Removal of Antibiotics from Synthetic Wastewater by Advanced Oxidation Processes (AOPs)

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Abstract—In this study, Fenton’s Reagents process was used for the treatment of water containing Amoxicillin antibiotics of 100 mg/L concentration. The molar concentration (0.1M, 0.2M, 0.3M, 0.35M, and 0.4M) of Fenton’s reagents was varied with different ratio of ferrous sulphate to hydrogen peroxide. COD and BOD reduction after treatment were calculated and optimum ratio was determined. Total Suspended Solid (TSS) formed after treatment was also measured.

Keywords— Amoxicillin antibiotics, Fenton’s reagent, Advanced Oxidation Processes (AOPs)

1. Introduction

One of pharmaceutical compound namely antibiotics have been detected in worldwide in environmental matrices (e.g. water and wastewater) and their presence on ecosystem has been known for 30 years[1]. The presence of the antibiotics in environment is at low level, but with continuous input of this compound may lead potential risk to the aquatic and terrestrial organism[2]. Hence, antibiotics have emerged as one of pollutant to the environment. Common antibiotics (with their concentration) found in wastewater are Sulphonamides: sulfamethoxazole (0.02–0.58 (µg/L), fluoroquinolones: ofloxacin (6–52 ng/L), ciprofloxacin (6–60 ng/L) bacteriostatic: trimethoprim (0.11–0.37 µg/L) Penicillin group; penicillin G (0.025 µg/L)[2].

The using of antibiotics is to sustain health condition of living organism such as human, animal and plant. The consumed antibiotics by organisms are excreted through urine or feces and sent to the sewage or manure[3]. This is one of route how antibiotics can emerge on the environment. Besides that, antibiotics are also can be found in different environmental compartment such as hospital effluent resulted from heavy used from human and veterinary medicine[4]. The emerging antibiotics in environment cause the formation of resistant microorganisms that causing problem to public health and also result to imbalance microbial ecosystem. Antibiotics could produce toxic effect that affects microbial community and other living organism in environment.

The objective of this study is to investigate removal of the antibiotics from synthetic wastewater by degrade it using Fenton’s reagent process. The degradation is measured in term of COD and BOD reduction after the treatment at different molar concentration of Fenton’s reagent.

2. Methodology

2.1 Preparation of Synthetic Wastewater

This synthetic wastewater contains amoxicillin antibiotics at 100 mg/L concentration. Amoxicillin antibiotics was obtained from Poliklinik Cahaya, Selangor is dissolved in distilled water and stored at 4°C.

2.2 Fenton’s Reagent Experiment

500 mL of synthetic wastewater was poured into 4 500mL beaker. The pH of this synthetic wastewater was adjusted to pH between 2 – 3 by adding several drops of 30% of sulfuric acid for optimum Fenton’s reaction[5]. Fenton’s reagent molar concentration of 0.1M with different ratio (1:2, 1:4, 1:8, and 1:10) of ferrous sulphate to hydrogen peroxide was added for each beaker and stir with stirrer for 30 minutes. After 30 minutes sample was taken for COD, BOD, and TSS test. Similar procedure was repeated for 0.2M, 0.3M, 0.35M, and 0.4M of molar concentration of Fenton’s reagent.

2.3 Chemical Oxygen Demand (COD) Test

COD reagent such as sulfuric acid reagent, standard ferrous ammonium sulfate titrant (FAS) (0.10M), potassium dichromate digestion solution (0.01667M), and ferroin indicator solution was prepared. The 1.5ml sample mixed with 3.5ml sulfuric acid reagent and 1.5ml potassium dichromate was heated for 2 hours at 150°C. The heated mixture then was cooled at room temperature and mixed with 2 drops ferroin indicator and titrate with 0.10M until color change from blue-green to reddish brown. Blank sample also was prepared in same manner as sample. Below is the equation to calculate the COD value:

\[
\text{FAS Molarity (M)} = \frac{\text{volume of 0.01676 K2Cr2O7 titrated (ml)}}{\text{Volume FAS used in titration (ml)}} \times 0.10
\]

Where: A = FAS for blank
B = FAS for sample
M = FAS molarity
8000 = miliequivalent weight of oxygen x 1000 mL/L

2.4 Biochemical Oxygen Demand (BOD)

BOD reagent (iodine azide solution, starch indicator, standard sodium thiosulfate titrant, manganous sulfate solution and dilution water) was prepared. 2 BOD 300 ml bottle was prepared. 1.5ml sample is diluted with dilution water inside each BOD bottles. One bottle is labelled with day-0 and another bottle with day-5. The day-0 bottle was mixed with 1ml of manganous sulfate, 1ml iodine azide and 1ml concentrated sulfuric acid. 200 mL was taken from day-0 bottle for titration with sodium thiosulfates solution till colourless. The day-5 bottle was wrapped with aluminum foil and incubates at 20°C for 5 days. After 5 days titration was done as day-0 bottle. Below is the equation to calculate BOD value:

\[
\text{BOD (mg/L)} = \frac{(A-B) \times M \times 8000}{\text{ml sample}}
\]

Where: \( A = \text{DO diluted sample for day-0} \)
\( B = \text{DO diluted sample for day-5} \)
M = FAS molarity
8000 = miliequivalent weight of oxygen x 1000 mL/L

2.5 Total Suspended Solid (TSS)

Weight of a filter paper was measured. By using the filter paper, sample was filtered to obtain the suspended solid. The filtrate was sent to the sewage or manure. Below is the equation to measure the TSS value:

\[
\text{TSS (mg/L)} = \frac{(A-B) \times 1000}{V}
\]
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Where

\[ A = \text{weight of filter + dried residue, mg} \]
\[ B = \text{weight of filter, mg} \]
\[ V = \text{volume of sample, mL} \]

3. Result and Discussion

3.1 COD Reduction at Different Ratio of \( \text{Fe}^{2+}/\text{H}_2\text{O}_2 \).

Value of COD of the synthetic wastewater before the treatment was 125.33 mg/L. After treatment with 0.1M and 0.2M of Fenton’s reagent COD value of the synthetic wastewater was reduced to certain value. Treatment was done for 30 minutes and foam was produced during the reaction and the color of water also was changed to reddish color. Figure 1 below shows reduction of COD after treated with 0.1M and 0.2M of Fenton’s reagent.

![Figure 1 COD Reduction at Different Ratio of Ferrous Sulphate to Hydrogen Peroxide of Different Concentration of Fenton’s Reagent](image)

For 0.1M of Fenton’s reagent, the COD reduction is increased as the ratio of \( \text{Fe}^{2+}/\text{H}_2\text{O}_2 \) decreases. However, further decreasing of \( \text{Fe}^{2+}/\text{H}_2\text{O}_2 \) will not increase the reduction of COD due to higher concentration of \( \text{Fe}^{2+} \) that will directly react with hydroxyl radical[6]. The maximum COD reduction at 0.1M is occurred at 1/10 ratio of \( \text{Fe}^{2+}/\text{H}_2\text{O}_2 \). The value of COD at this ratio is 27.00 mg/L and the reduction was 78%.

At 0.2M of Fenton’s reagent, the COD reduction is showed similar behavior as at 0.1M where, the COD reduction is increased as the ratio of \( \text{Fe}^{2+}/\text{H}_2\text{O}_2 \) decreases. However, the COD reduction at 0.2M is higher than COD reduction at 0.1M. Other study also shows that, the COD reduction is increased as the concentration of Fenton’s reagent increase[7]. The maximum COD reduction was occurred at 1/10 where the COD value is 26.50 mg/L and percentage of reduction is 79%. From literature review the COD reduction will be increased as the Fenton’s reagent concentration increased[7].

4. Conclusion

From this study, the COD reduction is increase as the ratio of \( \text{Fe}^{2+}/\text{H}_2\text{O}_2 \) decreases. However as reported in other study, further decreasing of \( \text{Fe}^{2+}/\text{H}_2\text{O}_2 \) will not increase the COD reduction due to higher concentration of \( \text{Fe}^{2+} \).

This study also shows that, as the concentration of Fenton’s reagent increase, the COD reduction also increases. This behavior also was reported in other study. Maximum of COD reduction for 0.1M occurred at 1/10 at the percentage of 78%. At 0.2M the maximum COD reduction also occurred at 1/10 at the percentage of 79%. This study result only up to 0.2M. However, result for 0.3M, 0.35M, 0.4M will be done.

References


