrade this compound. Besides, since exoelectrogenic bacteria are anaerobic and these processes can accept high CODs with complex compounds, it could be concluded that if supplementary research is conducted, it is possible to use ACMFC to reduce drug compounds.

Producing lower voltage, current, and electrical power in the tetracycline reactor, compared to the tetracycline-free reactor, can be a function of the interference of tetracycline drug compound with the duplication of exoelectrogenic bacteria. In the later operation runs, the voltage rate slightly increased, showing that the exoelectrogenic bacteria might have been adapted to the tetracycline-filled wastewater environment.

Because 2 g/L of sodium acetate trihydrate was used as a carbon resource in the wastewater, considering the following formula, the produced H^+ ions reduced the pH level.

$$CH_3COO^- + 4H_2O \rightarrow 2HCO_3^- + 9H^+ + 8e^-$$

The amount of pH reduction in both reactors was roughly similar (about pH 5). The important issue here is that the produced voltage by the reactors swiftly decreased when the pH level reached 5. Both reactors included a carbon resource (sodium acetate). However, the reduction of pH level in wastewater decreased the activity of the bacteria and the voltage together. Research has observed that the activity of exoelectrogenic bacteria decreases considerably in pH 5 compared to pH 7 (24). Thus, one of the main reasons for the reduction of pH level in the wastewater of microbial fuel cell reactors could be attributed to inequality of the capacity of cation exchange membrane in transferring produced H^+ ions, compared to the ions produced by degradation of the carbon resource.

Based on these findings, the sensitivity of the bacteria to low pH levels is one of the most important limitations in operation of microbial fuel cells. Putting electrodes in chambers that are separated from each other by ion-exchange membranes causes an imbalance in the pH level. The process of oxidation in anodes produces organic proton materials. In cathodes, however, the oxidation in neutral pH produces water.

If no buffer is used in the catholyte, pH level increases and reaches about 12 (30). The pH effect on the maximum possible potential of electrode reactions is measured by the following formula:

$$E = E^0 + \frac{RT}{ZF} ln \frac{\{ox\}}{\{Red\}}$$

This reaction for hydrogen is as follows:

$$2H^+ + 2e^- \rightarrow H_2$$

$E^0(H_2)=0$

$$E^{0/} = E^0 - \frac{RT}{ZF} ln \frac{[H_2]}{[H^+]^2}$$

$$E^{0/} = 0 - \frac{\left(\frac{8.31J}{mol.K}\right).(298.15K)}{2\left(\frac{96500C}{mol}\right)} ln \frac{1}{[10^{-7}M]^2} = -0.414V$$

In previous sections, E^0 (O_2) was measured equal to 0.805 V. Therefore, because $E^0/(O_2) > E^0/(H_2)$, so oxygen is reduced by hydrogen.

The influent of sodium ions, which is produced by sodium acetate degradation in cathode

chamber by cation exchange membrane, into wastewater chamber considerably, increased the catholyte EC in both reactors.

$$CH_3COO^- Na^+ + 4H_2O \rightarrow 2HCO_3{}^- + 9H^+ + 8e^- + Na^+$$

The produced electrodes entered the catholyte chamber via wire and the produced H^+ and Na^+ ions entered the catholyte chamber via cation membrane. The amount of EC increased twice as much after each operation run in both reactors. The amount of EC increase in the tetracycline-free reactor was more than the amount in the tetracycline reactor. This shows that sodium acetate is degraded more in the wastewater chamber. The faster reduction of COD in the tetracycline-free reactor in a shorter time also corroborates this issue.

The influent of sodium ions from the wastewater chamber into the cathode chamber and the consumption of needed compounds by the bacteria caused a drastic reduction in the EC of the influent wastewater of the reactors. In line with other findings, the amount of EC reduction in the tetracycline-free reactor was more than the amount in the tetracycline reactor, showing that more carbon resource (sodium acetate) was degraded in the tetracycline-free reactor.

The rate of COD removal in the tetracycline-free reactor was remarkably higher than the rate in the tetracycline reactor. This phenomenon is due to the existence of tetracycline antibiotic in the reactor that increased the retention time in the reactor. Consequently, the rate of COD removal considerably decreased.

Finally, results showed that the Coulombic effectiveness of the tetracycline reactor was two times more than the effectiveness of the tetracycline-free reactor. One of the reasons for such a higher Coulombic effectiveness in the tetracycline reactor is the longer voltage production time in similar situations.

Conclusion

It seems that based on the 50% rate of reduction in tetracycline antibiotic by microbial fuel cells and the production of electricity as well as treating wastewater, it is possible to reach three goals (wastewater treatment, electricity production, and tetracycline removal) by improving wastewater treatment systems.

Footnotes

Conflict of Interest:

The authors declared no conflict of interest.

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