Comparison of Azithromycin COD removal from wastewater by Fenton, Fenton like and Electro-Fenton processes

Ahmad Reza Yazdanbakhsh, Mahdieh Sardar, Sima Eliasi and Amir Sheikh Mohammadi*

Department of Environmental Health Engineering
Shahid Beheshti University of Medical Sciences
Teran City 11369, Iran

Key Words: Azithromycin, Electro-Fenton, Fenton, Fenton like, wastewater

ABSTRACT

This work makes a comparison between Fenton (Fe^{2+}/H_2O_2), Fenton like (Fe^0/H_2O_2) and Electro-Fenton processes to investigate the removal of Chemical Oxygen Demand (COD) from synthetic wastewater. The effects of operational parameters such as initial pH, current density, amount of hydrogen peroxide, amounts of Fe^0 and Fe^{2+}, applied voltage and electrolysis time on COD removal efficiency were investigated. The optimum values were determined for the Electro-Fenton process: current density = 20 mA cm^{-2}, hydrogen peroxide concentration = 2 mM, electrolysis time = 60 min, and pH 3.0. Desired pH in both Fenton and Fenton like processes was 7.0 and hydrogen peroxide concentrations for the Fenton and Fenton like processes were found to be 0.2 and 0.4 mM, respectively. In the optimum operating range for each of these operating variables, the Azithromycin COD removal efficiency was in order of Electro-Fenton > Fenton like > Fenton.

INTRODUCTION

In recent years pharmaceutical drugs have emerged as a novel class of water contaminants for which public and scientific concern is increasing steadily because of their persistence in the environment, impacts of pharmaceuticals for human and veterinary use in the environment [1]. Huge amounts of these chemicals in terms of thousands of tons are annually used to prevent or treat microbial infections in each European country, and may be excreted both unmetabolized and as active metabolites [2-5]. Their presence in the effluent of sewage treatment plants (STPs) indicates their poor biodegradability in municipal sewage and STPs that can be emitted into the receiving water systems [6,7]. Microbial resistance is one of the greatest concerns related to use and antibiotics disposal in environment. The Azithromycin (Formula: $C_{38}H_{69}NO_{13}$, Brand names: Zithromax, Sumamed, Zitrocin), is a member of the macrolide antibiotics that is more effective in destroying Gram-negative bacteria, especially *Haemophilus* influenza [8]. Hence, biological treatment processes have a low effect on the degradation of the medicine (because of the toxicity of this medicine for microbes). It is a cause for concern, because of theirs large-scale production and their common use in the treatment of bacterial infections. Therefore a pretreatment process is often required prior to discharge into the biological treatment processes. Advanced oxidation processes (AOPs) are one of the new technologies used for various applications in wastewater treatment, water reclamation, indirect potable water reuse, drinking water production, and recently in micro-pollutant control of sewage treatment effluents. Compared to other technologies (e.g., membrane filtration, adsorption, ion exchange, evaporation, and stripping), the organic compounds in water are degraded rather than concentrated or transferred into different phases. AOPs have the ability to generate elevated concentrations of hydroxyl radical ($^{\bullet}OH$), a strong oxidant capable of complete oxidation of most organic compounds into carbon dioxide, water, and mineral acids or salts [9-12]. The free radical chemistry makes AOPs interesting to the destruction of recalcitrant, anthropogenic and toxic organic pollutants, bacteria, viruses, and last but not least, the emerging

*Corresponding author
Email: amir.sheikh123@yahoo.com

micropollutants also called as trace pollutants/organics. The advantage of AOPs is the relative high reaction power of hydroxyl radical. In literature, this reaction power is often expressed in terms of electrode potential versus hydrogen electrode of redox reaction, but this is chemically not correct since the standard redox potential is only related to electron transfer, whereas the $^{\bullet}OH$ radical reacts by three different pathways and mostly on hydrogen abstraction pathway in the field of water treatment. This study attempted to examine the possibility of COD removal from pharmaceutical wastewater by Fenton, Fenton like and Electro-Fenton.

The most important advanced oxidation treatment based on the use of H_2O_2 is Fenton [13-16]. Fenton and Fenton-like processes use Fe^0 and Fe^{2+} in reaction with H_2O_2 respectively, to produce $^{\bullet}OH$ in which important reactions are as follows:

$$Fe^0 \rightarrow Fe^{2+} + 2e^- \quad (1)$$

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + {}^{\bullet}OH$$

$$Fe^{3+} + H_2O_2 \leftrightarrow Fe\text{-}OOH^{2+} + H^+ \leftrightarrow \quad (2)$$

$$Fe^{2+} + HO_2^{\bullet} + H^+ \quad (3)$$

In Electro-Fenton process H_2O_2 is externally applied into the electrocoagulation system while a sacrificial Fe anode is used as Fe^{2+} source and Fenton reactive was formed in the process [17-19]. During electrocoagulation process, Fe^{2+} ions are the common ions generated via the dissolution of iron that are produced at the anode and in contrast, OH^- ions are produced at the cathode. By mixing the solution, metal hydroxide species are produced which are beneficial for adsorption and coprecipitation of soluble organic compounds and trapping of colloidal particles, and these flocs are removed easily from aqueous medium by sedimentation or flotation [20,22]. Arslan-Alaton and Gurses utilized Fenton-like process for the COD removal of penicillin from the wastewater. At optimum reaction conditions 44% COD removal achieved by Fenton-like process [17]. Fan et al. used a Fenton process in the removal of sulfasalazine in which the removal of COD reached 84% in 60 min [19]. Babuponnusami and Muthukumar used electro Fenton process for advanced oxidation of phenol. The results showed 62.5% phenol degradation for Electro-Fenton process at 16 mA cm^{-2} in 60 min [23].

MATERIALS AND METHODS

1. Materials

Antibiotic was purchased from the Farabi Pharmaceutical Company. The chemical and physical properties of antibiotic are summarized in Table 1. All of the materials and reagents used in this work were of analytical grade and were purchased from Merck. The simulated antibiotic wastewater was prepared by dissolving 0.1 g Azithromycin in 1000 mL distilled water. Based on our analysis 1 mg L^{-1} of Azithromycin is equivalent to 1.95 mg L^{-1} COD.

2. Experimental Set-up

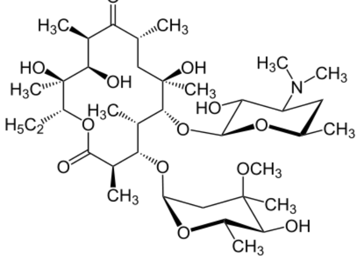
2.1. Fenton and Fenton like processes

Fenton and Fenton like processes were performed in a cylindrical glass reactor with a magnetic stirrer using a constant speed of 200 rpm. To determine the effects of pH, iron (Fe^{2+} or Fe^0) and H_2O_2 concentrations on the removal efficiency of COD, experiments were designed in three stages. In the first stage, Fe^{2+} and H_2O_2 concentrations were kept constant (from run to run) to determine the optimum pH for maximum COD removal. In the second stage by considering the optimum pH of first stage and the Fe^{2+} or Fe^0 concentration at a fixed level, the optimum level for H_2O_2 was determined. Finally by considering the optimum amounts of pH and H_2O_2 of the first and second stages, the optimum concentration for the Fe^{2+} or Fe^0 was measured and determined in the third stage.

2.2. Electro-Fenton process

The electrochemical oxidation experimental setup consisted of a cylindrical glass cell, which was 24 cm in height and 8 cm in internal diameter, 4 metal plates (monolar electrodes) from iron, a direct current power

Table 1. Physical and chemical properties of Azithromycin [21]

Properties	Azithromycin
Molecular Weight (g mol^{-1})	749
Classification	Antibacterial agent, antibiotic
Purity	96%
Solubility in Water	Insoluble
Physical State	White crystalline powder
Vapor Pressure	20-30 (25 °C)
Melting Point	113 °C
Molecular Structure	
Stability	Stable at normal temperatures and pressures
Toxicity	Rat LD_{50} (Oral): 1050 mg kg^{-1}

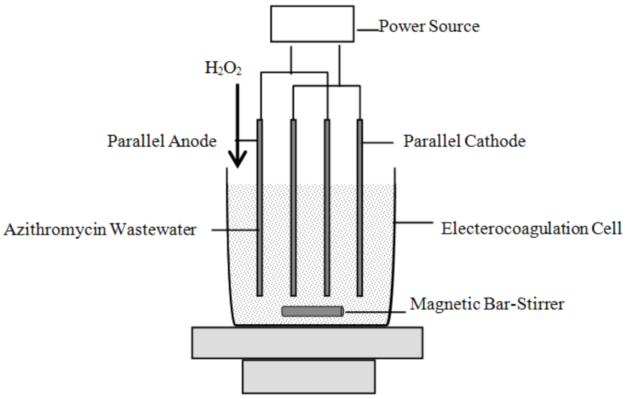


Fig. 1. A schematic diagram of the electrochemical reactor.

supply with current wires, a magnetic stirrer using a constant speed of 400 rpm. The plate dimensions were 150 × 160 × 10 mm and the electrodes were installed parallel to each other in the cell at a fixed distance of 3 cm apart from each other. The batch experimental setup is schematically shown in Fig. 1. The effects of the main operational variables (included the water pH, current density (0-20 mA cm^{-2}), and reaction time (10-90 min)) on the electrochemical process in treating COD, were investigated in various runs. At the beginning of a run, the antibiotic solution was fed into the reactor and the pH and conductivity were adjusted to a desirable value. Then the electrodes were connected to the DC power supply with the current density of 0-20 mA cm^{-2}, and the reaction was started. The H_2O_2 was added at preset dosage.

3. Chemical Analysis

The COD samples were measured by a Hach spectrophotometer (DR 5000) after filtration through a Millipore membrane filter with a pore size of 0.45 µm. Initial and residual H_2O_2 amounts were determined by the spectrophotometry method (presence of H_2O_2 leads to overestimating COD). The samples withdrawn were immediately added into an Erlenmeyer flask containing sodium hydroxide solution to quench the reaction. This was carried out to correct the quantitative effect of the concentration of hydrogen peroxide. Also iron amount in the solution for Fenton and Fenton like was determined by the phenanthroline method. The electrical conductivity was determined using Hach instrument, and temperature and solution pH monitored using the ultrameter II from Myron L Company in the electrolysis. The tests were carried out according to Standard Methods for the Examination of Water and Wastewater [24]. Each experiment was conducted three times.

RESULTS AND DISSCUSION

1. Factors Affecting the Performance of Fenton, Fenton like and Electro-Fenton Processes

1.1. The effect of solution pH on the performance of Fenton, Fenton like and Electro-Fenton processes

The pH value influences in the generation of $^{\bullet}OH$ and the oxidation of organic substances. Figure 2 shows the influence of initial pH on different processes in the decrease of final COD. To elucidate the role of pH (in the range of 3 to 11) was examined as one of the main variables affecting in removal of COD from wastewater by AOPs. The minimum final COD for Fenton and Fenton like processes, at pH 7 and for Electro-Fenton process at pH 3 was determined. The increase in the final COD in alkaline pH (for the all processes), could be due to precipitation of ferric ions as ferric hydroxides and decomposition of H_2O_2 to O_2 and H_2O. Consequently the degradation of organic chemicals in solution is decreased. The increase in the final COD below optimum pH is due to the high level of H^+ ions which inhibit the generation of hydroxyl and

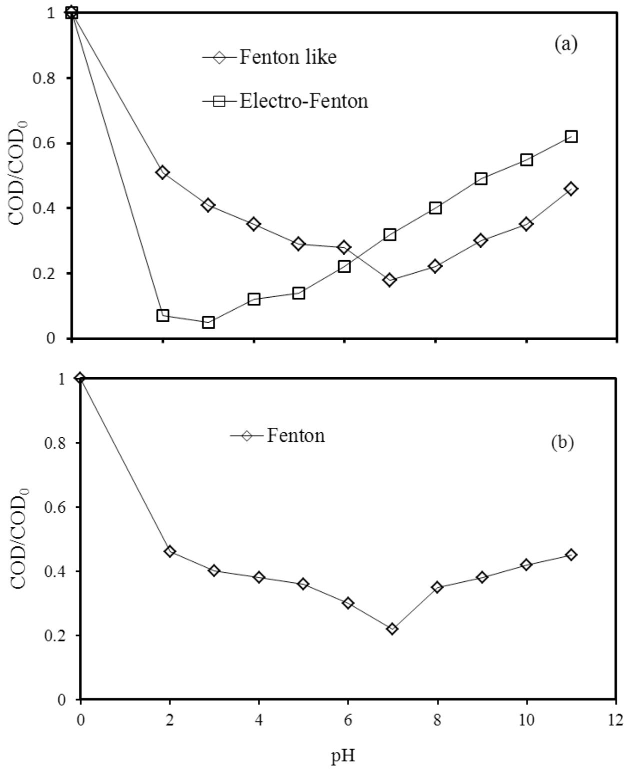


Fig. 2. Effect of pH on COD removal efficiencies obtained for Azithromycin antibiotic by Fenton like and Electro-Fenton (a), Fenton (b) processes (conditions: initial antibiotic concentration, 100 mg L^{-1}; H_2O_2 concentration, 0.1 mM; Fe^0 or Fe^{2+} concentration, 0.1 mM; current density, 20 mA cm^{-2}; reaction time, 60 min).

perhydroxyl radicals. It also decreases degradation of H_2O_2 because of the presence of $Fe(OH)^{2+}$, $Fe(OH)_2^{+}$ and $Fe_2(OH)_2^{4+}$ which react more slowly with H_2O_2 and are disadvantageous for antibiotic precipitation [25-27]. Xing et al. utilized a combination of coagulation and AOP to remove the COD related to antibiotic fermentation wastewater that the optimum pH of the Fenton process was determined to be 4.0 [10]. Elmolla and Chaudhuri used Fenton oxidation for treatment of antibiotic from wastewater with the best operating conditions for treatment at pH 3.0 [27]. Su et al. used Fenton and Electro-Fenton processes for degradation of Acetaminophen (ACTP). The ACTP degradation efficiency increased to 72% at pH 2 for the Fenton process and 74% at pH 4 for the Electro-Fenton process [7].

1.2. Effect of the solution H_2O_2 on the performance of Fenton, Fenton like and Electro-Fenton processes

The determination of optimum H_2O_2 amount is quite important in the Fenton based process, because the main cost of the method is the cost of H_2O_2 and excessive dose of H_2O_2 triggers adverse effects. To determine optimum H_2O_2 concentration, experiments were conducted by varying the amount of H_2O_2 from 0 to 3.5 mM. Figure 3 shows the amount of H_2O_2 required for the decreasing final COD using Fenton, Fenton like and Electro-Fenton processes. The results indicated that the minimum final COD of Azithromycin was 0.2, 0.4 and 2 mM for Fenton, Fenton like and Electro-Fenton respectively. For the Electro-Fenton process, the low COD removal can be attributed to the fact that the system suffered from both reactions (electrocoagulation and Fenton) simultaneously. The increase of final COD with decrease of H_2O_2 concentration could be due to partial oxidation of Fe^{2+} under less than optimum values of H_2O_2 concentration conditions. The increase in the concentration of H_2O_2 up to optimum did not significantly affect on the removal of COD due to, the combination of $^{\bullet}OH$ radicals with H_2O_2 [7,17]. Klavarioti et al. used the Fenton process for removal of residual pharmaceuticals COD from aqueous systems. The removal efficiency of COD was 56% at 3 M H_2O_2 [28].

1.3. Effect of Fe^0 and Fe^{2+} on the performance of Fenton, Fenton like process

To obtain the optimum concentration of Fe^0 and Fe^{2+}, some experiments were carried out changing this parameter between 0.05 and 0.8 mM. Figure 4 presents the COD removal of Azithromycin under different concentrations of Fe^0 and Fe^{2+}. Within the range studied, it was found that the addition of Fe^{2+} or Fe^0 concentrations enhanced the efficiency of

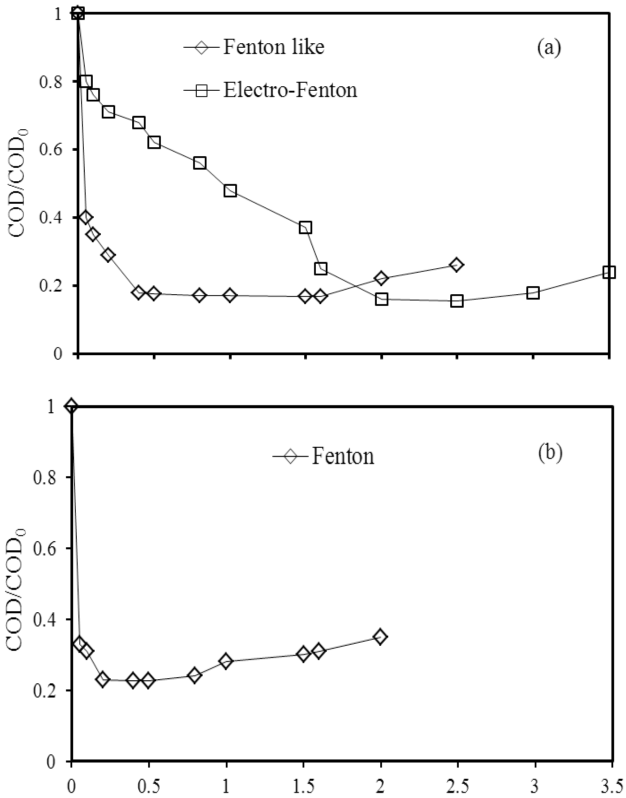


Fig. 3. Effect of H_2O_2 on COD removal efficiencies obtained for Azithromycin antibiotic by Fenton-like and Electro-Fenton (a), Fenton (b) processes (conditions: initial antibiotic concentration, 100 mg L^{-1}; Fe^0 or Fe^{2+} concentration, 0.1 mM; current density, 20 mA cm^{-2} ; reaction time, 60 min).

Azithromycin COD removal. The minimum final COD were observed at the Fe^0 and Fe^{2+} dosages of 0.36 and 0.42 mM for Fenton like and Fenton respectively. The addition of the iron above these amounts did not affect the final COD. An enormous increase in the ferrous ions will contribute to an increase in the total dissolved solids content of the effluent stream which is undesirable. Also higher dosages of Fe^{2+}or Fe^0 caused the recombination of OH radicals, as Fe^{2+} reacted with $^{\bullet}OH$ radicals as a scavenger [29]. Also it shows that in the absence of Fe^0 and Fe^{2+}, hydrogen peroxide seems to inhibit the Azithromycin degradation. Priambodo et al. used semi batch (Photo)-Electro-Fenton method for treatment of real wastewater. The removal efficiency of total organic carbon was 99% at the optimum Fe^{2+} dosage of 2000 mg L^{-1} [30].

1.4. Effect of current density on Electro-Fenton process

One of the most important parameters that can affect the removal efficiency in the electrochemical process is current density, which determines the coagulant dosage. To examine the influence of the

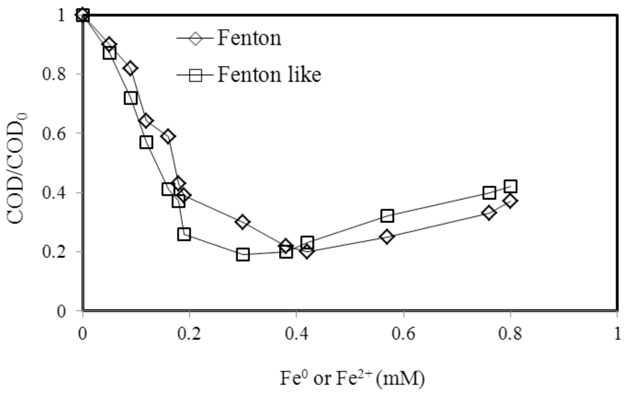


Fig. 4. Effect of Fe^0 or Fe^{2+} on COD removal efficiencies obtained for Azithromycin antibiotic by Fenton and Fenton-like processes (conditions: initial antibiotic concentration, 100 mg L^{-1}; H_2O_2 concentration, optimum dosage; reaction time, 60 min).

current density on the COD removal during the electrolysis with the iron electrodes, experiments were conducted by varying the current density from 2.5 to 20 mA cm^{-2}. The results are presented in Fig. 5. From the results, it is found that the COD removal efficiency was increased to 60% at 20 mA cm^{-2} from 20% at 0.5 mA cm^{-2} after 30 min and the removal efficiency of COD at higher current densities than 20 mA cm^{-2} stayed at the constant value**.** The amount of COD removal depends upon the quantity of ferric hydroxide produced, which is related to the time and current density [31-33]. The minimum energy consumption was 1 kWh g^{-1} COD at 20 mA cm^{-2} current density for 30 min that was calculated using the Eq. 4:

$$E = U\ I\ t_{EC} \quad (4)$$

where E is the electrical energy in Wh, U the cell voltage in volt (V), I the current in ampere (A) and t_{EC} is the time of electrocoagulation process (h).

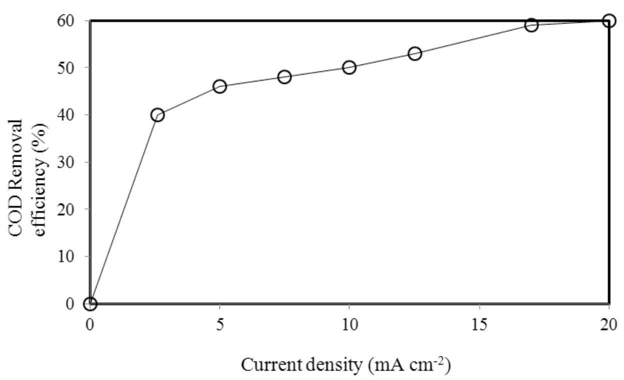


Fig. 5. Effect of current density on COD removal (initial antibiotic concentration, 100 mg L^{-1}; reaction time, 30 min; pH, 3.0; H_2O_2, 2 mM; initial conductivity, 390 µS cm^{-1}; distance between plates, 20 mm).

Wang et al. studied COD removal from real dyeing wastewater by Electro-Fenton technology using an activated carbon fiber cathode. In their studies the COD removal efficiency was 75% under optimum conditions of current density, 32 mA cm^{-2} [34].

2. Change in the Characteristics of the Solution for Electro-Fenton

Figure 6 shows the change in the electrical conductivity of the solution, total dissolved solids (TDS) and pH with the electrolysis time. It was observed that solution conductivity and the TDS in EF

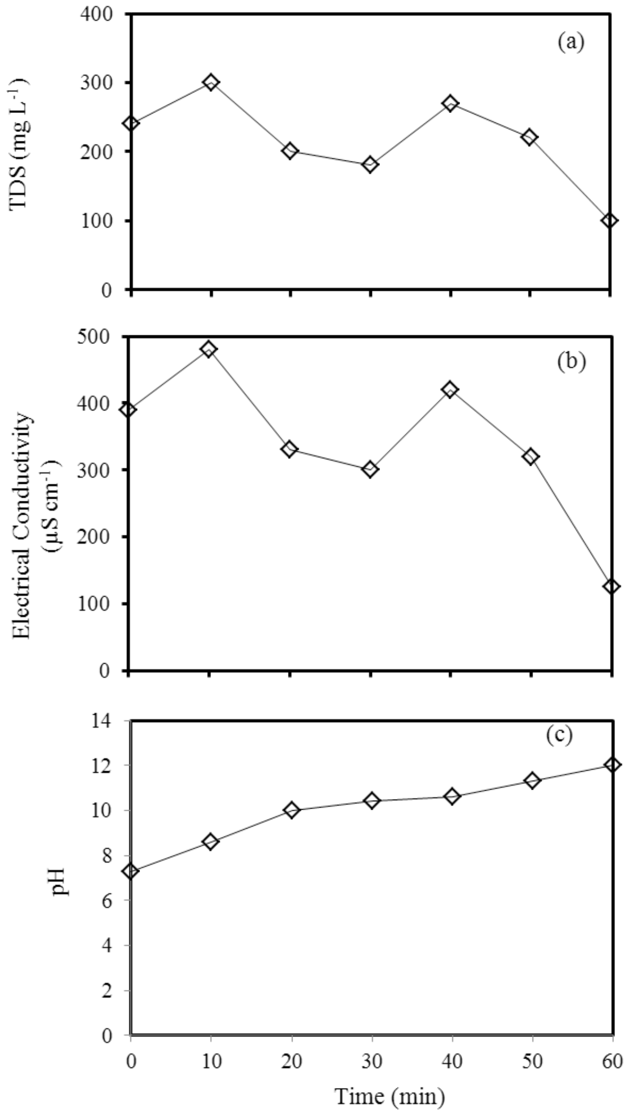


Fig. 6. Change in the characteristics of the solution (a) total dissolved solids, (b) electrical conductivity (c) pH (current density, 20 mA cm^{-2}; initial conductivity, 390 µS cm^{-1}; distance between plates, 20 mm).

process decreased from 390 to 110 μS cm^{-1} and 250 to 100 mg L^{-1} respectively after 60 min of reaction. This decrease in solution conductivity and the TDS with the electrolysis time may be due to removal of some ion species present in the wastewater. Also pHs are altered with the electrolysis time, and increased from 7 to 12 at the end of 60 min. It is due to the production of OH^- in solution [35].

3. Comparison among Different Processes Studied

The comparison of various processes was studied for the removal of Azithromycin COD from synthetic wastewater. Under optimum reaction conditions, 76, and 82 and 96% COD removals were achieved by Fenton, Fenton like and Electro-Fenton processes, respectively. In the optimum operating range for each of these operating variables, the Azithromycin COD removal efficiency was in order of Electro-Fenton > Fenton like > Fenton.

CONCLUSIONS

The Fenton, Fenton like and Electro-Fenton processes were tested on the synthetic wastewater and the effects of operational parameters such as pH, amounts of hydrogen peroxide, Fe^0 and Fe^{2+}, current density and electrolysis time on COD removal efficiency were investigated. Based on the results obtained, Electro-Fenton process was selected as the best process for the COD removal and the maximum COD removal efficiency was 96% for the initial COD concentration 195 mg L^{-1} at 60 min. Therefore, the pretreatment of the wastewater containing the antibiotic by these processes leads to a significant decrease in organic load for subsequent biological process. The optimal conditions for the removal of Azithromycin COD by Electro-Fenton process was determined to be current density = 20 mA cm^{-2}, hydrogen peroxide concentration = 2 mM, electrolysis time = 60 min, and pH 3. Finally, given the results obtained from this research, it is clear that, these processes are technologically feasible.

REFERENCES

1. Elmolla, E.S. and M. Chaudhuri, The feasibility of using combined Fenton-SBR for antibiotic wastewater treatment. Desalination, 285, 14-21 (2012).
2. Yazdanbakhsh, A.R., M.R. Massoudinegad, S. Eliasi and A.S. Mohammadi, The influence of operational parameters on reduce of azithromyin COD from wastewater using the peroxi-electrocoagulation process. J. Wat. Process. Engin. 6, 51-57 (2015).
3. Elmolla, E.S. and M. Chaudhuri, Combined photo-Fenton-SBR process for antibiotic wastewater treatment. J. Hazard. Mater., 192(3), 1418-1426 (2011).
4. Elmolla, E.S. and M. Chaudhuri, Photo-Fenton treatment of antibiotic wastewater. Nat. Environ. Pollut. Technol., 9(2), 365-370 (2010).
5. Ternes, T.A., J. Stüber, N. Herrmann, D. McDowell, A. Ried, M. Kampmann and B. Teiser, Ozonation: A tool for removal of pharmaceuticals, contrast media and musk fragrances from wastewater? Water Res., 37(8), 1976-1982 (2003).
6. Westerhoff, P., Y. Yoon, S. Snyder and E. Wert, Fate of endocrine-disruptor, pharmaceutical, and personal care product chemicals during simulated drinking water treatment processes. Environ. Sci. Technol., 39(17), 6649-6663 (2005).
7. Su, C.C., A.T. Chang, L.M. Bellotindos and M.C. Lu, Degradation of acetaminophen by Fenton and electro-Fenton processes in aerator reactor. Sep. Purif. Technol., 99, 8-13 (2012).
8. Chisholm, S.A., T.J. Neal, A.B. Alawattegama, H.D.L. Birley, R.A. Howe and C.A. Ison, Emergence of high-level azithromycin resistance in *Neisseria gonorrhoeae* in England and Wales. J. Antimicrob. Chemoth., 64(2), 353-358 (2009).
9. Zaharia, C., D. Suteu, A. Muresan, R. Muresan and A. Popescu, Textile wastewater treatment by homogeneous oxidation with hydrogen peroxide. Environ. Eng. Manag. J., 8(6), 1359-1369 (2009).
10. Xing, Z.P., D.Z. Sun and X.J. Yu, Treatment of antibiotic fermentation wastewater using the combined polyferric sulfate coagulation with Fenton-like oxidation. Environ. Prog. Sustain., 29(1), 42-51 (2010).
11. Babuponnusami, A. and K. Muthukumar, A review on Fenton and improvements to the Fenton process for wastewater treatment. J. Environ. Chem. Eng., 2(1), 557-572 (2014).
12. Barbusinski, K., The modified Fenton process for decolorization of dye wastewater. Pol. J. Environ. Stud., 14(3), 281-285 (2005).
13. Arsene, D., C.P. Musteret, C. Catrinescu, P. Apopei, G. Barjoveanu and C. Teodosiu, Combined oxidation and ultrafiltration processes for the removal of priority organic pollutants from wastewaters. Environ. Eng. Manag. J., 10(12), 1967-1976 (2011).
14. Lee, E., H. Lee, Y.K. Kim, K. Sohn and K. Lee, Hydrogen peroxide interference in chemical oxygen demand during ozone based advanced oxidation of anaerobically digested livestock

wastewater. Int. J. Environ. Sci. Te., 8(2), 381-388 (2011).
15. Hong, S.H., B.H. Kwon, J.K. Lee and I.K. Kim, Degradation of 2-chlorophenol by Fenton and photo-Fenton processes. Korean J. Chem. Eng., 25(1), 46-52 (2008).
16. Wang, S., A comparative study of Fenton and Fenton-like reaction kinetics in decolourisation of wastewater. Dyes Pigments, 76(3), 714-720 (2008).
17. Arslan-Alaton, I. and F. Gurses, Photo-Fenton-like and photo-fenton-like oxidation of Procaine Penicillin G formulation effluent. J. Photoch. Photobio. A, 165(1-3), 165-175 (2004).
18. Homem, V., A. Alves and L. Santos, Microwave-assisted Fenton's oxidation of amoxicillin. Chem. Eng. J., 220, 35-44 (2013).
19. Fan, X.Q., H.Y. Hao, X.X. Shen, F. Chen and J.L. Zhang, Removal and degradation pathway study of sulfasalazine with Fenton-like reaction. J. Hazard. Mater., 190(1-3), 493-500 (2011).
20. Cortez, S., P. Teixeira, R. Oliveira and M. Mota, Evaluation of Fenton and ozone-based advanced oxidation processes as mature landfill leachate pre-treatments. J. Environ. Manage., 92(3), 749-755 (2011).
21. Yazdanbakhsh, A.R., A.S. Mohammadi, M. Sardar, H. Godini and M. Almasian, COD removal from synthetic wastewater containing azithromycin using combined coagulation and a Fenton-like process. Environ. Eng. Manag. J., 13(12), 2929-2936 (2014).
22. Yazdanbakhsh, A.R., M. Kashefiasl, H. Zareh, E. Agaiani, M. Sardar and A. Sheikhmohammadi, Thickening of biological sludge by Electro-Coagulation-Flotation process. Int. J. Electrochem. Sci., 10, 3746-3756 (2015).
23. Babuponnusami, A. and K. Muthukumar, Advanced oxidation of phenol: A comparison between Fenton, electro-Fenton, sono-electro-Fenton and photo-electro-Fenton processes. Chem. Eng. J., 183, 1-9 (2012).
24. APHA, Standard Methods for the Examination of Water and Wastewater. 21st Ed., American Public Health Association, Washington, DC (2005).
25. Chou, W.L., C.T. Wang and K.Y. Huang, Investigation of process parameters for the removal of polyvinyl alcohol from aqueous solution by iron electrocoagulation. Desalination, 251(1-3), 12-19 (2010).
26. Cho, J.H., J.E. Lee and C.S. Ra, Effects of electric voltage and sodium chloride level on electrolysis of swine wastewater. J. Hazard. Mater., 180(1-3), 535-541 (2010).
27. Elmolla, E.S. and M. Chaudhuri, Comparison of different advanced oxidation processes for treatment of antibiotic aqueous solution. Desalination, 256(1-3), 43-47 (2010).
28. Klavarioti, M., D. Mantzavinos and D. Kassinos, Removal of residual pharmaceuticals from aqueous systems by advanced oxidation processes. Environ. Int., 35(2), 402-417 (2009).
29. Kabdasli, I., B. Vardar, I. Arslan-Alaton and O. Tunay, Effect of dye auxiliaries on color and COD removal from simulated reactive dyebath effluent by electrocoagulation. Chem. Eng. J., 148(1), 89-96 (2009).
30. Priambodo, R., Y.J. Shih, Y.J. Huang and Y.H. Huang, Treatment of real wastewater using semi batch (Photo)-Electro-Fenton method. Sustain. Environ. Res., 21(6), 389-393 (2011).
31. Vasudevan, S., J. Lakshmi, J. Jayaraj and G. Sozhan, Remediation of phosphate-contaminated water by electrocoagulation with aluminum, aluminum alloy and mild steel anodes. J. Hazard. Mater., 164(2-3), 1480-1486 (2009).
32. Apaydin, O., U. Kurt and M.T. Gonullu, An investigation on the treatment of tannery wastewater by electrocoagulation. Global Nest J., 11(4), 546-555 (2009).
33. Cheng, W., M. Yang, Y.Y. Xie, B. Liang, Z.Q. Fang and E.P. Tsang, Enhancement of mineralization of metronidazole by the electro-Fenton process with a Ce/SnO_2-Sb coated titanium anode. Chem. Eng. J., 220, 214-220 (2013).
34. Wang, C.T., W.L. Chou, M.H. Chung and Y.M. Kuo, COD removal from real dyeing wastewater by electro-Fenton technology using an activated carbon fiber cathode. Desalination, 253(1-3), 129-134 (2010).
35. Farhadi, S., B. Aminzadeh, A. Torabian, V. Khatibikamal and M.A. Fard, Comparison of COD removal from pharmaceutical wastewater by electrocoagulation, photoelectrocoagulation, peroxi-electrocoagulation and peroxi-photoelectrocoagulation processes. J. Hazard. Mater., 219, 35-42 (2012).

Discussions of this paper may appear in the discussion section of a future issue. All discussions should be submitted to the Editor-in-Chief within six months of publication.

Manuscript Received: December 10, 2014
Revision Received: April 22, 2015
and Accepted: May 18, 2015